



Spatial variation of heavy metals and their ecological risk and health risks to local residents in a typical e-waste dismantling area of southeastern China

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Abstract There is an increasing concern that soils in e-waste recycling regions are severely contaminated by unregulated e-waste dismantling activities. Hence, it is urgent to reveal the spatial variation of hazardous elements in arable lands close to e-waste stacking and dismantling areas and their potential risks to human beings. We collected 349 topsoil samples based on an intensive grid of 100 m × 100 m in southeastern China. The average concentrations of heavy metals were 1.25 (Cd), 35.44 (Ni), 77.68 (Cr), 77.38 (Pb), 122.14 (Cu), 203.39 (Zn), 0.21 (Hg), and 4.74 (As) mg kg⁻¹, respectively. Compared to the risk screening values of hazardous elements in Chinese agricultural land, Cd and Cu were severely accumulated in the soils. The results of ecological risk analysis revealed that Cd posed the crucial risk among the studied elements. However, the levels of non-carcinogenic and carcinogenic risk were still within the acceptable quantity for adults. Spatial distribution by kriging interpolation displayed that the heavy metals were mainly distributed close to e-waste dismantling sites.

Keywords Anthrosols · Heavy metals · Spatial distribution · Human health risk · E-waste dismantling

Introduction

Environmental pollution caused by anthropogenic activities has attracted great attention from environmental researchers worldwide, among which soil heavy metal contamination is an ongoing concern for its high toxicity, non-biodegradability, and persistence (Kamunda et al., 2016; O'Connor et al., 2018; Purchase et al., 2020; Shi et al., 2020; Zhang et al., 2013). Harmful heavy metals threaten the agricultural ecosystem health and impair human health through food intake, inhalation, and skin contact (Lü et al., 2021; Vatanpour et al., 2020; Zheng et al., 2020). Long-term intake of low-dose heavy metals by residents has potential carcinogenic health risk, causing them to develop a variety of cancers. Residents who are exposed to Cd for a long time can develop lung cancer, breast cancer, and kidney dysfunction (Zhou et al., 2021); As may result in skin cancer (Zhao et al., 2021) and liver cancer (Lin et al., 2013); Pb and Hg have adverse effects on central nerve system (Yu et al., 2019a). Long-term consumption of vegetables contaminated by heavy metals is related to chronic kidney disease in the North Central Province of Sri Lanka (Kulathunga et al., 2021). Ni, Cr, Cu, and Zn exceed safe thresholds and contribute to

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non-carcinogenic risks such as neuropathy and cholesterol unbalanced (USEPA, 2000; Zhang et al., 2012).

The sources of heavy metals in soils mainly include parent materials and anthropogenic activities such as industrial production, traffic emissions (road dust, vehicle exhaust), mining, and agricultural activities. For the transportation issue, the main pathways include dry, wet deposition and wastewater dispersion. Some researchers found that wind dispersion affected soil heavy metal concentration; however, this effect was often more significant in arid regions such as deserts for enough dry sand particles to be carried by the wind to transport the heavy metals into the soil (El Hadi et al., 2015; Mokhtari et al., 2018). Their contamination in arable soils is more severe, so it is easy to accumulate in the food chain. Heavy metals in rural soils are mainly derived from pesticide and fertilizer application, irrigation with contaminated water, mineral ore extraction, sewage sludge, atmospheric deposition, and waste disposal (Hou et al., 2017; Huang et al., 2018; Yu et al., 2019b). With the speedy development of electronic technology, electronic equipment waste pollutants become a major factor affecting the quality of local farmland soil (Anselm et al., 2021; Olusegun et al., 2021; Purchase et al., 2020).

The amount of e-waste such as unwanted, non-working, and obsolete electronic products is gradually increasing as its lifespan becomes shorter due to the rapid development of persistently technological innovation (Damrongsiri et al., 2016; Li et al., 2011). In 2019, the total amount of e-waste worldwide reached 53.6 Mt and was expected to approach 74 Mt by 2030 (Forti et al., 2020). China generated the largest amount and imported 50% of e-waste in the world (Zhao et al., 2019). The e-waste dismantling points of family-operated recycling facilities are scattered in various villages. They are concentrated, scattered, and located near the dismantling sites. They belong to non-point source pollution rather than point source pollution. Basic recycling practices include manual dismantling of electronic products, plastic clipping and melting, burning to recover precious metals, and using chemical dissolution to recover gold and other precious metals, causing a large number of contaminants to be released into the surroundings (Tang et al., 2010; Song & Li,

2014; Wang et al., 2015; Salam & Varma, 2019). Specifically, Tang et al. (2014) found that the mean soil Cd and Cu values were high in some e-waste disposal areas. Zhao et al. (2019) found that heavy metals were significantly accumulated in the topsoil of Wenling city (another famous e-waste dismantling area) in China. Heavy metal pollution caused by electronic waste dismantling activity is an environmental issue not only in China, but also in Asian and African countries such as India (Subramanian et al., 2015), Thailand (Amphalop et al., 2020), Indonesia (Soetrisno & Delgado-Saborit, 2020), and Ghana (Michael Ackah, 2019; Cao et al., 2020).

Hazardous metal contamination in arable lands causes potential risks to the environment and imposes a great threat to public health (Li et al., 2011; Yin et al., 2018; Zhao et al., 2019). Pollution indices: pollution index (PI), potential ecological risk index (PERI), risk index (RI), geoaccumulation index (I_{geo}) were generally regarded as useful tools for comprehensively assessing soil contamination levels (Mazurek et al., 2017; Joy & Uchennam, 2017; Zhao et al., 2020; Jin et al., 2021; Fu et al., 2022). Meanwhile, carcinogenic and non-carcinogenic models were applied to quantify the human health risk of polluted soils (Du et al., 2019; Lü et al., 2021).

Considering the mix of natural and anthropogenic factors, geographic information system and other geostatistical analyses (such as ordinary kriging spatial interpolation) are powerful methods for evaluating heavy metal spatial distribution patterns due to their visual display and geostatistical derivation (Ha et al., 2014; Jin et al., 2019; Wu et al., 2014). Researchers are growing to use these powerful tools in combination to measure and predict pollution status (Majumdar & Biswas, 2016; Zhao et al., 2020).

The main source of soil heavy metal pollution in the study area is e-waste, which is a specific issue. Comprehensive and diversified spatial analysis methods and health risk assessment methods were adopted to study heavy metals in the farmland soil in the study area. The main objectives of this study were (1) to characterize the status and the spatial patterns of heavy metals in arable land soils of the study region and (2) to analyze the heavy metal ecological risk and the health risks to local residents through various pathways.

Materials and methods

Study area

A typical e-waste recycling area in southeastern Zhejiang Province, China, was chosen as the study site. The annual mean precipitation is about 1583 mm, and the annual mean temperature is around 17.6 °C. The topographic type of the study area is mainly plain, and high hilly land distributed in the northwest part, and lowland in the southeast part, and suitable for the growth of a variety of crops. It is also an intensive e-waste recycling area with massive electronic waste, including discarded television sets, computers, phones, microwave ovens, and old refrigerators. Prior informal dismantling activities are ubiquitous, which may cause serious soil contamination and severe potential risks to residents.

