Research Article

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# Spatial distribution and risk assessment of toxic metals in agricultural soils from endemic nasopharyngeal carcinoma region in South China

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**Abstract:** Sihui city (South China) is much affected by nasopharyngeal carcinoma (NPC). To investigate the relationships between the toxic metals in soil and NPC incidence in Sihui, 119 surface soil samples were collected from agricultural fields and analyzed. The soil As–Cr contents in Longjiang (high-incidence area) are significantly lower than those in Weizheng and Jianglin (low-incidence areas), whereas the soil Pb content in Longjiang is significantly higher than that in Weizheng. The Nemerow pollution indices ($PI_N$) of soils decrease in the order of Jianglin > Weizheng > Longjiang. The enrichment factor (EF) of Cd indicates that the Cd enrichment is contributed by human activities. Potential toxic metal-related ecological risk values decrease in the order of Jianglin > Weizheng > Longjiang. The mean hazard index (HI) value of Longjiang was lower than those of Weizheng and Jianglin. There are no adverse noncarcinogenic health effects of soil toxic metals to adults in the study areas. Carcinogenic risks of As and Cr via ingestion and dermal contact and total carcinogenic risk are within the warning range, from $10^{-6}$ to $10^{-4}$. Hence, we suggest that toxic metals in the soil may not be major geochemical carcinogenic factors of high NPC incidence in Sihui.

**Keywords:** toxic metals, nasopharyngeal carcinoma, risk assessment, Pearl River Delta

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## 1 Introduction

Toxic metals in soil usually possess significant toxicity because of their persistence in soil, and then affect human health through the food chain. Thus, diet is the most significant pathway of toxic metals exposure in humans. Moreover, other exposure routes of toxic metal in humans include absorption through skin, respiratory intake, and accidental ingestion of heavy metal-contaminated substances [1–3]. Toxic metal can interfere with the oxidation–reduction potential and disrupt the reactions in living cell. Notably, the toxic metals in the soil pose a notable risk for skin, skeletal, immune, cardiovascular, digestive, and other systems, which may ultimately result in cancer according to the previous studies [2,4–8]. The exposure of Pb in topsoil is significantly associated with gastric cancer in Anhui province, China [9]. The toxic metals are thought be linked to the high gastrointestinal cancer incidence in Van province of Turkey [10]. The joint effects of Cr–Cd, Cr–As, Cd–As, and Cr–Pb have significant impact on the distribution of stomach cancer in Hangzhou, China [11]. Sb and Sr contents in soil are much higher in area with high esophageal cancer in Golestan province, Iran [12].

Nasopharyngeal carcinoma (NPC) is a rare malignancy in most areas of the world, whereas it has a high incidence rate in certain parts of Asia, especially in China [13,14]. Roughly 65,000 new cases of NPC are reported annually worldwide, and over 80% come from the South China and Southeast Asia. In particular, the Guangdong province in South China has the highest NPC incidence (more than 20 per 1,00,000 in male annually). Other provinces in South China, such as Fujian, Guangxi, Jiangxi, and Hunan, have also reported high NPC incidence, whereas incidence in northern China is low (1–5 per 1,00,000 annually) [15]. Epidemiological studies of NPC revealed that genetic susceptibility, Epstein–Barr virus (EBV) infection, and different environmental impacts are likely the major exposure factors of NPC in South China [16,17]. Most epidemiological studies of NPC focus on etiological factors including EBV, genetic susceptibility, and consumption of food with nitrosamines, but there are few attentions on the obviously geographical heterogeneity of NPC incidence. In fact, the pathogenic mechanism of heavy metals on NPC has not been fully understood [18]. Limited studies reported that nickel (Ni) in drinking water had a significant positive correlation with nasopharyngeal cancer mortality rate in China, and arsenic and cadmium (Cd) levels in the blood were associated with the risk of nasopharyngeal cancer in Tunisian population [19]. Ecologic studies showed trace elements in soils are related to NPC incidence or mortality [19–23]. A map-based ecologic study in China presented a geographic correlation between NPC mortality and low soil levels of the alkaline elements magnesium, calcium, and strontium as well as high soil levels of radioactive thorium and uranium [21,22]. Notably, these findings need to be further confirmed by the analytic epidemiologic studies. Sihui, an NPC endemic region in Guangdong, South China, has the highest incidence of NPC in China. A cancer registry including NPC was established in Sihui to report the incidence and mortality from the 1970s [13,24]. According to the secular trend analysis of NPC incidence in Sihui, males have a higher incidence of NPC than females, with a sex ratio of 2.16:1. NPC has the higher incidence in those around 35–60 years of age in Sihui; this phenomenon has not been well understood.

