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Source apportionment and health risk assessment of heavy metals in soil for a township in Jiangsu Province, China

Yanxue Jiang [a, b], Sihong Chao [a, b], Jianwei Liu [a, b], Yue Yang [a, b], Yanjiao Chen [a, b], Aichen Zhang [a, b], Hongbin Cao [a, b, *]

[a] Beijing Area Major Laboratory of Protection and Utilization of Traditional Chinese Medicine, Beijing Normal University, Beijing, China
[b] College of Resource Science & Technology, Beijing Normal University, Beijing, China

HIGHLIGHTS

- Soil heavy metals caused unacceptable health risks, mainly through homegrown food.
- Arsenic and chromium were the predominant hazardous elements.
- Waste incineration, textile/dyeing industries were the main anthropogenic inputs.
- Electroplating and livestock/poultry industries produced the highest health risks.

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ABSTRACT

Human activities contribute greatly to heavy metal pollution in soils. Concentrations of 15 metal elements were detected in 105 soil samples collected from a typical rural-industrial town in southern Jiangsu, China. Among them, 7 heavy metals—lead, copper, zinc, arsenic, chromium, cadmium, and nickel—were considered in the health risk assessment for residents via soil inhalation, dermal contact, and/or direct/indirect ingestion. Their potential sources were quantitatively apportioned by positive matrix factorization using the data set of all metal elements, in combination with geostatistical analysis, land use investigation, and industrial composition analysis. Furthermore, the health risks imposed by sources of heavy metal in soil were estimated for the first time. The results indicated that Cr, Cu, Cd, Pb, Ni, and Co accumulated in the soil, attaining a mild pollution level. The total hazard index values were 3.62 and 6.11, and the total cancer risks were 9.78×10^{-4} and 4.03×10^{-4} for adults and children, respectively. That is, both non-carcinogenic and carcinogenic risks posed by soil metals were above acceptable levels. Cr and As require special attention because the health risks of Cr and As individually exceeded the acceptable levels. The ingestion of homegrown produce was predominantly responsible for the high risks. The potential sources were apportioned as: a) waste incineration and textile/dyeing industries (28.3%), b) natural sources (45.4%), c) traffic emissions (5.3%), and d) electroplating industries and livestock/poultry breeding (21.0%). Health risks of four sources accounted for 23.5%, 32.7%, 7.4%, and 36.4% of the total risk, respectively.

1. Introduction

Along with the remarkable achievement of rapid economic development, environmental costs are also significantly increasing. Soil contamination by heavy metals (HM) has been increasing worldwide (Facchinelli et al., 2001; Frangi and Richard, 1997; Giller and McGrath, 1988; Huang et al., 2007; Li et al., 2014; Solgi et al., 2012) and has become the focus of attention in recent years (Chen et al., 2016; Wang et al., 2016; Zou et al., 2015). Heavy metals have generally high toxicity with low concentration thresholds, long residence times (often exceeding decades), and persistent bioavailability (Alloway, 2013). They could be hazardous to human health and ecosystems at a trace level due to their ubiquity, toxicity, and persistence (Burges et al., 2015; Guney et al., 2010). The United

* Corresponding author. College of Resource Science & Technology, Beijing Normal University, No.19 Xinjiekouwai Street, HaiDian District, Beijing 100875, China.
E-mail address: caohongbin@bnu.edu.cn (H. Cao).

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States Environmental Protection Agency (USEPA) considers heavy metals such as Cd, Cr, As, Hg, Pb, Cu, Zn, and Ni as priority control pollutants (USEPA, 2014). Many studies have focused on pollution levels and risk assessments of HM in soil environments (Chabukdhara and Nema, 2013; Li et al., 2014; Zhao et al., 2012; Zheng et al., 2010). According to the first National Survey of Soil Contamination conducted by the Ministry of Environmental Protection and Ministry of Land and Resources of China, concentrations of heavy metals in 82.8% of the soil samples exceeded the standard limit (MEPPRC and MLRPRC, 2014). It is necessary to evaluate human health risks from exposure to soil heavy metals, and identify the contamination sources to improve the soil environment and protect human health.

Compared with research involving pollution investigation, risk assessment, or remediation of soil heavy metals, few have been conducted for qualitative source identification, and even fewer for quantitative source apportionment. Qualitative methods used in previous source studies have included geostatistical models based on geographic information systems (Davis et al., 2009; Facchinelli et al., 2001; Nanos and Rodríguez Martín, 2012; Sun et al., 2013; Zhang, 2006), multivariate statistics analyses (principal component analysis, PCA; cluster analysis, CA) (Qu et al., 2013), and isotopic signatures (Cheng and Hu, 2010; Luo et al., 2011). Quantitative methods have included mostly receptor models, such as a Chemical Mass Balance (CMB) model, PCA related methods (Absolute Principal Component Scores, APCS; Multiple Linear Regression, PCA-MLR; (A)PCS-MLR; UNMIX model), and Positive Matrix Factorization (PMF) (Luo et al., 2014; Mijić et al., 2010; Wang et al., 2016). PMF has been widely applied for source apportionment of pollutants in the atmosphere (Amil et al., 2016; Hsu et al., 2016; Kim et al., 2007; Lee et al., 1999), water (Li et al., 2015; Li and Zhang, 2011; Rodenburg et al., 2011; Soonthornnonda and Christensen, 2008), and sediment (Bzdusek et al., 2006; Chen et al., 2013; Comero et al., 2014; Sundqvist et al., 2010). There are also some successful cases of source apportionment used for soil heavy metals (Schaefer and Einax, 2016; Vaccaro et al., 2007; Xue et al., 2014). Apportioning sources of soil heavy metals could help in understanding the characteristics and contributions of different sources, so that appropriate control measures can be effectively targeted to reduce anthropogenic metal inputs to soil. However, the composition of metals in emissions differs depending on the source, and different metals have different toxicities. Thus, the sources should be prioritized for control and management based on the potential health risk of each source, rather than the contribution of each source to the environmental levels of heavy metals, which we proposed here for the first time.

Jiangsu Province has a highly developed economy, being located in the eastern coastal region of China. The study area, atypical rural-industrial town in southern Jiangsu, has a complex industry structure, with agricultural activities of planting, feeding, and aquaculture, and various industrial activities, such as production of photovoltaic electronics, bio-pharmaceutical products, mechanical metallurgy, electronic components, textile clothing, as well as printing and dyeing industries. The town also has the advantage of good transportation, with two first class roads passing through the core of the territory, connecting the commercial cities of Suzhou and Wuxi. Diverse industries and heavy traffic, which provide various heavy metal emission sources, made challenging to quantify the appropriate sources. Fortunately, the contamination history and related information of source changes were captured during our 10 years of researching this area (Cao et al., 2010; Jiang et al., 2015). Combining this knowledge with auxiliary methods, such as spatial analysis, correlation analysis, and others, apportioned sources could be correctly interpreted. Previous investigations have found that inhabitants were confronted with potential health risks from exposure to heavy metals in soils and self-planted rice and garden vegetables (Cao et al., 2010; Jiang et al., 2015). However, these samplings were conducted in 2008 and 2009. Several policies, such as rectification of polluted industries, transformation of the energy structure, and the termination or emigration of heavily polluting enterprises have been implemented since September 2010. In order to measure the reform effect, heavy metal concentrations in soil samples needed to be analyzed, and the relative health risks estimated.