Soil sampling and analysis

A grid of 100 m×100 m was used (Fig. 1), and a total of 349 topsoil samples (0–20 cm) were collected from farmland. All samples were dried at room temperature and sieved through <2-mm nylon mesh after clearing the twigs and pebbles away. Part of the soils was ground to pass the 0.149-mm nylon mesh.

Chemical properties of soil—pH, organic matter, alkaline hydrolyzed nitrogen (HN), available phosphorus

(AP), and available potassium (AK)—were determined based on the *Analysis Methods of Soil and Agricultural Chemistry* by Lu (2000). Each sample was tested in duplicate. The indicator of relative deviations of the duplicates analysis for heavy metals was controlled within 10%. To ensure analytical quality, the soil standard materials such as GSS-22 were used in the analysis. The concentrations of heavy metals were 0.065 ± 0.012 (Cd), 26 ± 1 (Ni), 57 ± 3 (Cr), 26 ± 2 (Pb), 18.3 ± 0.8 (Cu), 59 ± 2 (Zn), 0.020 ± 0.002 (Hg), and 7.8 ± 0.5 (As) mg kg⁻¹ for soil standard materials.

Soil quality assessment methods

The degree of single soil heavy metal contamination can be calculated as follows: (Zhao et al., 2020)

$$P_i = C_i/S_i$$

where P_i represents the pollution index of pollutant i , C_i is the actual concentration of target pollutant (mg kg⁻¹), and S_i represents the background values of pollutant i in soil. The P_i can be used to determine the level, which can be classified into five levels to indicate heavy metal contamination in soil, as described by Yang et al. (2013).

Nemerow multi-factor index is a comprehensive pollution index, which might be essential and scientific in reflecting soil contamination levels

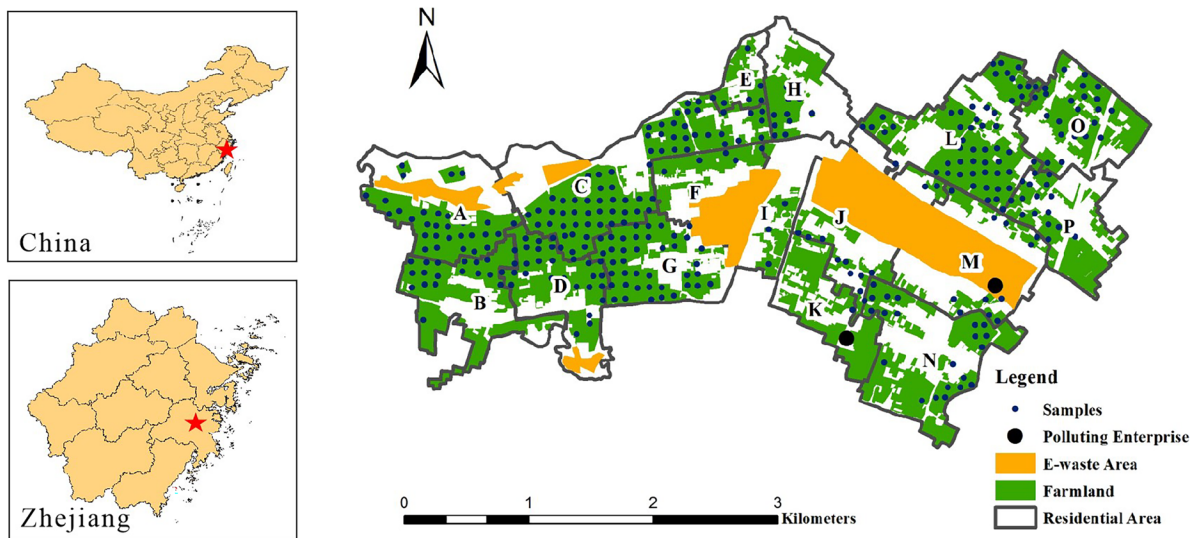


Fig. 1 Location of the study area and samples

(Zhao et al., 2015). The index can be formulated as:

$$P = \sqrt{(P_{iMax}^2 + P_{iAve}^2)/2}$$

where P represents the integrated pollution degree of soil, P_{iMax} is the maximum value of P_i , and P_{iAve} is the mean of P_i . The soil quality can be classified into four levels (Yang et al., 2013; Zhao et al., 2019).

Ecological risk evaluation

Hakanson (1980) provided the potential ecological risk index (PERI) to indicate the degree of ecological risk. The PERI for individual element (E_i) and comprehensive risk index (RI) can be calculated by the following equations:

$$RI = \sum_{i=1}^n E_i = \sum_{i=1}^n T_r^i \times P_i$$

where P_i represents the single pollution index of heavy metal i and T_r^i is the toxic-response factor for a given metal. The reference values of heavy metal toxicity and the grading standard of E_i and RI can be found in other studies (Hakanson, 1980; Yuan et al., 2014).

Human health risk assessment

Adult farmers are most exposed to contamination for spending most of their daily time in fields (He et al., 2019). In general, the human health risk assessment in this study was carried out considering three pathways: accidental oral ingestion of soil, inhalation of air, and dermal exposure (Xiao et al., 2015; Zheng et al., 2020) which may lead farmers to be at risk of being exposed to heavy metals in soil. The equations of average daily doses (ADDs) to adults through three pathways and related information could be found in Han et al. (2018).

The reference values of ADDs are represented in previous studies (USEPA, 2001; Yin et al., 2018).

The hazard quotient (HQ) and hazard index (HI) were applied to evaluate the non-carcinogenic effect, while the carcinogenic risk (CR) index was adopted to assess the carcinogenic effect on human health (USEPA, 1986, 1989). The equations of HI, CR, and related information can be found in Xiao et al. (2015). $HI < 1$

suggests that the heavy metal has no non-carcinogenic risk to humans. $HI > 1$ indicates that non-carcinogenic risk to humans can be identified from heavy metals.

Spatial autocorrelation analysis

Moran's I typically can be used to indicate the degree of spatial autocorrelation, which includes global Moran's I and local Moran's I. Local spatial clustering patterns and spatial outliers can be identified based on the local Moran's I index, which measures the degree of correlation at each specific spatial location (Anselin, 1995). The high-high and low-low spatial clusters indicate that the high and low values are surrounded by their counterparts, respectively. Spatial outlier (high-low and low-high) is used to indicate that the target value is different from its surrounding values (Anselin, 1995). Detailed information about the calculation formula related to Moran's I can be found in published literature (Dong et al., 2021; Zhao et al., 2014).

Geostatistics, including semi-variogram and kriging, is generally employed to study the spatial pattern of environmental variables (Blanchet et al., 2017; Dai et al., 2018b; Fu et al., 2014; Goovaerts, 1999). In this study, ordinary kriging was adopted to interpolate the soil heavy metal values of the farmland in the study area.