To make a better understanding of the relationship with toxic metals (Cd, Cr, Cu, Ni, Zn, Pb, As, and Hg) and NPC in Sihui city, 119 surface soil samples were collected from one high-incidence area and two low-incidence areas. This study aims to (1) compare toxic metal contents including Cd, Cr, Cu, Ni, Zn, Pb, As, and Hg and evaluate the soil pollution levels between high and low NPC incidence areas; (2) identify the potential sources of the toxic metals using multivariate analysis such as correlation matrix, cluster analysis, and principal component analysis (PCA); (3) assess the potential ecological risk and health risk of toxic metal pollution in soil; and (4) provide insights for policymakers with respect to pollution and NPC prevention measures.

# 2 Materials and methods

## 2.1 Study area and sampling

The study areas including Longjiang, Weizheng, and Jianglin are located in Sihui city, Guangdong (South China), in the northwestern part of the Pearl River Delta (23°11′40″ ~ 23°41′42″N and 112°25′25″ ~ 112°51′35″E), covering an area of 1,258 $km^2$ (Figure 1). The area is influenced by subtropical climate. Sihui is a county-level city. Sihui had a population of 4,18,097 at the end of 2011 and over 75% of the inhabitants live in the rural area. Rice paddies and oranges are the main economic crops in Sihui [25]. Longjiang district is in the central and southern part of the study area. This area is in hilly region located along the Longjiang River. Weizheng and Jianglin districts are located in the northern and western part of Sihui, respectively, and both are of mountainous topography. Sihui has the highest NPC morbidity (19.23 per 100,000 each year) and mortality (14.43 per 1,00,000 each year) in China [26]. However, the morbidity of NPC in Sihui is not uniform. The mortality of NPC in Longjiang district (23.48 per 1,00,000 each year) is significantly higher than the peripheral regions, such as Weizheng district (9.83 per 1,00,000 each year) and Jianglin district (5 per 1,00,000 each year).

In order to compare the levels of toxic metal pollution and health risk in agricultural soil of Sihui, 94 samples were collected from the high-incidence area of NPC (HC), Longjiang district, including 7 towns along the Longjiang river. In the low-incidence areas of NPC (LC), 17 samples were collected from Weizheng and 8 samples were from Jianglin. The surface soil samples were collected at an interval of about 200 m according to the topographical characteristics and avoiding obvious pollution sources. Soil samples were collected from agricultural land including paddy field, vegetable land, and orchard. Five subsamples were collected within 30 m of the sampling point to form a composite sample (about 1 kg). The spatial distribution of sample points should be as uniform as possible. All the surface soil samples were collected at 0–20 cm depth. The samples were first dried in an oven at 60°C and then sieved for 2 mm using a nylon sieve to remove larger particles and plant debris. The samples were then grinded using an agate ball-grinder and further sieved using a 74 μm nylon sieve and powdered. These soil samples were used to measure the pH, soil organic matter, and toxic metal concentrations.