The three main objectives of this study were: (1) to investigate the concentration distribution of major and trace metals in soil in the study area; (2) to assess the health risk of residents exposed to seven heavy metals—lead, copper, zinc, arsenic, chromium, cadmium, and nickel—in soils; and (3) to apportion potential sources of these common heavy metals and quantify their contributions using a PMF model combined with geostatistical analysis, land use investigation, and historical industrial information, and further to evaluate the health risks imposed by each source.

2. Materials and methods

2.1. Study area

The studied town, X, with an area of 104.26 km^2, belongs to Changshu City, Jiangsu Province, China. The region has a north subtropical humid monsoon climate with an average annual temperature of 16.6 °C and mean annual precipitation of about 1320 mm. The predominant and secondary prevailing wind directions are east-southeast and east-northeast, respectively. The annual average wind speed is 3.7 m/s. The main soil type is gleyic clayey paddy soil derived from lacustrine deposits. The study area belongs to the well-developed regions in the east of China. The area has been undergoing rapid and intense industrialization and urbanization over the past two decades. Primary, secondary, and tertiary industries currently coexist in the area.

2.2. Soil sampling and chemical analysis

A total of 105 topsoil samples (0–20 cm) were taken from the centers of the 1 km grid squares in October 2014. The average temperature was 19.6 °C during sampling period. Soil samples were collected on non-rainy days. The distribution of all samples is presented in Fig. 1. Each sample (about 1 kg dry weight) was a composite of five subsamples from nearby sites (approximately 5 m apart). Samples were packed into polyethylene bags and brought back to the lab. The soil samples were then air-dried, ground, sieved, and digested with a typical concentrated acid mixture (HNO_3, HF, and $HClO_4$). The details of the laboratory analyses of soil were the same as described in our previous research (Cao et al., 2010; Jiang et al., 2015). Concentrations of 15 elements (Mg, K, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb, and Pb) were measured by inductively coupled plasma-mass spectrometry (ICP-MS; Agilent 7500a, USA). For quality control (QC) and quality assurance (QA), blank control, duplicate samples, and standard reference soils (GBW07419; Center for Certified Reference Materials, China) were used. The detection limits of Mg, K, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Cd, Sb, and Pb were 0.228, 0.218, 0.010, 0.070, 0.009, 0.230, 0.010, 0.018, 0.023, 0.020, 0.009, 0.021, 0.010, 0.023, and 0.023 mg/kg, respectively. The results measured for the standard reference soils were within uncertainty ranges of the certified values. The relative standard deviations (RSD) for soil properties of duplicates were <3%.

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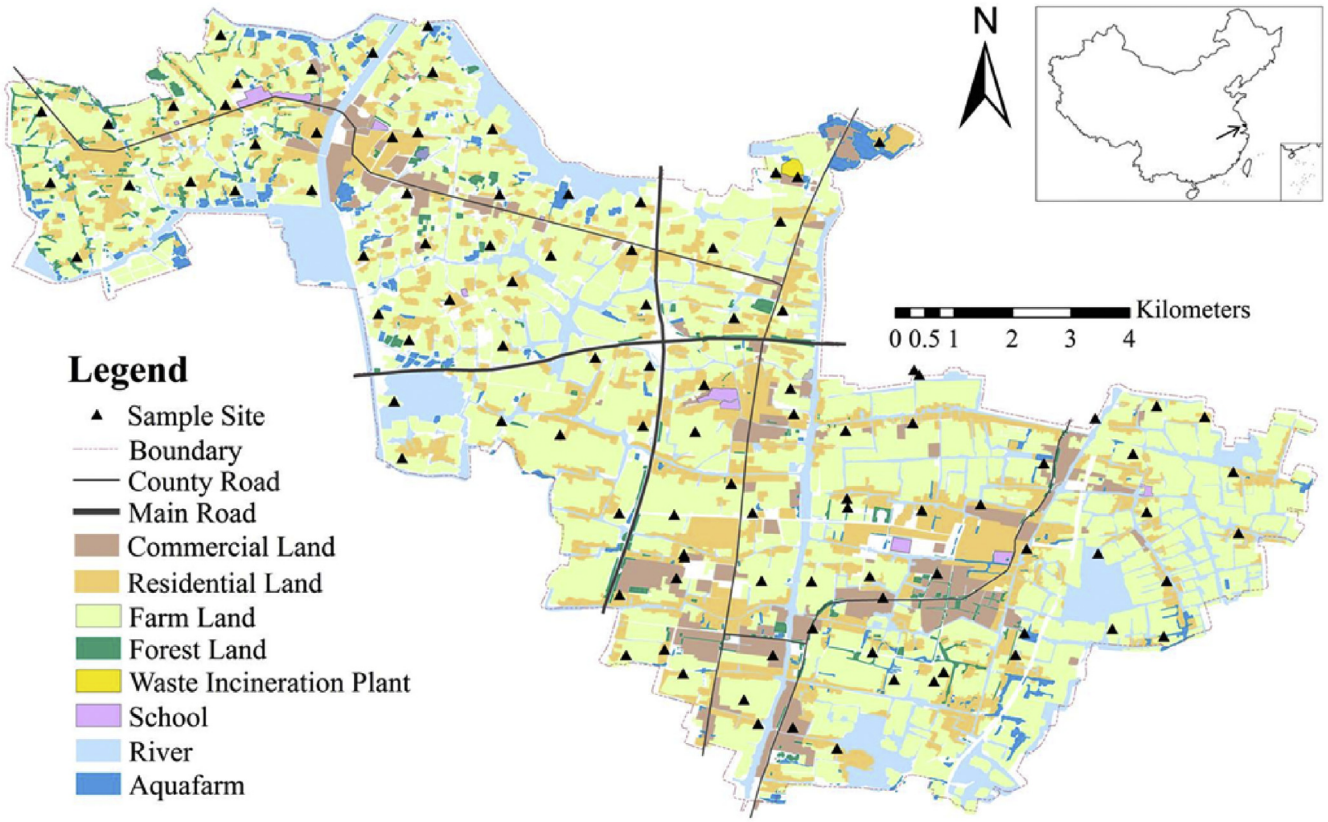

Fig. 1. Map of study area and location of soil sampling sites in town X (n = 105).