Furthermore, disjunctive kriging is applied to assess the probability that the concentration of hazardous metals may exceed the threshold value (here the risk screening values in GB15618-2018), which can be calculated as:

$$\Omega[z(x_0) \geq Z_c] \approx P[z(x_0) \geq Z_c | z(x_1), z(x_2), \dots, z(x_N)]$$

where $z(x)$ is a random variable, which means the concentrations of single heavy metal; x represents the spatial coordinates in two dimensions, and z_c is the threshold values of heavy metals. If a threshold concentration z_c is defined, making the limit of what is acceptable, then the scale is dissected into two classes which is less and more than z_c , respectively. The values 0 and 1 can be assigned to two classes; a new binary variable, or indicator, is denoted by $\Omega[z(x) \geq z_c]$. At the sampling points, the values of z are known, and so the values 0 and 1 can be assigned with certainty. Elsewhere, one can at best estimate $\Omega[z(x_0) \geq z_c]$. In fact, it is necessary to do this in such

a way that the estimate at any place x_0 approximates the conditional probability, given the data that $z(x)$ equals or exceeds z_c (Zhao et al., 2020).

The nugget (C_0), sill ($C_0 + C$), and range were the three main factors to identify the spatial structure. The nugget-to-sill ratio was applied to reflect the extent of spatial dependence of soil variables (Guan et al., 2017; Yao et al., 2020). The ratio of <25% indicated the target variable is primarily controlled by soil type, topography, parent material, and other soil intrinsic factors, and >75% indicated the soil element is mostly affected by human activities and other extrinsic factors (Cambardella et al., 1994).

Softwares for data analysis

All statistical analyses were conducted by SPSS 19.0. The results of E_i and correlation analyses were mapped in R. Semi-variogram was analyzed by the software of GS+9.0. Moran’s I was performed by GeoDa. The spatial distributions of heavy metals were mapped with Arc GIS 10.7 software.

Due to the large variation in heavy metal concentration at the sampling points, when interpolating heavy metals in the study area, we used a manual classification method to classify the heavy metal concentrations (Fig. 7). The background value of soil heavy metal in Zhejiang Province and the soil pollution risk

screening value of agricultural land were introduced as the classification limit for the classification of pollution levels. The range of HM concentrations in the legend of Fig. 8 was automatically classified by the probability kriging method in Arc GIS 10.7.

Results and discussion

Heavy metal status in topsoil

The soil pH value in this study area varied from 3.71 to 8.55, with more than half of the pH values in the 5.5–6.5 range (Table 1). The organic matter (OM) values ranged from 17.3 to 57.49 g kg⁻¹. The mean concentrations of HN, AP, and AK were 154.39, 327.86, and 164.51 mg kg⁻¹, respectively. The mean concentration values of heavy metals were 1.25 (Cd), 35.44 (Ni), 77.68 (Cr), 77.38 (Pb), 122.14 (Cu), 203.39 (Zn), 0.21 (Hg), and 4.74 (As) mg kg⁻¹, respectively. Each Hg and As value was lower than the background values (BVs), indicating no contamination of Hg and As in soil. The average values of heavy metals, especially Cd and Cu, exceeded their BVs in Zhejiang Province by 9.62- and 3.99-fold, respectively, and exceeded the risk screening value (RSV) with 87.68% and 79.94% respectively, reflecting the soil in this region was severely contaminated

Table 1 Descriptive statistical analysis for the heavy metal concentrations in soils (mg kg⁻¹; OM: g kg⁻¹)

Metals	Range	Mean	SD	CV (%)	Skewness	Kurtosis	K-S _p	BV	RSV	Percent (%)
pH	3.71–8.55	6.13	0.80	13.05	0.090	0.295	0.629	-	-	-
OM	17.30–57.49	32.14	8.35	25.98	-0.019	-0.436	0.622	-	-	-
AN	28.00–345.50	154.39	62.32	40.37	1.018	0.933	0.001 (0.506)	-	-	-
AP	32.30–1536.64	327.86	335.56	102.35	2.135	5.218	0.000 (0.800)	-	-	-
AK	40.10–400.49	164.51	66.39	40.36	0.882	0.904	0.014 (0.875)	-	-	-
Cd	0.07–7.38	1.25	1.67	133.60	2.636	6.414	0.000 (0.114)	0.13	0.30	87.68
Ni	5.73–65.34	35.44	9.47	26.72	0.169	-0.269	0.922	36.48	70.00	0
Cr	9.00–152.99	77.68	22.35	28.77	0.588	2.022	0.017 (0.020)	95.91	150.00	2.87
Pb	19.70–278.02	77.38	40.32	52.11	2.099	7.808	0.000 (0.108)	30.46	90.00	20.34
Cu	13.20–608.43	122.14	105.78	86.61	1.833	4.216	0.000 (0.076)	30.54	50.00	79.94
Zn	51.10–875.54	203.39	128.72	63.29	2.151	8.975	0.000 (0.072)	107.79	200.00	32.09
Hg	0.04–1.00	0.21	0.12	57.14	2.884	11.926	0.000 (0.373)	0.75	1.80	0
As	0.37–9.35	4.74	1.44	30.38	0.480	0.480	0.250	17.10	40.00	0

SD, standard deviation; CV, coefficient of variation; K-S_p, Kolmogorov–Smirnov test for normality, the value in bracket were calculated after Box-Cox transformation; BV, background values of heavy metals for agricultural soil in Zhejiang province (CNEMC, 1990); RSV, risk screening values for soil contamination of agricultural land based on GB15618-2018 Soil environmental quality; percent: percentage of soil samples exceeded the RSV

mainly by these metals. The results were in line with some reports that Cd, Pb, Cu, and Zn were the main polluting heavy metals near the e-waste recycling area (Zhao et al., 2020). Cd, Cu, and Pb were the dominating pollutants in relation to e-waste recycling activities (Fu et al., 2013). According to He et al. (2017), Cd, Cu, and Zn were the main polluting heavy metals in soils around the e-waste dumping areas.

The coefficient of variation (CV) with a value < 20% means weak variation, which is controlled by natural sources. CV with a value > 50% indicates that the variation of the target variable is mainly influenced by anthropogenic activity (Zhao et al., 2019). The CVs of soil heavy metals were 133.60% (Cd), 52.11% (Pb), 86.61% (Cu), 63.29% (Zn), and 57.14% (Hg), indicating these five heavy metals had extensive variability. This result showed that the above-mentioned hazardous elements were mainly affected by anthropogenic activities (e.g., e-waste dismantling activities) (Han et al., 2018). Ni, Cr, and As had moderate variability with CVs of 26.72%, 28.77%, and 30.88%.

Pollution risk assessment

The P_i values varied greatly among the elements (Table 2). The mean P_i -BV values were ranked as follows: Cd > Cu > Pb > Zn > Ni > Cr > Hg, As. According to the maximum values of P_i -BV, some parts of the study area were heavily polluted by Cd, Cu, Zn, and Pb, while some areas were slightly polluted by Ni, Cr, and Hg. The P_i of As revealed that there was no contamination of As. The average P_i -RSV values of Cd and Cu were still within the level of serious pollution and the mean P_i -RSV values of

Fig. 2 The potential ecological risk index (E_i). Black point represents the mean distribution value. (The solid black line represents the 95% confidence interval, the width of the colored elements represents frequency, the black dot indicates the median, and the white square represents the four-point range)

Pb and Zn belonged to the level of heavy pollution, indicating that Cd and Cu accumulated at high levels in the soil in the study area, followed by Pb and Zn. Tang et al. (2014) also found that high levels of Cd and Cu were accumulated in soils of the e-waste region and their concentrations were 2.16 mg kg⁻¹ and 69.2 mg kg⁻¹, respectively.