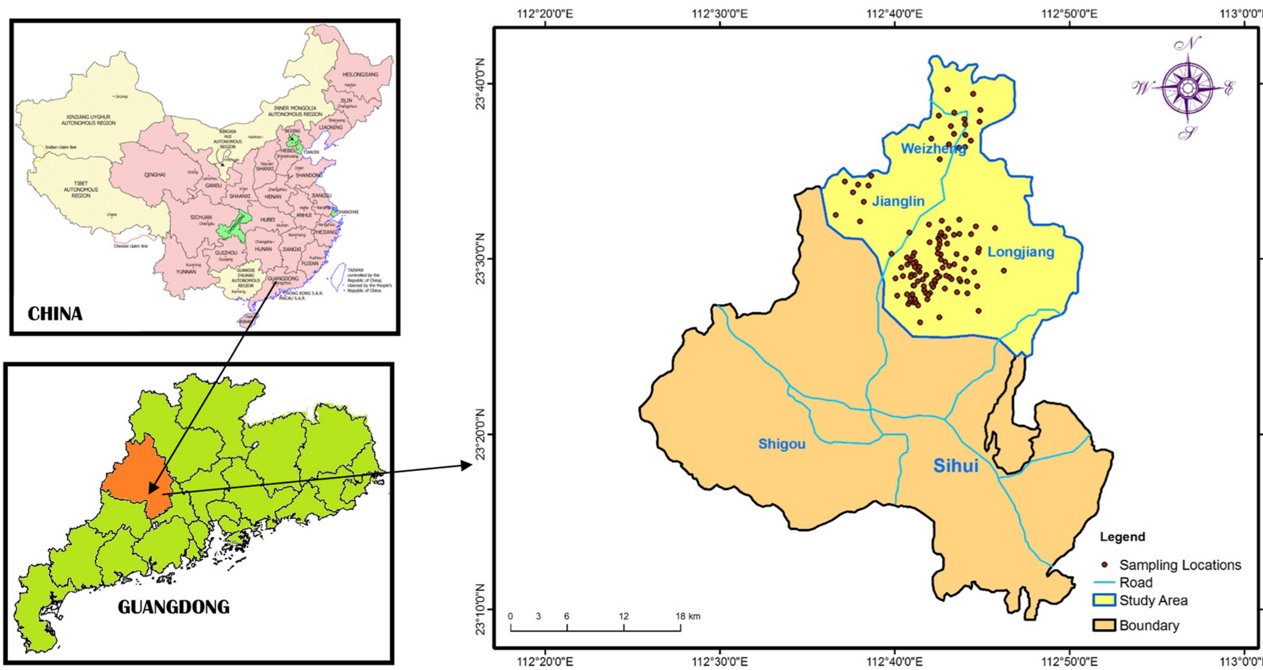


**Figure 1:** Locations of study area and sample locations in Sihui city of Guangdong, China.

## 2.2 Chemical analysis

For the toxic metal concentration measurement, 0.5 g of each soil sample was digested with a concentrated mixture of $HCl–HNO_3–HF$ using a microwave digestion system (MARS, CEM, USA) according to EPA method 3052 [27]. After evaporating the digested solution to near dryness, the residuals were redissolved to 5 ml with 1 N $HNO_3$. Subsequently, the concentrations of most elements (e.g., Cd, Cr, Cu, Ni, Zn, Pb, and Mn) in the samples were analyzed by inductively coupled plasma-optical emission spectrometry (ICP-OES; Optima 2000 DV, Perkin Elmer, USA), and the As–Hg contents were analyzed by atomic fluorescence spectrometry (AFS230, China). The $Al_2O_3$ and $Fe_2O_3$ contents were analyzed by X-ray fluorescence spectrometer (XRF; ZSX Primus, Rigaku, Japan). Soil pH value was measured with a pH meter (HACH, USA) in the soil suspension (water:soil = 2.5:1). The organic matter (OM) content was measured by oxidation of potassium dichromate [28]. The reagents and chemicals were of analytical grade. The accuracy and precision were controlled by using China geochemical reference materials (GSS-1, GSS-2, GSS-3, and GSS-8). The recoveries of target toxic metals in the standard references are of 85–119%. The relative standard deviations (RSDs) were less than 5% for all target toxic metals.