2.3. Enrichment factor

Enrichment factors (EFs) are used to assess pollution levels, and to evaluate the degree of human impact (Loska et al., 2004). It was calculated as Eq. (1) based on the equation suggested by Buat-Menard and Chesselet (1979).

$$EF = \frac{\left(C_i / C_{ref}\right)_{sample}}{\left(C_i / C_{ref}\right)_{background}} \quad (1)$$

where C_i is the concentration of the ith metal element (mg/kg); C_{ref} is the concentration of reference element for normalization (mg/kg). Elements such as Sc, Mn, Ti, Al, and Fe were usually chosen as references (Amil et al., 2016; Hsu et al., 2016; Kara et al., 2014; Namaghi et al., 2011; Szolnoki et al., 2013). In this study, Mn was adopted as a reference because of its relatively high concentration and stability in the crust (Tasdemir and Kural, 2005). According to the value of EF, soils can be classified into 5 levels: minimal enrichment ($EF < 2$); moderate enrichment ($2 \leq EF < 5$); significant enrichment ($5 \leq EF < 20$); very high enrichment ($20 \leq EF < 40$); and extremely high enrichment ($40 \leq EF$) (Sutherland, 2000).

2.4. Health risk assessment of heavy metals in soil and their emission sources

Health risk assessment connects the levels of contaminants in the environment with a probability of toxic effects for a human population. The hazard quotient (HQ) and cancer risk (CR) were used to quantitatively characterize non-carcinogenic and carcinogenic risks, respectively, of exposure to individual heavy metals (USEPA, 2011). For the soil, three exposure pathways were considered: (1) ingestion—directly through soil ingestion and indirectly through consumption of homegrown produce; (2) dermal contact; and (3) air inhalation via soil vapor. For non-carcinogens, the average daily intake (ADD) of a heavy metal via each pathway could be calculated as Eqs. (2)–(4) (USEPA, 2011). Components of the HQ were determined by dividing ADD by the corresponding reference dose (RfD) of each exposure pathway. The sum of all components was finally applied to assess any non-carcinogenic effects (Eq. (5)). For carcinogens, the lifetime average potential daily dose (LADD) was multiplied by the corresponding cancer slope factor (SF) to yield a level of carcinogenic risk via each exposure pathway (Eqs. (7)–(9)) (USEPA, 2011). CR was determined by summing the carcinogenic risks for each element (Eq. (10)). Since there is a lack of evidence indicating interactive effects of heavy metal mixtures, a total hazard index (THI) can be defined by adding the HQ of each element, as described by Eq. (6) (USEPA, 2011). Similarly, the total cancer risk (TCR) is calculated by summing the individual CR across all heavy metals using Eq. (11) (USEPA, 2011).

$$ADD_{ing} = \left(\frac{C \times IR_s \times EF \times ED}{BW \times AT} + \frac{C \times TF \times IR_p \times EF \times ED}{BW \times AT}\right) \times 10^{-6} \quad (2)$$

$$ADD_{dermal} = \frac{C \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \quad (3)$$

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$$ADD_{inh} = \frac{C \times IR_i \times EF \times ED}{PEF \times BW \times AT} \quad (4)$$

$$HQ = \sum \frac{ADD_x}{RfD_x} \quad (5)$$

$$THI = \sum HQ_y \quad (6)$$

$$LADD_{ing} = \left(\frac{C \times IR_s \times EF \times ED}{BW \times LT} + \frac{C \times TF \times IR_p \times EF \times ED}{BW \times LT}\right) \times 10^{-6} \quad (7)$$

$$LADD_{dermal} = \frac{C \times SA \times AF \times ABS \times EF \times ED}{BW \times LT} \quad (8)$$

$$LADD_{inh} = \frac{C \times IR_i \times EF \times ED}{PEF \times BW \times LT} \quad (9)$$

$$CR = \sum (LADD_x \times SF_x) \quad (10)$$

$$TCR = \sum CR_y \quad (11)$$

where ADD_{ing}, ADD_{dermal}, and ADD_{inh} are the average daily intake from ingestion, dermal contact, and inhalation, respectively (mg/kg/day); C is the concentration of trace metal in the soil (mg/kg); IR_s and IR_p are the ingestion rate of soil and homegrown produce, respectively (mg/day); TF is a soil-to-plant transfer factor (unitless); IR_i is the inhalation rate of soil (m^3/day); EF is the exposure frequency (day/year); ED is the exposure duration (year); BW is the body weight of the exposed individual (kg); AT is the time period over which the dose is averaged (day); PEF is the emission factor (m^3/kg); SA is the exposed skin surface area (cm^2); AF is the adherence factor (kg/cm^2/day); ABS is the dermal absorption factor (unitless); x and y indicate the number of exposure pathways and contaminants, respectively; RfD is the corresponding reference dose (mg/kg/day); SF is the slope factor (per mg/kg/day); and LT is the average lifetime (day). Parameter values for children (aged 1–17) were different from those for adults (aged 18-). Details of parameters and the values used in the health risk assessment model are given in Table S1. Among various parameters, TF was calculated from detected concentrations in soil and in several species of homegrown produce, and IR_p was determined from surveyed food consumption data for this study area in our previous study (Cao et al., 2010; Jiang et al., 2015). Table S2 lists the reference dose and slope factor of trace metals for use in the health risk assessment.

For the emission sources, only direct exposure pathways (soil ingestion, inhalation, and dermal contact) were considered. Both non-carcinogenic and carcinogenic risks were estimated as mentioned above by using the concentration of the individual metal (profile) of each source for C in Eqs. (2)–(9).

2.5. Positive matrix factorization

Positive matrix factorization (PMF), as a typical receptor model, has been recommended by the U.S. Environmental Protection Agency (USEPA) as a general apportionment modeling tool. It can be used without source profiles as inputs. Its remarkable features are non-negativity constraints and using uncertainty to weigh each data point individually (Norris et al., 2014). A correlation matrix and covariance matrix were used to simplify initial high-dimensional variables. The original matrix X(i × j), was decomposed into a contribution matrix, G(i × k), a source profile matrix, F(k × j) and a residual error matrix, E(i × j), as follows:

$$X_{ij} = \sum_{k=1}^{p} G_{ik} F_{kj} + E_{ij} \quad (12)$$

where x_{ij} is the concentration of the jth chemical element in the ith sample; g_{ik} is the contribution of the kth source for i number of samples; and f_{kj} is the concentration of the jth chemical element in the kth source. The residual error matrix e_{ij} is obtained by minimizing the object function Q:

$$Q = \sum_{i=1}^{n} \sum_{j=1}^{m} \left(\frac{e_{ij}}{u_{ij}}\right)^2 \quad (13)$$

where u_{ij} is the uncertainty in the jth chemical element of sample i. When the concentration of a chemical element was below or equal to the corresponding method detection limit (MDL), the uncertainty was calculated as:

$$Unc = 5/6 \times MDL \quad (14)$$

Otherwise, it was calculated as:

$$Unc = \sqrt{(\sigma \times c)^2 + MDL^2} \quad (15)$$

where σ is relative standard deviation; and c is the concentration of chemical element.