The Nemerow multi-factor pollution indices based on background values and risk screening values in the soil around the study area are shown in Table 3. According to P -BV values, only one of 349 samples was non-polluted, while 99.71% of the soil samples were contaminated, indicating widespread accumulation of soil heavy metals. More than 50% of the soil samples were heavily polluted. More than 80% of samples were above the level of

Table 3 Nemerow multi-factor pollution index (%)

Standard	Unpolluted	Slightly polluted	Moderately polluted	Heavy polluted
BV	0.29	12.61	23.21	63.89
RCS	17.48	37.53	16.05	28.94

P -BV: The soil background value of Zhejiang province was used as the reference ratio to calculate the P

P -RSV: The risk screening values for soil contamination of agricultural land were used as the reference ratio to calculate the P

Table 2 Single-factor pollution index for soil heavy metals

P_i -BV: The soil background value of Zhejiang province was used as the reference ratio to calculate the P_i
 P_i -RSV: The risk screening values for soil contamination of agricultural land were used as the reference ratio to calculate the P_i

Metals	P_i -BV				P_i -RSV			
	Mean	Max	Min	Level	Mean	Max	Min	Level
Cd	9.65	57.21	0.56	Serious	4.15	24.60	0.24	Serious
Ni	0.97	1.79	0.16	Slight	0.51	0.93	0.08	Clean
Cr	0.81	1.60	0.09	Slight	0.52	1.02	0.06	Slight
Pb	2.54	9.13	0.65	Serious	0.86	3.09	0.22	Heavy
Cu	4.00	19.92	0.43	Serious	2.44	12.17	0.26	Serious
Zn	1.89	8.12	0.47	Serious	1.02	4.38	0.26	Heavy
Hg	0.28	1.33	0.05	Slight	0.12	0.56	0.02	Clean
As	0.28	0.55	0.02	Clean	0.12	0.23	0.01	Clean

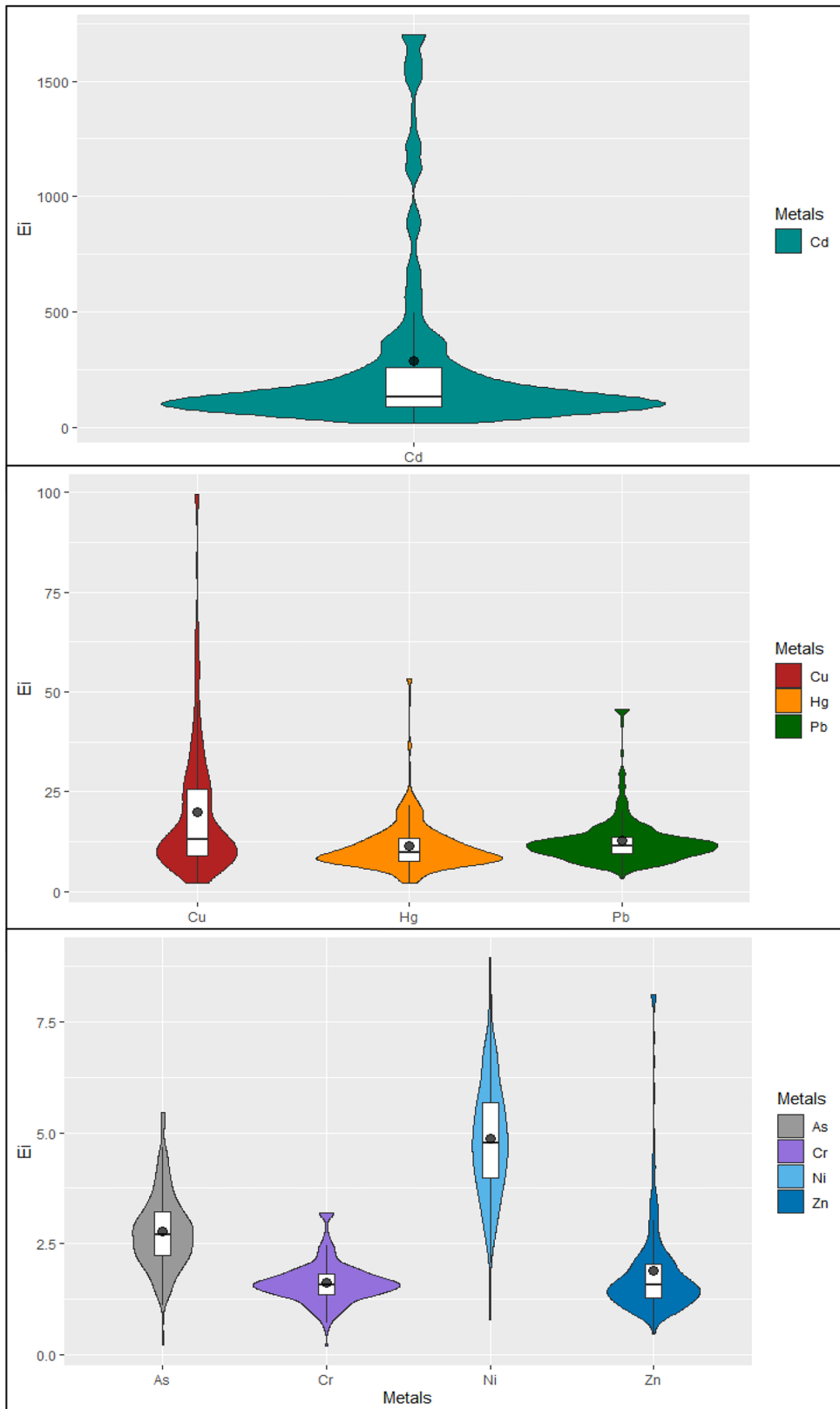
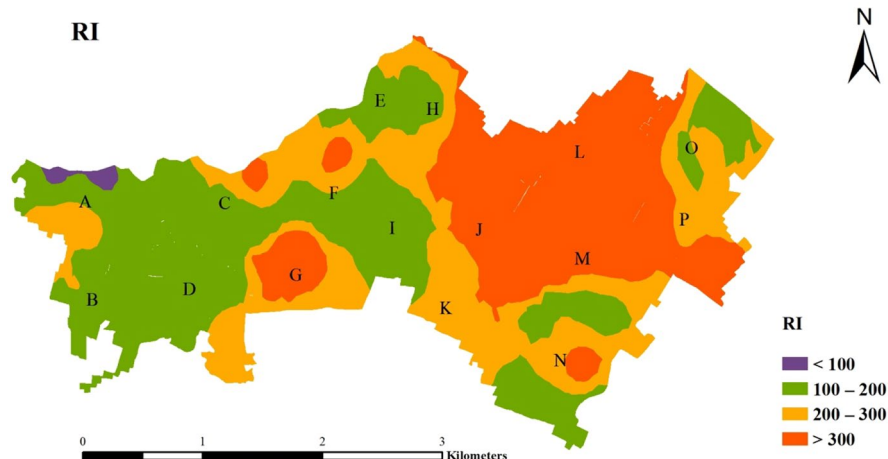


Fig. 3 The potential ecological risk index (The letters indicate the villages)



slightly polluted according to P -RSV values. Therefore, the potential ecological and human health risk should be further studied.