## 2.3 Toxic metal pollution assessment

The level of metal pollution was assessed by pollution indices of toxic metals ($i$), which was evaluated with the single pollution index ($PI_i$):

$$PI_i = C_i/S_i \tag{1}$$

where $C_i$ is the toxic metal elements concentration (mg kg$^{-1}$), and $S_i$ is its reference value (mg kg$^{-1}$). The values for toxic metal elements were based on the standards for Soil Environmental Quality of China (GB15618-2018). The risk control standard of heavy metals for soil contamination of agricultural land is listed in Table S1. The overall pollution level of toxic metals in the soils was assessed by the Nemerow pollution index ($PI_N$) using the following formula [29,30]:

$$PI_N = \sqrt{[(C_i/S_i)^2_{\max} + (C_i/S_i)^2_{\text{ave}}]/2} \tag{2}$$

where $PI_N$ is overall pollution index, and $(C_i/S_i)_{\max}$ and $(C_i/S_i)_{\text{ave}}$ are maximum and mean pollution indices for individual toxic metal, respectively. Based on the corresponding $PI_N$ values, surface soil pollution is classified into five levels, i.e., clean ($PI_N \le 0.7$), warning limit ($0.7 < PI_N \le 1$), slight polluted ($1 < PI_N \le 2$), moderately polluted ($2 < PI_N \le 3$), and seriously polluted ($PI_N > 3$).

Anthropogenic influence on toxic metal accumulation in the surface soils was evaluated by the enrichment factor (EF), which was calculated to identify the anthropogenic and natural contributions, using the formula below [31]:

$$\mathrm{EF} = \left[\frac{(C_i/C_n)_{\mathrm{sample}}}{(C_i/C_n)_{\mathrm{background}}}\right] \tag{3}$$

where $C_i$ is the target metal concentrations (mg kg$^{-1}$), $C_n$ is the concentration of the reference metal (mg kg$^{-1}$), and $(C_i/C_n)_{\mathrm{sample}}$ and $(C_i/C_n)_{\mathrm{background}}$ are the ratios of the target metal and the reference metal in the determined soil and background soil. The natural toxic metal background of soils in Guangdong was referenced from the National Environmental Monitoring Centre (1990) (Table 1). EF values of all toxic metals (except Pb) measured in this study were calculated using $Fe_2O_3$ as a reference for normalization, and $Al_2O_3$ as a reference for Pb [32,33]. The average soil background $Fe_2O_3$ and $Al_2O_3$ contents in Guangdong are 3.46% and 13.62%, respectively. Based on their EF values, the toxic metals of the soils were classified into six categories, i.e., no enrichment (0–1), minimal enrichment (1–2), moderate enrichment (2–5), significant enrichment (5–20), very high enrichment (20–40), and extremely high enrichment (>40) [31].

Potential ecological risk posed by toxic metal pollutants was quantitatively assessed using the ecological risk index (RI) [34,35], which was calculated using the sum of risk factors of the toxic metals:

$$\mathrm{RI} = \sum_{i=1}^{n} E_{\mathrm{f}}^{i} \tag{4}$$

$$E_{\mathrm{f}}^{i} = T_i \times \frac{C_i}{C_0} \tag{5}$$

where $E_{\mathrm{f}}^{i}$ is the single risk factor for toxic metal $i$. The toxic-response factor $T_i$ for toxic metal $i$ was determined for Hg = 40, Cd = 30, As = 10, Ni = Cu = Pb = 5, Cr = 2, and Zn = 1 [34,36–38]. $C_i$ is the concentration of metal $i$ (mg kg$^{-1}$), and $C_0$ is the background concentration (mg kg$^{-1}$). Five ecological risk levels of the toxic metals in the soils were classified based on the calculated $E_{\mathrm{f}}^{i}$ and RI (Table S2).