The PMF software (Ver. 5.0, USEPA) was used for source apportionment. Multilinear engine-2 (ME-2) and a PMF script file were used in the software to solve the problem of minimizing the Q value (Norris et al., 2014).

2.6. Data processing and spatial analysis

Geostatistical methods are common approaches to characterize the spatial distribution of contaminant concentrations in soil (Davis et al., 2009; Facchinelli et al., 2001; Zou et al., 2015). They are also used for describing the spatial distribution of source contributions in this study. The basic principle is to make an unbiased estimate for the values of unsampled locations by considering their spatial correlation with the sampled points and minimizing the variance in the estimation error. Ordinary kriging (OK), the most robust and common interpolation method, was used. A spherical model was chosen to fit the experimental semi-variogram.

The statistical software package SPSS version 22 for Windows (SPSS Inc., Chicago, IL, USA) and Microsoft Excel, 2007 (Microsoft, 2007) were used for all of the statistical computations. ArcGIS version 10.2 (ESRI Inc., Redlands, CA) was used for mappings.

3. Results and discussion

3.1. Concentrations of metal elements in soil samples

Basic statistics for several metals in top soils are shown in Table 1. The mean and median concentrations of Co, Pb, Cu, Cr, Cd, and Ni were higher than their corresponding average background values (ABV) of soils in Jiangsu (CNEMC, 1990). The average accumulation rate (C_{mean}/ABV) for Co, Pb, Cu, Cr, Cd, and Ni were 1.06, 1.07, 1.37, 1.51, 1.22, and 1.07, respectively. It was therefore inferred that the study area has been mildly contaminated by human activities. Among the samples, concentrations of Cu in 4 samples (3.8%) and Ni in 12 samples (11.4%) were beyond the corresponding Grade II criterion of the National Environmental Quality standards

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Table 1
Statistical summary of element concentrations in soil samples (n = 105, % for Mg, K, and Fe, mg/kg for other elements).

	Mg	K	V	Se	Mn	Fe	Co	Sb	Pb	Cu	Zn	As	Cr	Cd	Ni
Minimum	0.12	1.06	60.77	0.03	181.25	0.83	10.33	0.80	14.46	18.41	40.23	5.36	55.21	0.06	26.05
Mean	0.22	1.64	82.77	0.12	352.77	1.16	12.76	4.14	31.41	31.60	61.13	7.46	86.38	0.11	34.93
Median	0.21	1.66	83.13	0.12	347.91	1.16	12.87	3.87	30.48	30.86	57.21	7.36	86.24	0.11	34.31
Maximum	0.45	2.09	101.98	0.21	692.00	1.42	15.14	9.47	55.91	55.58	102.14	10.86	120.66	0.27	60.06
Standard deviation	0.07	0.22	7.55	0.03	102.10	0.10	1.32	2.57	8.65	8.83	14.19	1.17	12.17	0.03	5.18
Coefficient of variation	0.31	0.13	0.09	0.25	0.29	0.09	0.10	0.62	0.28	0.28	0.23	0.16	0.14	0.27	0.15
Average background of Jiangsu	0.84	1.81	83.4	0.222	585	2.95	12.6	1.04	26.2	22.3	62.2	10.0	77.8	0.126	26.7
Average of China [a]	–	–	–	–	–	–	–	–	31.2	27.1	79	12.1	68.5	0.225	29.6
Average of China [b]	0.78	1.9	82	0.29	582	3	13	1.2	27	23	74	11	61	0.097	27
Average of China [c]	–	–	–	–	–	–	–	–	145.77	93.53	301.72	26.67	114.00	3.53	–
Chinese soil criteria (Grade II)	–	–	–	–	–	–	–	–	250	50	200	30	150	0.3	40
Dutch target values	–	–	42	0.7	–	–	9	3	85	36	140	29	100	0.8	35

[a] Data taken from Chen et al. (2015).
[b] Data taken from Teng et al. (2014).
[c] Data taken from Duan et al. (2016), which focused on soil pollution.

for soils in China (CEPA, 1995). The highest concentrations of Cu and Ni were about 1.1 and 1.5 times their corresponding limits. Concentrations of other heavy metals in all samples were less than the standard limits. The results compared with the Chinese soil standard indicated that no obvious heavy metal pollution was found in the study area soil. However, in comparison with target values of Dutch soil guidelines (VROM, 2000), V, Co, Sb, and Cr also showed mild pollution. The coefficients of variation for Sb and Mg were relatively high, which might be a consequence of human activities. Metal concentrations in this study area are of the same order of magnitude as the national average level reported by other studies (Table 1), much less than those at typical pollution sites, such as mines, smelters, and electronic waste dumpsite soils (Li et al., 2014; Roy and McDonald, 2015; Song and Li, 2014).

The enrichment factors (EFs) of trace metals in soils have been calculated to differentiate elements originating from human activities and those from natural provenance and to assess the degree of anthropogenic influence. Results showed that the average EF values for Mg, K, V, Se, Fe, Co, Sb, Pb, Cu, Zn, As, Cr, Cd and Ni were 0.46, 1.63, 1.81, 0.97, 0.71, 1.80, 7.05, 2.17, 2.57, 1.76, 1.32, 2.03, 1.57, and 2.35, respectively, with the ranges of 0.19–1.04, 0.73–3.36, 0.76–3.66, 0.38–2.38, 0.37–1.26, 0.95–3.58, 1.07–26.23, 0.68–5.01, 0.85–7.21, 0.68–4.69, 0.59–2.25, 0.87–4.05, 0.51–3.10 and 1.14–6.09, respectively. The EF values of Mg and Fe were less than 1, indicating a natural origin. The values for K, V, Co, Zn and Cd were between 1.5 and 2, suggesting that the sources are likely to be anthropogenic (Zhang and Liu, 2002). The Pb, Cu, Cr and Ni values showed moderate enrichment, which indicated anthropogenic influences. In addition, with the highest EF value at greater than 5, Sb showed significant enrichment. This finding suggested that the soils in this study area were moderately affected by human activities.