The results of E_i and RI are shown in Figs. 2 and 3. The mean E_i values of eight elements decreased in the order of $E_{Cd} > E_{Cu} > E_{Pb} > E_{Hg} > E_{Ni} > E_{As} > E_{Zn} > E_{Cr}$ (Fig. 2). Overall, the mean E_i values of all studied variables were below 40, except for Cd, which was up to 287.31, indicating that Cd was the crucial influence element on ecological risk (Jiang et al., 2014). The range of RI values was from 41.57 to 1925.13, with an average value of 342.53, showing the uneven distribution of potential ecological risk (Fig. 3). The RI values of almost 90% of sampling points (313 soil samples) were more than 100, suggesting that the ecological risk level ranged from moderate to high in most of the study region.

Human health risk assessment

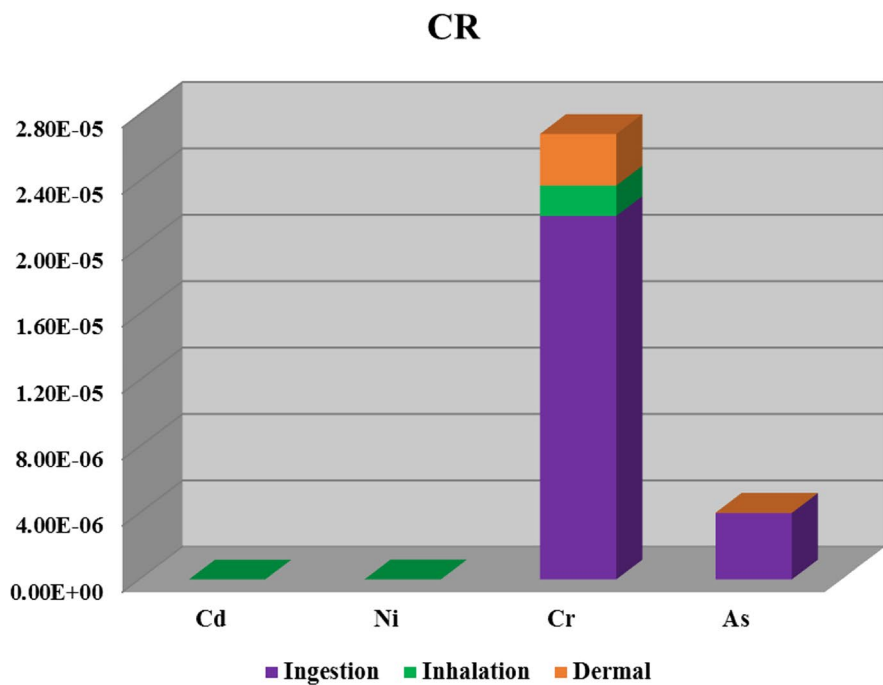
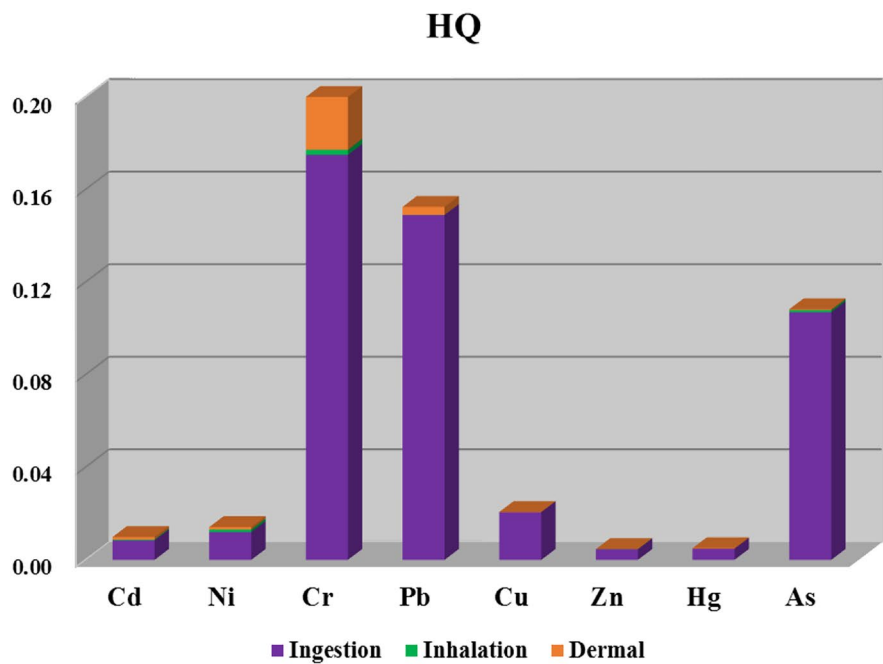
The non-carcinogenic risk for three exposure pathways to the same element increased in the following order: food intake > skin contact > inhalation for Cd, Cr, Pb, Cu, and Hg; food intake > inhalation > skin contact for Ni, Zn, and As (Fig. 4). In general, food intake of eight elements could cause serious damage to human health among the three main pathways (Yin et al., 2018). The HI value (0.517) indicated no non-carcinogenic risk to adults. The results of CR and CR_T values ($3.09E-05$) suggested that the carcinogenic risk was acceptable. Therefore, there was no health risk to local people. Even if contaminated soil with obvious potential ecological risk has no health hazard to human health,

people may be affected by heavy metals in agricultural products through food intake. Other researchers (Liu et al., 2013) reported that the intake of vegetables leads to an obvious risk to the residents and consumers, while the risk from soil creates relatively low hazards. The total carcinogenic risk value of heavy metals was within 10^{-6} – 10^{-4} , which indicated the acceptable carcinogenic risk, while the health risks caused by rice cannot be ignored (Yin et al., 2018).