**Table 1:** Descriptive statistics of physicochemical properties and toxic metal concentrations of agricultural soils in the study area (mg/kg). (HC and LC = high and low incidence area of NPC)

| Region | | pH | OM (%) | As | Cd | Cr | Cu | Hg | Ni | Zn | Pb |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | Background[1] | 5.2 | 2.93 | 8.90 | 0.056 | 50.5 | 17.0 | 0.078 | 14.4 | 47.3 | 36.0 |
| Weizheng (LC, $n$ = 17) | Mean[2] | 5.87 | 1.25 | $13.8^{a}$ | $0.15^{a}$ | $63.4^{a}$ | $20.3^{a}$ | $0.14^{a}$ | $16.2^{a}$ | $53.1^{a}$ | $26.1^{a}$ |
| | Min | 3.35 | 0.66 | 5.25 | 0.06 | 31.6 | 10.3 | 0.04 | 8.91 | 28.0 | 14.5 |
| | Max | 7.87 | 1.79 | 39.1 | 0.38 | 125 | 42.5 | 0.35 | 29.6 | 123 | 71.3 |
| | SD | 1.19 | 0.38 | 8.40 | 0.08 | 20.8 | 8.01 | 0.08 | 4.63 | 21.2 | 13.1 |
| | CV (%) | 20.7 | 32.2 | 70.2 | 54.7 | 34.3 | 41.9 | 69.3 | 29.6 | 42.4 | 54.0 |
| | Percentage of exceeding the background (%) | | | 64.7 | 100 | 70.6 | 70.6 | 88.2 | 52.9 | 58.8 | 11.8 |
| Jianglin (LC, $n$ = 8) | Mean[2] | 5.39 | 1.80 | $20.2^{a}$ | $0.12^{a}$ | $75.4^{a}$ | $21.5^{a}$ | $0.15^{a}$ | $20.8^{a}$ | $61.2^{a}$ | $28.1^{a,b}$ |
| | Min | 5.07 | 0.74 | 6.42 | 0.03 | 44.8 | 11.9 | 0.03 | 11.3 | 25.7 | 17.6 |
| | Max | 5.91 | 3.54 | 83.1 | 0.27 | 155 | 62.3 | 0.30 | 61.7 | 163 | 34.6 |
| | SD | 0.31 | 1.21 | 25.7 | 0.09 | 35.1 | 16.8 | 0.08 | 16.7 | 43.1 | 5.31 |
| | CV (%) | 5.8 | 67.0 | 127 | 72.0 | 46.5 | 78.2 | 54.7 | 80.4 | 70.3 | 18.9 |
| | Percentage of exceeding the background (%) | | | 62.5 | 75.0 | 75.0 | 37.5 | 87.5 | 62.5 | 37.5 | 0.0 |
| Longjiang (HC, $n$ = 94) | Mean[2] | 5.77 | 1.15 | $9.11^{b}$ | $0.14^{a}$ | $42.2^{b}$ | $15.1^{a}$ | $0.12^{a}$ | $13.3^{a}$ | $53.7^{a}$ | $33.5^{b}$ |
| | Min | 4.22 | 0.12 | 3.31 | 0.03 | 16.3 | 3.75 | 0.01 | 4.81 | 28.8 | 13.1 |
| | Max | 8.83 | 1.84 | 24.6 | 1.77 | 238 | 160 | 0.45 | 104 | 283 | 66.5 |
| | SD | 0.74 | 0.44 | 4.46 | 0.19 | 28.0 | 16.0 | 0.07 | 10.8 | 27.5 | 11.5 |
| | CV (%) | 12.8 | 38.8 | 48.9 | 137 | 66.4 | 106 | 58.8 | 81.3 | 51.3 | 34.3 |
| | Percentage of exceeding the background (%) | | | 46.8 | 88.3 | 26.6 | 26.6 | 69.1 | 29.8 | 52.1 | 39.4 |
| Kurtosis | | 1.95 | 6.46 | 43.32 | 71.07 | 16.04 | 67.87 | 3.57 | 43.74 | 40.30 | 0.55 |
| Skewness | | 0.65 | 1.42 | 5.53 | 7.69 | 3.17 | 7.45 | 1.55 | 5.76 | 5.44 | 0.82 |
| K–S p | | 0.32 | 0.56 | 0.00 | 0.00 | 0.01 | 0.00 | 0.03 | 0.00 | 0.00 | 0.28 |

[1]Background values of soils Guangdong Province (CNEMC 1990). [2]Mean values within the same column with the same letters are with no significant differences ($p$ > 0.05).