3.2. Health risk assessment of soil

Seven heavy metals (Cd, Cr, As, Pb, Cu, Zn, and Ni) were considered in the health risk assessment because of their relatively strong toxicity to humans, detailed and published dose-response relationships (USEPA, 2016), and a previously quantified TF (Cao et al., 2010; Jiang et al., 2015). The results of non-carcinogenic and carcinogenic health risks posed by these heavy metals in soils for adults (aged 18-) and children (aged 1–17) via different pathways are shown in Table 2. Missing values are due to a lack of corresponding RfD or SF values.

The THI values were 6.11 and 3.62 for children and adults, respectively. Values greater than 1 indicate that the public may experience non-carcinogenic effects (USEPA, 2001). Children tended to have a higher probability than adults did. The HQ values for different heavy metals followed the order: Cr > As > Cu > Pb > Zn > Ni > Cd. Among these elements, HQ values of Cr and As exceeded 1, accounting for 48.96% and 30.66% of THI for adults, respectively. The HQ value of Cu for adults was 0.33 (9.00%), which nearly balanced the sum of HQ values of the remaining four heavy metals (11.38%). The ratios of HQ values to THI for children were similar to those for adults. Homegrown produce ingestion was the major exposure pathway for both adults and children, accounting for about 98.7% and 95.8% of their corresponding THI, respectively. The order of hazard indices was: homegrown produce ingestion > dermal contact > soil ingestion > air inhalation. The percentage of HQ by air inhalation was only about 0.01%. The order of contributions of the last three pathways was the same as in other studies (Chabukdhara and Nema, 2013; Chen et al., 2016; Zheng et al., 2010). Children had higher non-carcinogenic risks than adults through every pathway of every element, indicating that they are more susceptible to environmental contaminants. This might be due to the behavioral and physiological characteristics of children, including hand-to-mouth activities in soil and higher respiration rates per unit body weight.

Since Cu and Zn were not included in the carcinogenic category (USEPA, 2016), only the carcinogenic risks of the other five metals were estimated. According to available slope factors, all three pathways were contained in the risk estimation of As, but only one or two pathways were included in the estimation of the other metals.

The TCR values for adults and children were 9.78×10^{-4} and 4.03×10^{-4}, respectively (Table 2). The carcinogenic risks for both adults and children were higher than the maximum tolerable or acceptable risk (1×10^{-4} (Fryer et al., 2006)). Adults showed higher carcinogenic risks than children. Similar to the trend of HI values, the CR values for different heavy metals decreased in the order: Cr > As > Pb > Ni > Cd. The CR values of Ni and Cd for both adults and children, and Pb for children were below the negligible risk level of 1×10^{-6}, indicating no significant health effects. The CR values of Pb for adults and As for children stood in the range of acceptable risk (1×10^{-6}–1×10^{-4}). The CR values of As for adults and Cr for adults and children were 1.56, 8.2, and 3.36 times of the maximum tolerable risk, respectively. Measures should be taken to reduce the carcinogenic risk.

The carcinogenic risks derived from homegrown produce ingestion accounted for 99.4% and 98.2% of the TCR for adults and children, respectively, which were much higher than the carcinogenic risks due to other pathways in total. This indirect pathway has not been considered in the health risk assessment of soils in most studies (Chabukdhara and Nema, 2013; Chen et al., 2016; Luo et al.,

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Table 2
Estimations of non-carcinogenic (Hazard Quotient, HQ) and carcinogenic health risks (Cancer Risk, CR) from heavy metals in soil.

	Adults (aged 18-)					Children (aged 1–17)				
	soil ingestion	home-grown produce ingestion	air inhalation	dermal contact	total pathways	soil ingestion	home-grown produce ingestion	air inhalation	dermal contact	total pathways
Hazard quotient										
Pb	2.73E-03	1.16E-01		1.04E-03	1.20E-01	1.48E-02	1.87E-01		5.54E-03	2.07E-01
Cu	2.40E-04	3.25E-01		4.57E-05	3.25E-01	1.31E-03	5.43E-01		2.44E-04	5.44E-01
Zn	6.20E-05	1.15E-01		1.77E-05	1.16E-01	3.37E-04	1.93E-01		9.43E-05	1.93E-01
As	7.57E-03	1.10E+00		1.05E-03	1.11E+00	4.11E-02	1.84E+00		5.62E-03	1.89E+00
Cr	8.76E-03	1.74E+00	5.41E-04	2.50E-02	1.77E+00	4.76E-02	2.81E+00	5.58E-04	1.33E-01	2.99E+00
Cd	3.32E-05	6.13E-02	1.95E-06	1.89E-04	6.16E-02	1.80E-04	9.94E-02	2.01E-06	1.01E-03	1.01E-01
Ni	5.32E-04	1.14E-01	6.95E-05	1.12E-04	1.15E-01	2.89E-03	1.81E-01	7.17E-05	5.99E-04	1.85E-01
Total metals	1.99E-02	3.57E+00	6.12E-04	2.74E-02	3.62E+00	1.08E-01	5.86E+00	6.32E-04	1.46E-01	6.11E+00
Cancer risk										
Pb	2.55E-08	1.08E-06			1.10E-06	3.46E-08	4.36E-07			4.70E-07
As	1.07E-06	1.55E-04	6.32E-09	1.48E-07	1.56E-04	1.45E-06	6.50E-05	1.63E-09	1.98E-07	6.66E-05
Cr	4.12E-06	8.16E-04	2.04E-07		8.20E-04	5.59E-06	3.30E-04	5.25E-08		3.36E-04
Cd			3.85E-11		3.85E-11			9.94E-12		9.94E-12
Ni			1.65E-09		1.65E-09			4.25E-10		4.25E-10
Total metals	5.21E-06	9.72E-04	2.12E-07	1.48E-07	9.78E-04	7.08E-06	3.96E-04	5.46E-08	1.98E-07	4.03E-04