Correlation analysis

The correlation analysis can effectively reveal the relationship between soil properties and heavy metals (Fig. 5). There was a significantly positive correlation between pH with Ni, As ($P < 0.01$), and Zn ($P < 0.05$). Alkaline soil can strengthen the sorption of Ni and Zn in soil solution, which weakens the transfer of Ni and Zn from soil to plant, increasing their total contents in soil (Temminghoff et al., 1995). Arsenic is usually found in soil as arsenate and arsenite, not as cations. In general, adsorption of arsenate decreases with increasing pH, whereas adsorption of arsenite strengthens with increasing pH (Zhong et al., 2020). Not all heavy metals are positively or negatively correlated with soil pH (Sungur et al., 2014). The OM, HN, and AP were significantly correlated with some heavy metals. The soil physical and chemical properties can be used to indicate the soil productivity and heavy metals are susceptible to them. They can affect the availability of heavy metals in soil and influence the obligate adsorption and surface coordination to affect the behavior and

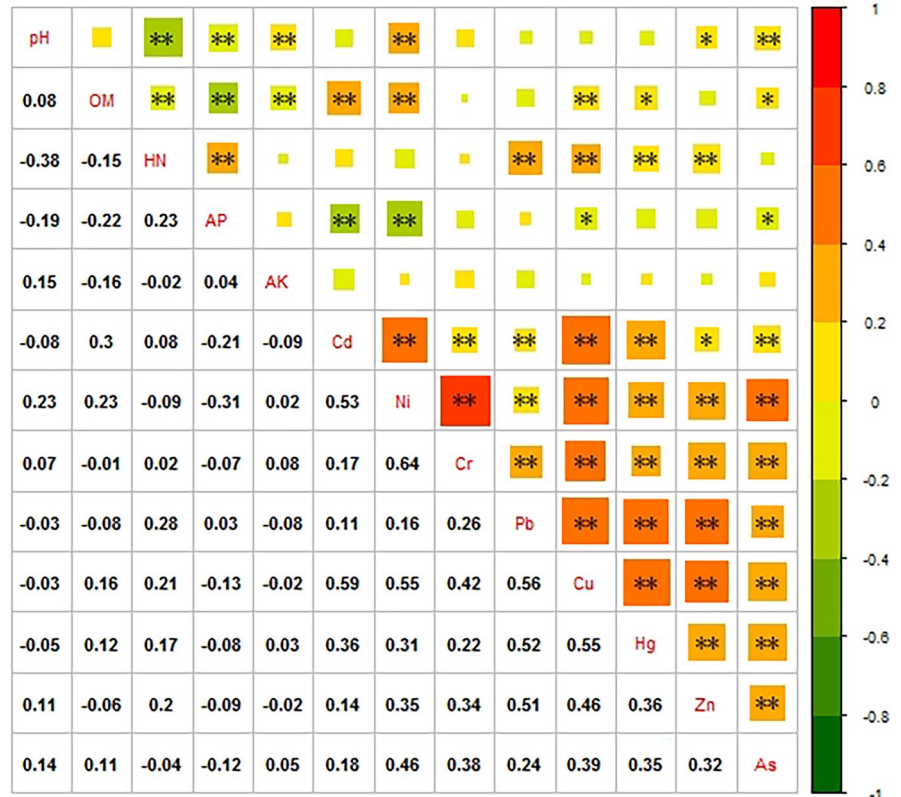
Fig. 4 Non-carcinogenic risk and carcinogenic risk of heavy metals for adults. (The values of the left bar indicate risk calculated by formulas to represent risk levels)



availability of heavy metals in soil (Covelo et al., 2007). It may be because our study area was agricultural land and long-term fertilization can affect the accumulation of heavy metals (Zhao et al., 2020).

All eight heavy metals considered were significantly positively correlated, suggesting that heavy metals could have a similar source, which was most likely e-waste in this study.

Fig. 5 Correlation analysis of soil heavy metal and physicochemical properties (double asterisks stand for significant correlation at $P < 0.01$; asterisk stands for significant correlation at $P < 0.05$). (The color scale on the right is for indicating the size of the correlation. Different confidence levels were applied to indicate the significance level at different significance levels. The size of the square indicates the strength of the correlation, the larger the correlation, the stronger the correlation, the smaller the weaker the correlation)



Spatial patterns of heavy metals in soil

Spatial clusters and outliers

The global Moran’s I values were 0.41 (Cd), 0.42 (Ni), 0.20 (Cr), 0.05 (Pb), 0.20 (Cu), 0.17 (Zn), 0.11 (Hg), and 0.37 (As) (Fig. 6), indicating that these elements had clear spatial patterns (Dai et al., 2018a). However, the spatial pattern of Pb was less obvious than other heavy metals. The high-high spatial clusters of Cd, Ni, and Cu were found in the northeast part of the study area and some obvious high-value spatial clusters of Ni, Cr, and Zn were observed in the southeast, while the low-value clusters of Cd, Ni, Cr, Cu, and Zn were mainly observed at southwest. The result of clusters of Pb was different from the other five heavy metals. The high-high clusters of Pb were mainly in the northwest part of the research area and low-value clusters were distributed in the northeastern and southwestern regions. The results (Fig. 6) indicated the high-high clusters were obviously correlated to unregulated e-waste dismantling sites and other polluting enterprises (Fig. 1), including e-waste

recycling, incineration, and burying. Some dismantling activities obviously affect the concentrations of soil heavy metals, which is similar to other reports (Han et al., 2017). The low-low spatial clusters are far away from the industrial activities and the areas were under traditional agricultural management.

Spatial structures

The semi-variograms can quantitatively indicate the spatial variability of heavy metals (Fang et al., 2016; Goovaerts, 1999). The best-fit model of Cd and Cr was the gaussian model, of Pb, Cu, and Zn was the exponential model, and of Ni was the spherical model (Table 4). The determination coefficients (R^2) of all heavy metals in soil were greater than 0.9, with the highest R^2 being 0.982 of Zn and the smallest R^2 being 0.915 of Pb, which indicated that all the soil variables had strong spatial structures.

In this study, the Cd and Cr had strong spatial dependence with the values of the nugget to sill ratios of Cd and Cr being 18.92 and 0.25, respectively. The family-operated recycling facility to deal with