## 2.4 Health risk assessment of toxic metals

The health risks assessment model recommended by United States Environmental Protection Agency (USEPA) was used in this study to quantify both noncarcinogenic effects and carcinogenic effects of toxic metals in soils to humans via ingestion, air inhalation, and dermal contact [39]. Considering NPC had rather low incidence in those of age below 30 in Sihui, only health risks to adults were discussed in this study. The average daily exposure dose of a toxic metal via ingestion, air inhalation, and dermal contact could be calculated [40–43].

$$\mathrm{ADD_{ing}} = \frac{\mathrm{C_s \times IngR \times CF \times EF \times ED}}{\mathrm{BW \times AT}} \tag{6}$$

$$\mathrm{ADD_{ing}} = \frac{\mathrm{C_s \times InhR \times EF \times ED}}{\mathrm{PEF \times BW \times AT}} \tag{7}$$

$$\mathrm{ADD_{derm}} = \frac{\mathrm{C_s \times AF \times CF \times SA \times ABS \times EF \times ED}}{\mathrm{BW \times AT}} \tag{8}$$

where ADD is the average daily exposure dose (mg $\mathrm{kg^{-1}\,day^{-1}}$); $C_s$ is the concentration of toxic metal in soil (mg $\mathrm{kg^{-1}}$). Exposure parameters used to calculate the average daily exposure dose and risk are shown in Table S3 [40–43].

The noncarcinogenic risk of exposure to individual toxic metals was assessed by calculating the hazard quotient (HQ). Total noncarcinogenic hazard index (HI) is the sum of HQ of each element. The carcinogenic risk of exposure was assessed by lifelong carcinogenic risk (CR) values. $\mathrm{CR_T}$ is the total carcinogenic risk.

$$\mathrm{HQ} = \frac{\mathrm{ADD}}{\mathrm{RfD}} \tag{9}$$

$$\mathrm{HI} = \Sigma \mathrm{HQ}_i \tag{10}$$

$$\mathrm{CR}_i = \mathrm{ADD} \times \mathrm{SF} \tag{11}$$

$$\mathrm{CR}_T = \Sigma \mathrm{CR}_i \tag{12}$$

where RfD is the reference dose of noncarcinogenic risk for the chemical (mg $\mathrm{kg^{-1}\,day^{-1}}$). SF is the carcinogenicity slope factor (kg day $\mathrm{mg^{-1}}$). Reference values of RFD and SF are listed in Table S4 [41,44,45]. The evaluation standards of HQ and CR are listed in Tables S5 and S6 [2,46,47]. When the HI value is <1, noncarcinogenic risk is accepted and the degree of risk increases as HI increases. When the HI value is between 1 and 10, the adverse health effects are possible. When the HI value is >10, high chronic risk may appear. Generally, when the CR value is $<10^{-6}$, the health risk caused by carcinogens from the soil can be considered as negligible. When CR > $10^{-4}$, significant health risk will be posed to human being. The acceptable range of CR value is $10^{-4}$–$10^{-6}$.

# 3 Results and discussion

## 3.1 Concentrations of toxic metals

Descriptive statistics of pH, OM content, and toxic metals concentrations of soils in three studied districts are listed in Table 1. The K–S test indicated that pH, OM, and toxic metals contents are log-normally distributed except for Pb, which is normally distributed. The results show that the soil pH in Weizheng, Jianglin, and Longjiang is 3.35–7.87 (avg. 5.87), 5.07–5.91 (avg. 5.39), and 4.22–8.83 (avg. 5.77), respectively, indicating that the soils in the study areas are on average acidic. Soil acidification may contribute to increased leaching of toxic metals. The soil OM contents in Weizheng, Jianglin, and Longjiang are 0.66–1.79% (avg. 1.25%), 0.74–3.54% (avg. 1.80%), and 0.12–1.84% (avg. 1.15%), respectively.