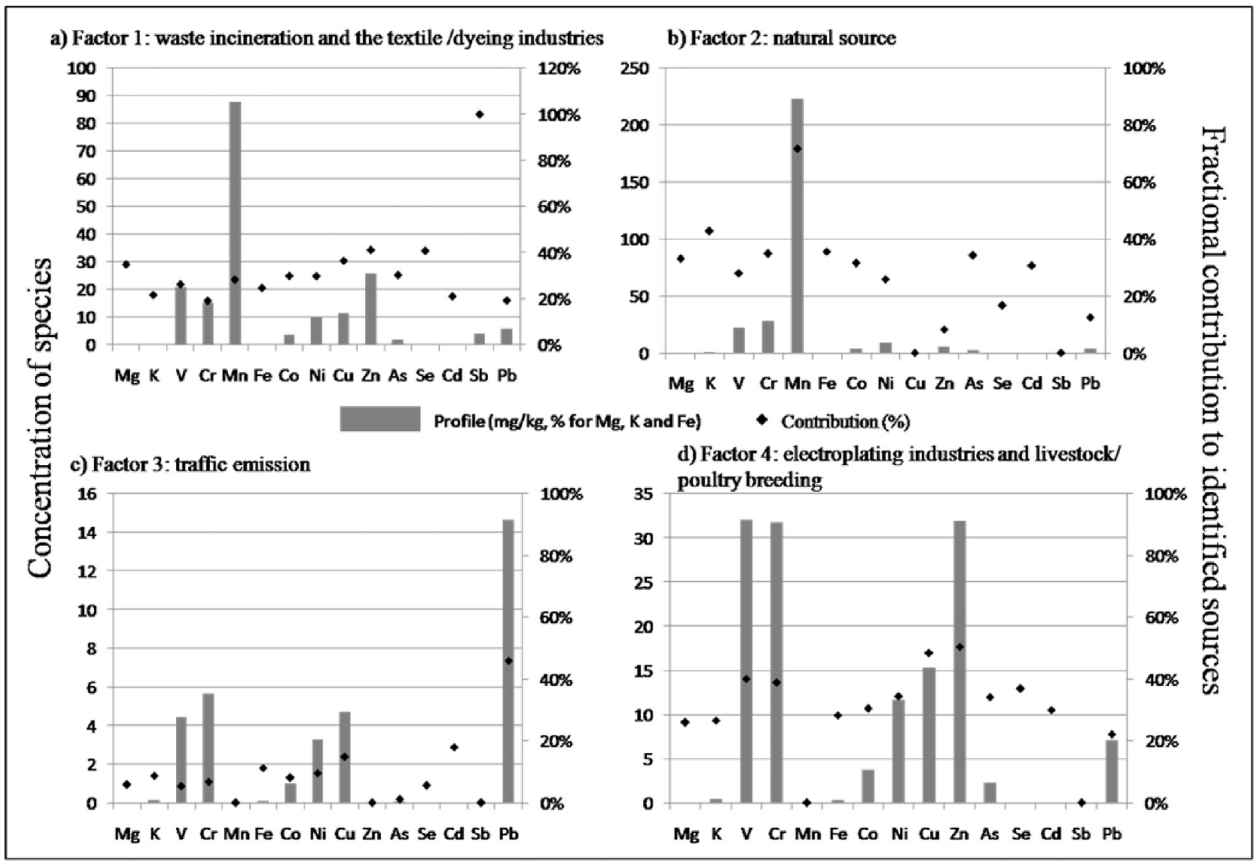

Fig. 2. Fractional concentrations and percentage contributions of four factor profiles derived from PMF.

2015), which may result in an underestimation of health risks due to metals in the soil. It may be reasonable for a health risk assessment of urban soil to ignore this indirect pathway, because daily food products in cities come from supermarkets and are grown elsewhere (Chabukdhara and Nema, 2013). However, in rural and rural-urban fringe zones, daily food, such as rice and vegetables, are self-planted and harvested to varying degrees. To assess the health risk of these soils, the exposure pathway from the soil to plants and ultimately to humans should not be ignored.

It should be noted that the health risks may have been overestimated. For example, we estimated the health risk of total As by using the RfD/SF of inorganic As; however, only approximately 60% of the total As in agricultural produce was inorganic As (Jiang et al., 2015), while the other 40% was organic As, which has a much lower

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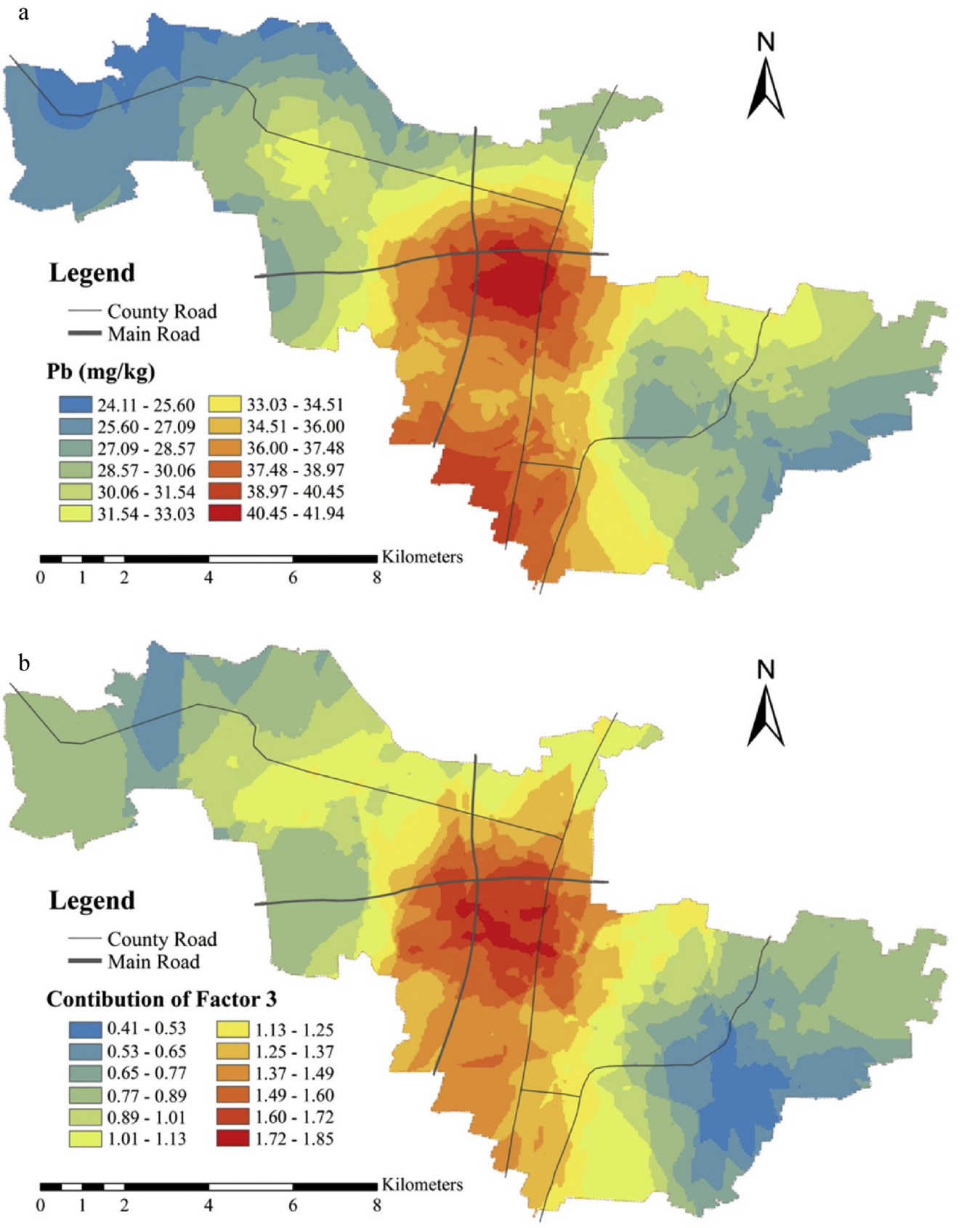

Fig. 3. Spatial distribution of: a) concentrations of Pb in soil; and b) normalized contributions of Factor 3 i.e. traffic emission source (the average of normalized contributions to all sample sites =1).