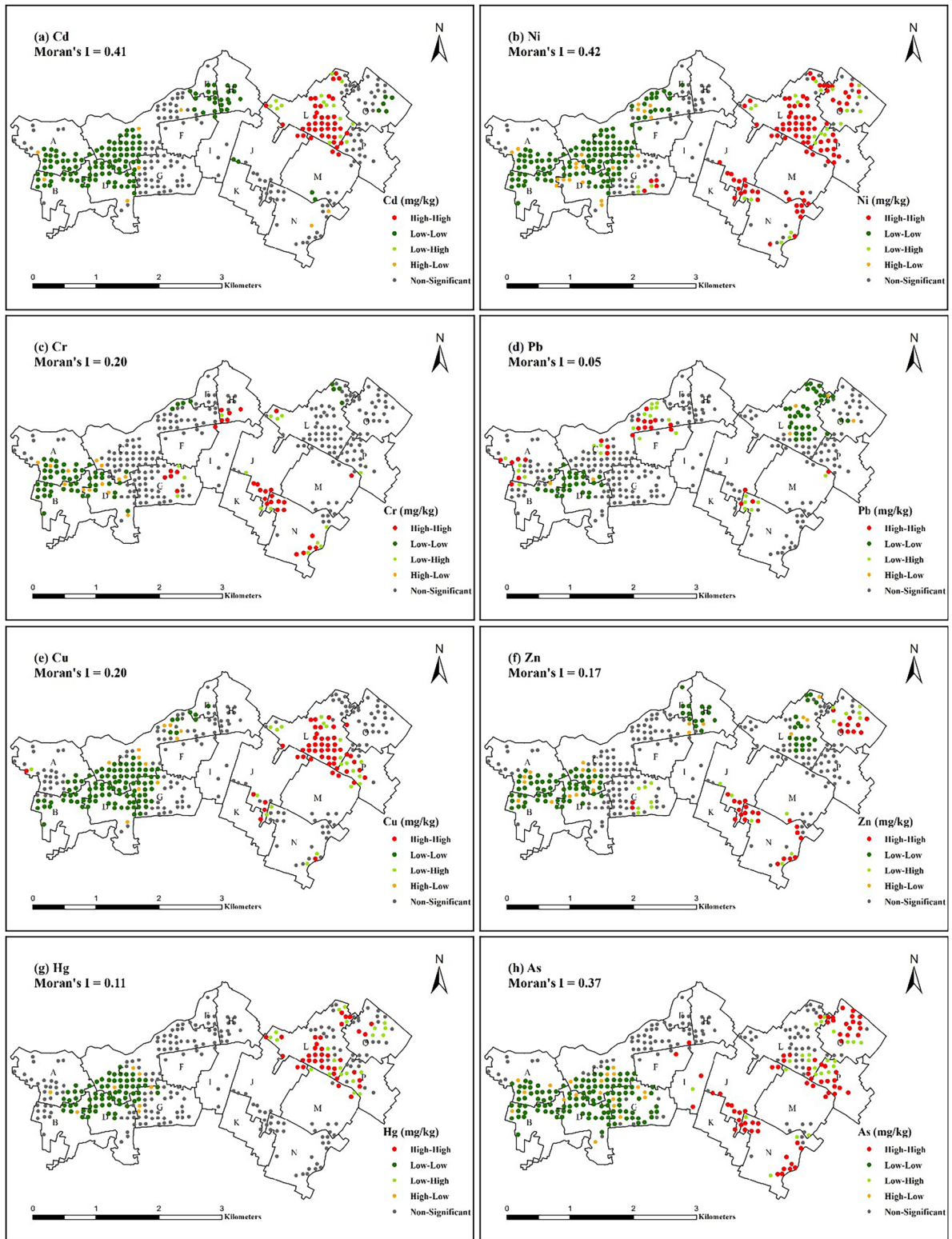


Fig. 6 Local Moran's I maps for soil heavy metals

Table 4 Theoretical semi-variance model and the corresponding parameters of soil heavy metals

Metal	Model	Nugget (C_0)	Sill ($C_0 + C$)	Nugget/sill (%)	Range (m)	R^2
Cd	Gaussian	1.22	6.449	18.92	1952	0.935
Ni	Spherical	44.3	118.62	47.35	5913	0.932
Cr	Gaussian	1	404.2	0.25	147	0.961
Pb	Exponential	1006	2380	42.27	5280	0.915
Cu	Exponential	5750	19,570	29.38	9078	0.929
Zn	Exponential	10,150	24,320	41.74	7542	0.982
Hg	Exponential	0.00001	0.01292	0.08	165	0.900
As	Spherical	1.026	2.185	46.96	2994	0.990

R^2 , determination coefficient

e-wastes in the study area was banned in the 1990s. Heavy metal pollution in the study area has existed for more than 30 years. The development of the parent rock is regarded as an internal factor and changes with other factors, so the ratio nugget/sill of Cr, Hg, Cd, and Cu is lower (<50%). The other soil heavy metals had moderate dependence (Table 4), indicating that target variables were affected by extrinsic and intrinsic factors together. The e-waste dismantling activities probably were the main extrinsic factor in changing the spatial structure of heavy metals. Soil properties could be the main intrinsic factor according to the correlation analysis shown in Fig. 5.

Spatial distribution of heavy metals

The spatial distribution trends of Cd and Cu were similar with the high concentration values clustering near villages of L area, while the low concentration values were patchy in the periphery of the study area (Fig. 7). The Cd and Cu values were gradually decreasing from the village of L area to the periphery of the area, indicating that Cd and Cu may have same sources: e-waste dismantling activities, such as refining metals from wire, pyrolyzing circuit board, and using acids to extract gold and other metals (Han et al., 2018; Tang et al., 2014). The Ni and Cr values decreased from the southeast part to the northwest except for a small part of higher Ni and Cr values which were found in A area. The high Pb values were mainly concentrated in the northern and eastern regions of the study area. Most of the high concentrations of soil Zn were found in the southeast part. Meanwhile, the spatial distribution of Pb and Zn displayed that the low Pb and Zn

values were found in the west and north parts of the study region, indicating that Pb and Zn had similar spatial distribution.

Almost all concentrations of all heavy metals around the e-waste dismantling sites were very high. However, the spatial distribution differences may be caused by different electronic dismantling activities and different kinds of e-wastes. Previous studies concluded that heavy metal pollution was primarily impacted by unregulated dismantling activities (Han et al., 2018; He et al., 2017). The primitive processes (e.g., strong acid leaching and stripping) could lead a significant quantity of heavy metals to be released into the soil (Zhao et al., 2019). Burning is usually associated with higher heavy metal contamination, and opening dumping is a typical method of heavy metal contamination in the area.

Disjunctive kriging to estimate the probability of excessive concentrations of Cd and Cu is shown in Fig. 8. If the probability exceeds 60%, the area is at considerable or high risk (Qishlaqi et al., 2009). The probability of Cd concentration exceeding the threshold was generally greater than 0.6, especially in C, F, and G areas in the west and L and J areas in the east with the probability of 0.94 to 1. Many lower probability patches were distributed in the western and middle parts. The areas with high risk of Cu were mainly distributed in the eastern regions (risk probability over 0.87); low-risk areas were mainly distributed in B and D areas. In general, high-risk areas of the two elements mentioned above were widely distributed in the study region, which was identical to the risk evaluation results. Therefore, high-risk areas should be paid great attention to ensure safety and improve the quality of local produce.