The mean soil toxic metal contents show the following decreasing order: Cr (63.4 mg $\mathrm{kg^{-1}}$) > Zn (47.3 mg $\mathrm{kg^{-1}}$) > Pb (36.0 mg $\mathrm{kg^{-1}}$) > Cu (17.0 mg $\mathrm{kg^{-1}}$) > Ni (14.4 mg $\mathrm{kg^{-1}}$) > As (8.90 mg $\mathrm{kg^{-1}}$) > Cd (0.056 mg $\mathrm{kg^{-1}}$) ≈ Hg (0.078 mg $\mathrm{kg^{-1}}$) in Weizheng; Cr (75.4 mg $\mathrm{kg^{-1}}$) > Zn (61.2 mg $\mathrm{kg^{-1}}$) > Pb (28.1 mg $\mathrm{kg^{-1}}$) > Cu (21.5 mg $\mathrm{kg^{-1}}$) > Ni (20.8 mg $\mathrm{kg^{-1}}$) > As (20.2 mg $\mathrm{kg^{-1}}$) > Hg (0.15 mg $\mathrm{kg^{-1}}$) > Cd (0.12 mg $\mathrm{kg^{-1}}$) in Jianglin; Zn (53.7 mg $\mathrm{kg^{-1}}$) > Cr (42.2 mg $\mathrm{kg^{-1}}$) > Pb (33.5 mg $\mathrm{kg^{-1}}$) > Cu (15.1 mg $\mathrm{kg^{-1}}$) > Ni (13.3 mg $\mathrm{kg^{-1}}$) > As (9.11 mg $\mathrm{kg^{-1}}$) > Cd (0.14 mg $\mathrm{kg^{-1}}$) ≈ Hg (0.12 mg $\mathrm{kg^{-1}}$) in Longjiang. Compared to the soil background values of Guangdong, the Longjiang soils are enriched in Cd, Hg, and Zn, with 50–90% of the soil samples exceeding the background values. Meanwhile, 50–100% of the soil samples in Weizheng have higher As, Cd, Cr, Cu, Hg, Ni, and Zn contents than the background values. 50–90% of the Jianglin soil samples have higher As, Cd, Cr, Hg, and Ni contents than the background values. The average Cd contents are over two times higher than the background in all the three districts, indicating distribution of Cd is likely affected by human activities. The toxic metals of Cd and Hg have higher coefficients of variation (CV > 36%) in the three districts, which also suggests the extrinsic factors have influenced the metal contents [48–50]. One-way analysis of variance (ANOVA) was employed to compare the toxic metal contents in the high NPC incidence area (Longjiang) with those in the low NPC incidence areas (Jianglin and Weizheng). As shown in Table 1, the soil As–Cr contents in HC area are much lower than those in LC areas, Weizheng and Jianglin, whereas the soil Pb content in HC area is significantly higher than that in LC area, Weizheng. Previous study found that significant association of soil nickel exposure with NPC mortality rate [19]. No significant differences of Ni contents were observed between the LC area and HC area in Sihui, besides Cd, Cu, Hg and Zn. Figure 2 shows the spatial distributions of toxic metals in Longjiang, Weizheng, and Jianglin. The Cd, Cr, Cu, Ni, and Zn highs are distributed in HC area, while the As highs are confined only in LC area, Jianglin. The Hg highs are distributed in LC areas, Jianglin and Weizheng, and Pb highs are found in both LC area, Weizheng, and HC area.