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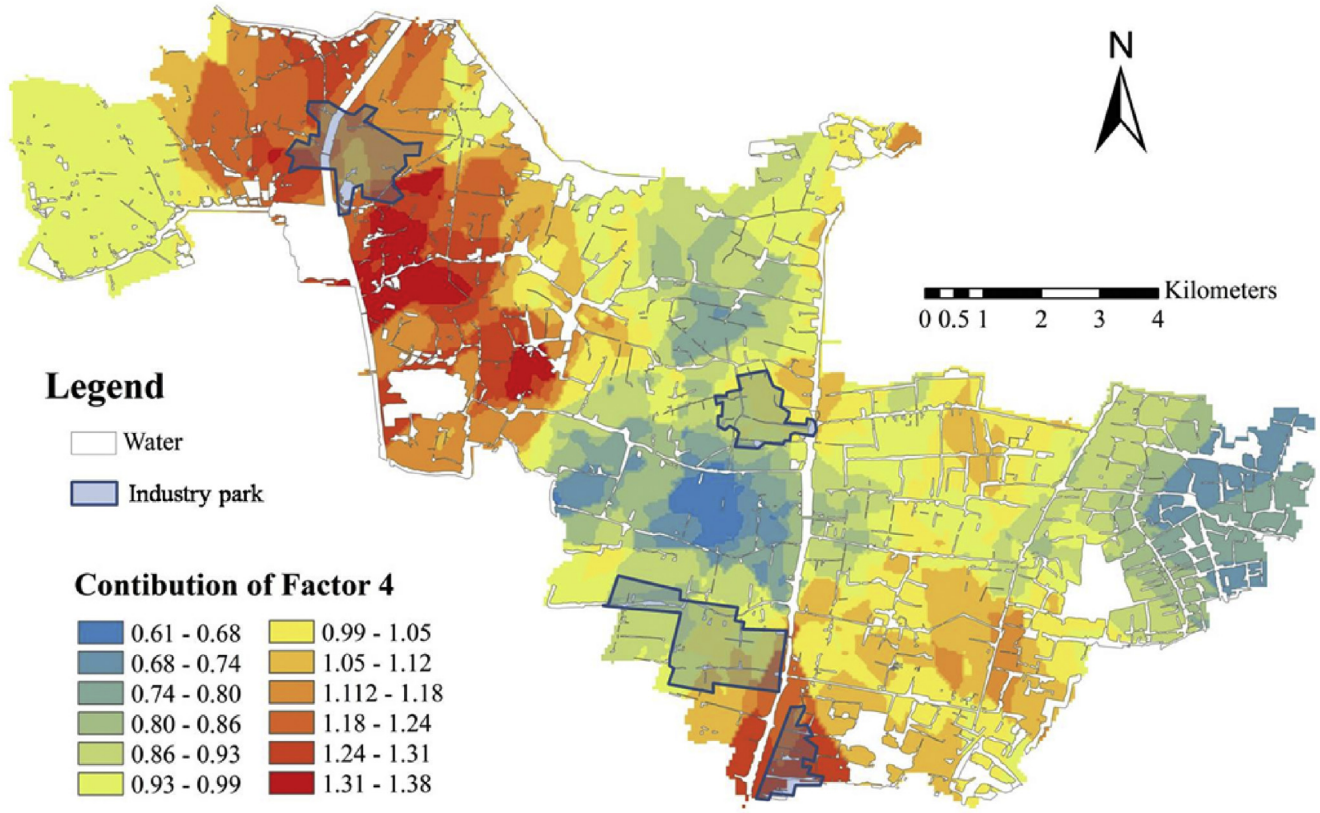

Fig. 4. Spatial distribution of normalized contributions of Factor 4 i.e. electroplating industries and livestock/poultry breeding (the average of normalized contributions to all sample sites =1).

Table 3
Estimations of non-carcinogenic (Total Hazard Indices) and carcinogenic (Total Cancer Risks) health risks via direct pathways (soil ingestion, inhalation, and dermal contact) for the apportioned sources.

	Factor 1: waste incineration and the textile/dyeing industries (28.3%)	Factor 2: natural source (45.4%)	Factor 3: traffic emission (5.3%)	Factor 4: electroplating industries and livestock/poultry breeding (21.0%)	Total risk
Total hazard indices of sources, THI_s					
Adults (aged 18-)	9.67E-03	1.47E-02	4.25E-03	1.66E-02	4.53E-02
Children (aged 1–17)	7.98E-02	7.80E-02	2.27E-02	8.85E-02	2.69E-01
Total cancer risks of sources, TCR_s					
Adults (aged 18-)	1.12E-06	1.80E-06	3.10E-07	1.98E-06	5.21E-06
Children (aged 1–17)	1.47E-06	2.37E-06	4.05E-07	2.60E-06	6.85E-06

toxicity compared to inorganic As. The RfD/SF of Cr(VI) was used in our risk assessment; however, Cr(VI) in agricultural produce might be reduced to Cr(III), a less toxic valence, under the acidic conditions in the stomach (Cao et al., 2010). At the current stage, a conservative estimation is often preferred to protect public health. The health risks of any given element, however, should be further analyzed by considering the toxicity of various species and valences as well as changes in species, valence and *in vivo* bioavailability.

3.3. Source apportionment and risk assessment

Concentration data (including 15 elements for 105 soil samples) and uncertainty data files (including sampling and analytical errors) were used as the input data for the PMF. Different values of the rotational parameter Fpeak (between −1 and +1, in steps of 0.1) were explored, and −0.1 was finally adopted to improve oblique edges. Four factors were identified, and their contributions are presented in Fig. 2.

The first factor was predominated by Sb, Zn, Se, Cu, Mg, and As, in descending order (Fig. 2a). This source accounted for 28.3% of the total variances. Antimony, with the highest weight, has an average concentration of 0.2 mg/kg in the earth's crust (Merian, 1991). The median concentration of Sb in soil is 0.1 mg/kg, ranging from 0.2 to 10 mg/kg globally, and is regularly less than 1 mg/kg (He and Wan, 2004). The average background value of Sb concentrations in China is in the range of 0.38–2.98 mg/kg (He and Wan, 2004). The average Sb content of soil in this study area was 4.14 mg/kg, with a coefficient of variation of 0.62, indicating a mild pollution compared with the background concentration. A connection with anthropogenic emissions was indicated.