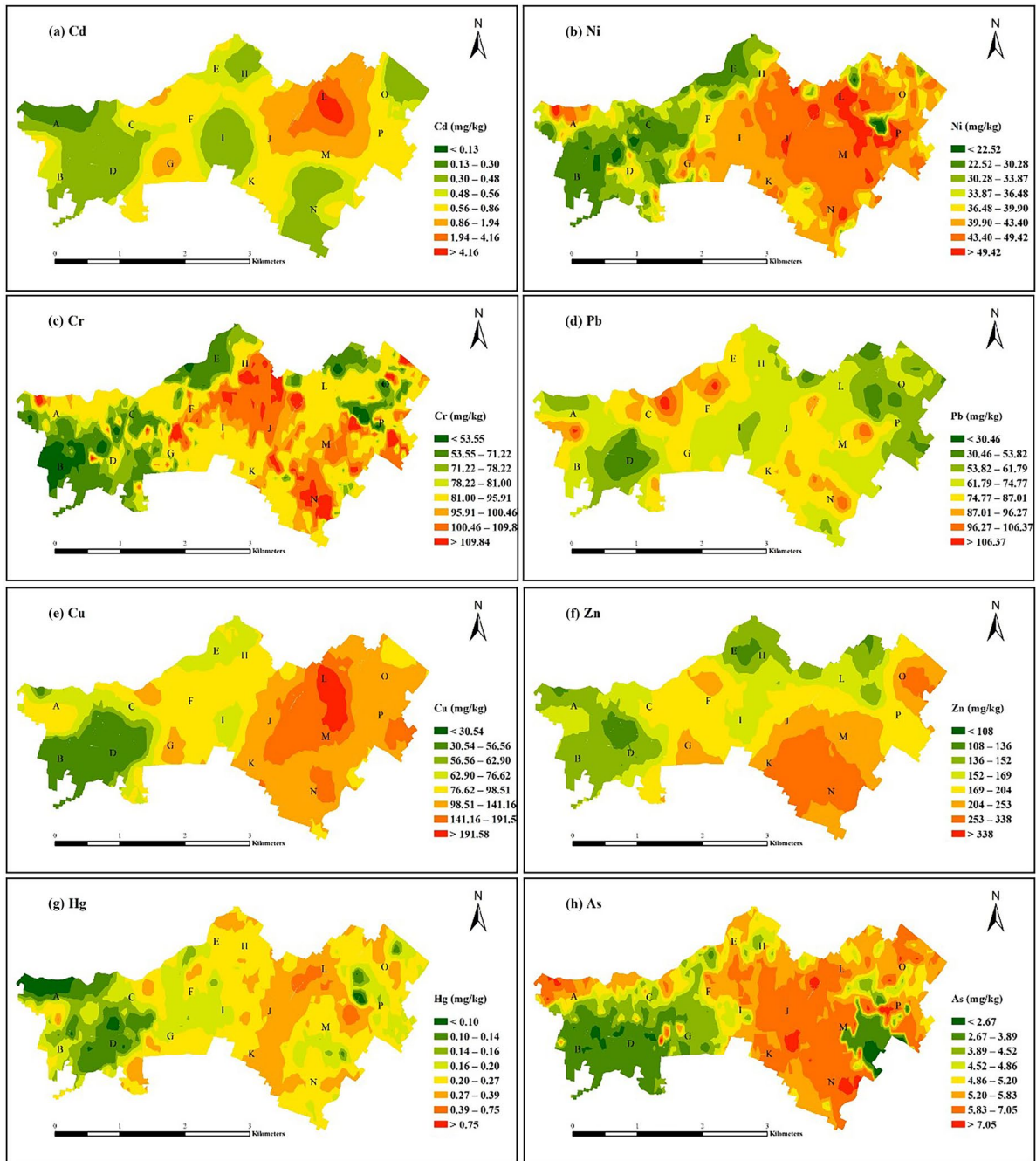
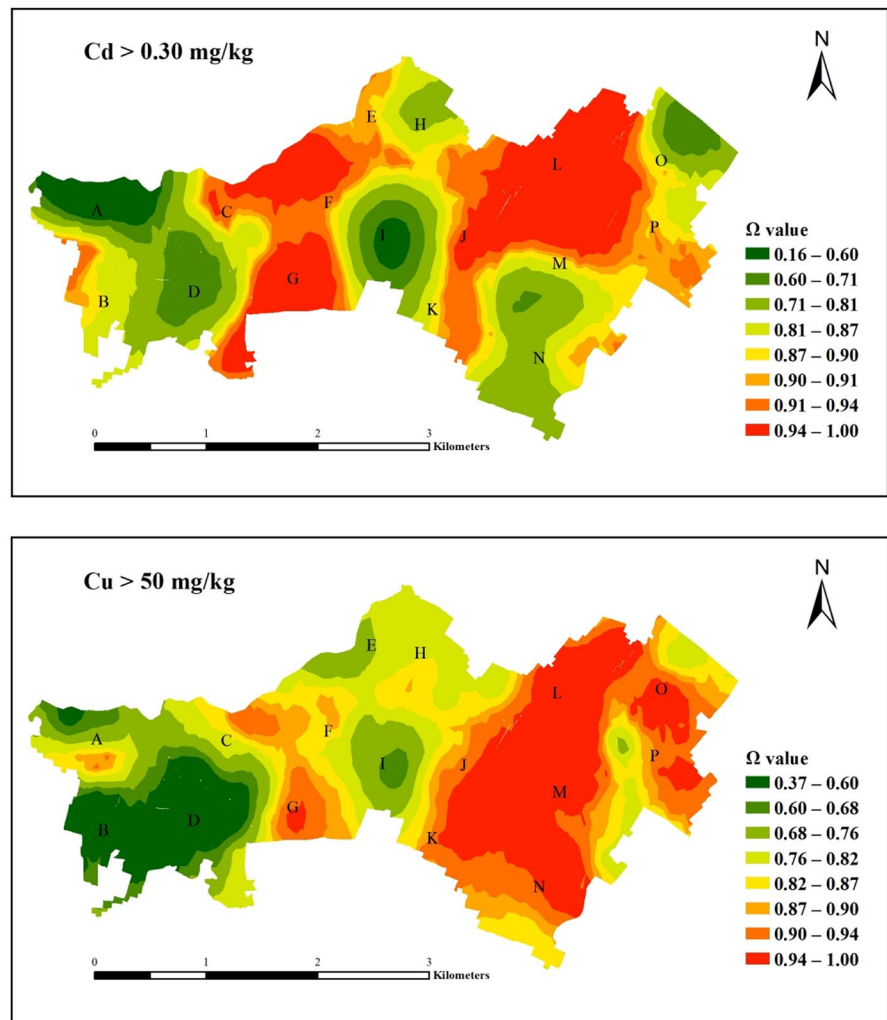


Fig. 7 Spatial distribution maps of the concentration of soil heavy metals

Fig. 8 The estimated probability of the concentration of soil Cd and Cu



Conclusions and implications

The Cd, Pb, Cu, and Zn were accumulated in soils compared with the BVs and the RSVs. The concentration of soil heavy metals above the risk screening value means that agricultural products may not meet quality and safety standards, and heavy metals can accumulate in the human body by eating edible agricultural products. Therefore, some safety measures such as alternative planting (e.g., low-accumulating vegetables) may be useful to reduce the damage of heavy metals to humans. Meanwhile, Cd and Cu severely contaminated farmland soils in the study area. The heavy metal ecological risk in the study area varied from moderate to high and Cd was the major factor causing the risk, as shown by E_i and RI . High

concentrations of heavy metals were mainly distributed close to e-waste dismantling areas.

For agricultural lands, it is essential to adopt efficient measures to improve the soil quality and remove the pollutants such as toxic metals. There are many family-operated e-waste dismantling sites in the study area. Although our study area is small, the sources and characteristics of regional heavy metal pollution are similar. This study is also representative. Therefore, our study of local pollution can also be extended to regional heavy metal pollution remediation. Therefore, it is urgent to describe the spatial distribution and identify sources of heavy metal contamination at the regional scale. E-waste pollution is not only a problem in China, but also a global problem for other countries such as India and Thailand, and it is a research hotspot

in the environmental field. Therefore, for agricultural lands around the e-waste dumping sites and dismantling fields, risk-based environmental management measures should be implemented to mitigate the soil heavy metal contamination and to further eliminate human health risk caused by soil heavy metal contamination (For example, for heavily-polluted areas, remediation measures such as planting hyperaccumulator (*Sedum alfredii* Hance) and converting the land use from farmlands to forestry-lands are necessary; for the slight-polluted areas, planting low-accumulating vegetables such as solanaceous is recommended).

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Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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