Anthropogenic Sb could originate from mining, smelting, pesticides, and other materials containing Sb. According to the field

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investigation, there were no mining or smelting industries in the study area. Antimony in pesticides comes from the extracting process of As, which is an associated mineral (Steely et al., 2007). However, the concentrations of Sb and As in the study soil did not show a significant correlation (Table S3). Antimony was widely used in many kinds of industries, for instance as a hardener in metal or alloy producing, in polycondensation catalysts for polyethylene terephthalate production, and in plastics, fire prevention materials, battery plates, chemical pipelines and others. Different sizes of textile and dyeing enterprises were widely distributed in the study area. Antimony is used as a catalyst during the synthesis of polyester and a fire retardant during dyeing and finishing. Textile wastes are combusted on site near small family factories. Several months before sampling, the dyeing and finishing industries in Wujiang City, Jiangsu province had been notified to stop or restrict production because the Sb concentration in the Taipu River had exceeded regulatory limits (NBD, 2014). In addition, high contributions of Factor 1 were located downwind of the waste incineration plant (Fig. S1). Other elements with a high load are common pollutants of leachate from house refuse landfills, and the bottom and fly ash of waste incineration (Cheng et al., 2015; Hu et al., 2013; Rimmer et al., 2006). According to the above analysis, Factor 1 was determined to be an anthropogenic component due to waste incineration and the textile and dyeing industries.

The second and biggest factor, which accounted for 45.4% of the total variance, consisted predominantly of Mn, K, Fe, Cr, As, Mg, and Co (Fig. 2b). Concentrations of these metals were not high in the study area, and the coefficients of variation were small. These elements, especially Mn, K, Fe, and Co, are associated with the crustal material source (Cass and McRae, 1983; Ostro et al., 2007). Much research has also reported that Mn, Cr, and Co in soils would originate from soil parent materials (Chen et al., 2016; Salonen and Korkka-Niemi, 2007; Xue et al., 2014). Thus, this factor was identified as a natural source.

The third and smallest factor, which accounted for 5.3% of the total variance, had higher relative concentrations for Pb than other elements. The contributions of Factor 3 (Fig. 3b) had a distribution similar to that of the Pb concentration (Fig. 3a). The high concentration of Pb near the main road agreed with other studies showing that Pb was a primary marker of transportation (Arditsoglou and Samara, 2005; Zhu et al., 2013). Additionally, for this factor, Cu and Ni may result from traffic emission sources because more than 90% of Cu from traffic emission were due to brake wear (Hjortenkrans et al., 2006), and more than 80% of Ni from traffic emission were due to exhaust emissions (Johansson et al., 2009). Factor 3 can therefore be interpreted as the traffic emission source.

The fourth factor, which explained 21.0% of the total variance, was associated with Zn, Cu, V, and Cr. These metals are widely used in electroplating industries in the study area, and the distribution of the contributions of Factor 4 (Fig. 4) indicate that high contributions occurred near industrial parks including businesses involved in electroplating materials, manufacture of electric appliances, electronic components, and plastics, and knitting factories. Although the polluting electroplating and chemical enterprises have been shut down or moved out of the study area, the impact of heavy metals on the soil will not disappear immediately. Factors containing Cu and Zn are also often attributed to farming related sources (Nicholson et al., 2003), such as pesticides and fertilizers, meaning that the contributions to farmland should be different from those to uncultured land. However, in this study, no significant difference was shown among the contributions of Factor 4 to different land use types (Table S4). The other potential source considered was family livestock and poultry breeding. Manure and effluents are directly emitted to the surroundings at will in these family-run, non-intensive livestock farms. Most Cu and Zn in animal feed would enter the soil along with other wastes generated by these operations (Cang et al., 2004). This source might have made a small contribution. Based on the available information, Factor 4 was taken to be a combination of electroplating industries and livestock and poultry breeding.

The health risk assessment of sources showed that for adults, Factors 1–4 accounted for 21.4%, 32.4%, 9.4%, and 36.8% of the non-carcinogenic risks, respectively, and 21.4%, 34.7%, 5.9%, and 38.0% of the cancer risks, respectively (Table 3). For children, Factors 1–4 accounted for 29.7%, 29.0%, 8.4%, and 32.9% of the non-carcinogenic risks, respectively, and 21.5%, 34.6%, 5.9%, and 38.0% of the cancer risks, respectively (Table 3). Factor 4 caused the greatest health risk, but was only third in explanation of the total variance with the four-source profile. It was therefore inferred that human health might not be protected if the prior control of pollution sources was simply determined by emission amounts. Health risk assessment of emission sources should be an important evaluation index of harmfulness and included in pollution source management. Since electroplating industries had already been shut down or moved out, the next steps will be to focus on regulation of waste incineration and the textile and dyeing industries, as well as livestock and poultry breeding.

4. Conclusions

Although the average concentrations of all 15 metals in soil did not exceed the corresponding national standard, accumulations of Cr, Cu, Cd, Pb, Ni, and Co were observed in the study area. Seven soil heavy metals (Cd, Cr, As, Pb, Cu, Zn, and Ni) in total posed potential non-carcinogenic risks to both adults (THI = 3.62) and children (THI = 6.11) via soil inhalation, dermal contact, and direct/indirect ingestion. The total carcinogenic risk levels of soil in the study area were about 10 times the acceptable risk limit, mainly due to As (1.56×10^{-4} for adults and 6.66×10^{-5} for children) and Cr (8.20×10^{-4} for adults and 3.36×10^{-4} for children). Homegrown produce ingestion, the most important exposure pathway, contributed about 98% to health risks of soil heavy metals. More attention should be paid to this indirect exposure route, especially in rural soils. The health risks of the sources of trace metals in soil were estimated for the first time based on quantitative source contributions apportioned by PMF. The natural source (Factor 2) explained the majority of the total variance of metal concentrations (45.4%), but its health risk was less than that of the electroplating industries and livestock and poultry breeding (Factor 4). The other anthropogenic sources were waste incineration and the textile/dyeing industries (Factor 1), ranking second and third in variance explanation (28.3%) and source health risk (23.5%), respectively. Traffic emission (Factor 3) contributed the least to both variance explanation (5.3%) and source health risk (7.4%). To protect the soil environment and public health in the study area, government regulators should make efforts to manage these anthropogenic sources and reduce the effect of soil metal pollution on human health risks.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2016.11.088.

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