## 3.2 Toxic metal pollution assessment

Table S7 shows the mean values of single pollution indices and the $PI_N$ of the surface soils of the three studied districts. The spatial distribution of $PI_N$ is shown in Figure S1. The results show that the toxic metal pollution levels decrease in the order of Cd > As ≈ Cr > Cu > Pb > Zn > Ni > Hg in Weizheng, As > Cr > Cu > Cd > Pb > Ni > Zn > Hg in Jianglin, and Cd > Pb > Cu > Cr > Zn > As > Ni > Hg in Longjiang. Overall, the mean $PI_N$ decrease in order of Jianglin > Weizheng > Longjiang. The $PI_N$ vary in all the districts. The $PI_N$ values are 0.19–1.14 (mean 0.49), 0.33–1.50 (mean 0.67), and 0.17–4.35 (mean 0.44) in Weizheng, Jianglin, and Longjiang, respectively. According to the $PI_N$ results, 92.6%, 4.3%, 2.1%, and 1.1% of the Longjiang soils are classified as "clean," "warning limit," "slightly polluted," and "seriously polluted," respectively. For the soils in Weizheng and Jianglin, 88.2% and 62.5% are "clean," 5.9% and 25% are "warning limit," and 5.9% and 12.5% are "slight polluted," respectively. The toxic metal pollution levels in the study area mostly range from clean to slightly polluted, suggesting the high soil quality. Several samples do not meet the soil quality standards in China and worth further study. The most polluted areas are found in the southern parts of Weizheng and in northern part of Longjiang.

## 3.3 Sources of toxic metals

The EF was used to discriminate the natural vs anthropogenic toxic metal sources. EF values of the toxic metals in this study are (Table S8) as follows: Cd: 0.23–16.73 (mean 1.67); As: 0.22–7.45 (mean 0.95); Hg: 0.33–6.40 (mean 1.50); Cu: 0.12–6.16 (mean 0.78); Ni: 0.15–4.73 (mean 0.80); Zn: 0.33–3.91 (mean 0.93); Cr: 0.16–3.08 (mean 0.78); Pb: 0.19–1.76 (mean 0.63). EF values of 0.5–1.5 indicate that the metals are totally from the natural source, whereas values > 1.5 suggest more likely the anthropogenic source [51]. Actually, our study area lacks industrial activities, thus presence of elevated soil metal contents is caused mainly by utilization of pesticide and fertilizer in agricultural soils. China consumes nearly one-third of the world's fertilizer. About 20% of toxic metals inputs (i.e., Cr) were from phosphate and compound fertilizers [50,52]. Moreover, with depletion of high-quality phosphate rock around the world, the metal content in phosphate fertilizer may increase. Moreover, the application of pesticides has also resulted in excessive toxic metals in agricultural soils, especially for copper and cadmium [50,52]. According to a statistics analysis (Table 1), the coefficients of variations of Cd and Hg are higher than 50%, and more than 50% of soil samples exceeding the background value of Cd and Hg in three districts. The mean EF values of Cd and Hg in the study area are more than 1.50. The EF values of Cd in 68.1% and Hg in 62.2% are higher than 1. The variation coefficients and EF indicate enrichments and anthropogenic inputs of Cd and Hg in soil of the study areas, which are possibly contributed by agricultural activities in the study area.

The Pearson's correlation coefficient matrix among toxic elements can reflect the bonding, sources, and migration of these elements [53,54]. Table 2 shows the Pearson's correlation matrix of the soil toxic metals in the study area. It shows that the coefficient between Cu and Ni is 0.934 ($p < 0.01$), indicating a strong linear correlation and a common origin for these metals. The other highly correlated pairs are Cu and Zn (0.893), Ni and Zn (0.888), Cr and Ni (0.872), followed by Cd and Zn (0.840, $p < 0.01$) [55,56]. OM exhibits distinct positive correlations with Cr (0.210), Ni (0.204), Zn (0.197), Cd (0.189), Cu (0.183) ($p < 0.05$), and Hg (0.240) ($p < 0.01$), indicating that OM may be an important factor for the distribution of Cu, Ni, Zn, Cd, Cr, and Hg in the soil of Sihui. Toxic metals can be retained in the organic matter of the soils [55,56]. No significant correlation between toxic metals and pH was found in the soils of Sihui ($p > 0.05$).

The hierarchical cluster analysis was performed to reveal any similarity in the sources and spatial distributions between different toxic metals [57]. Based on the furthest neighbor cluster method, results of the hierarchical cluster analysis are illustrated in Figure S2. Cu, Ni, Zn, Cr, and Cd are clustered into Group I. Many studies concluded that the soil Cd, Cu, and Zn contents are mainly influenced by mixed natural and anthropogenic sources or

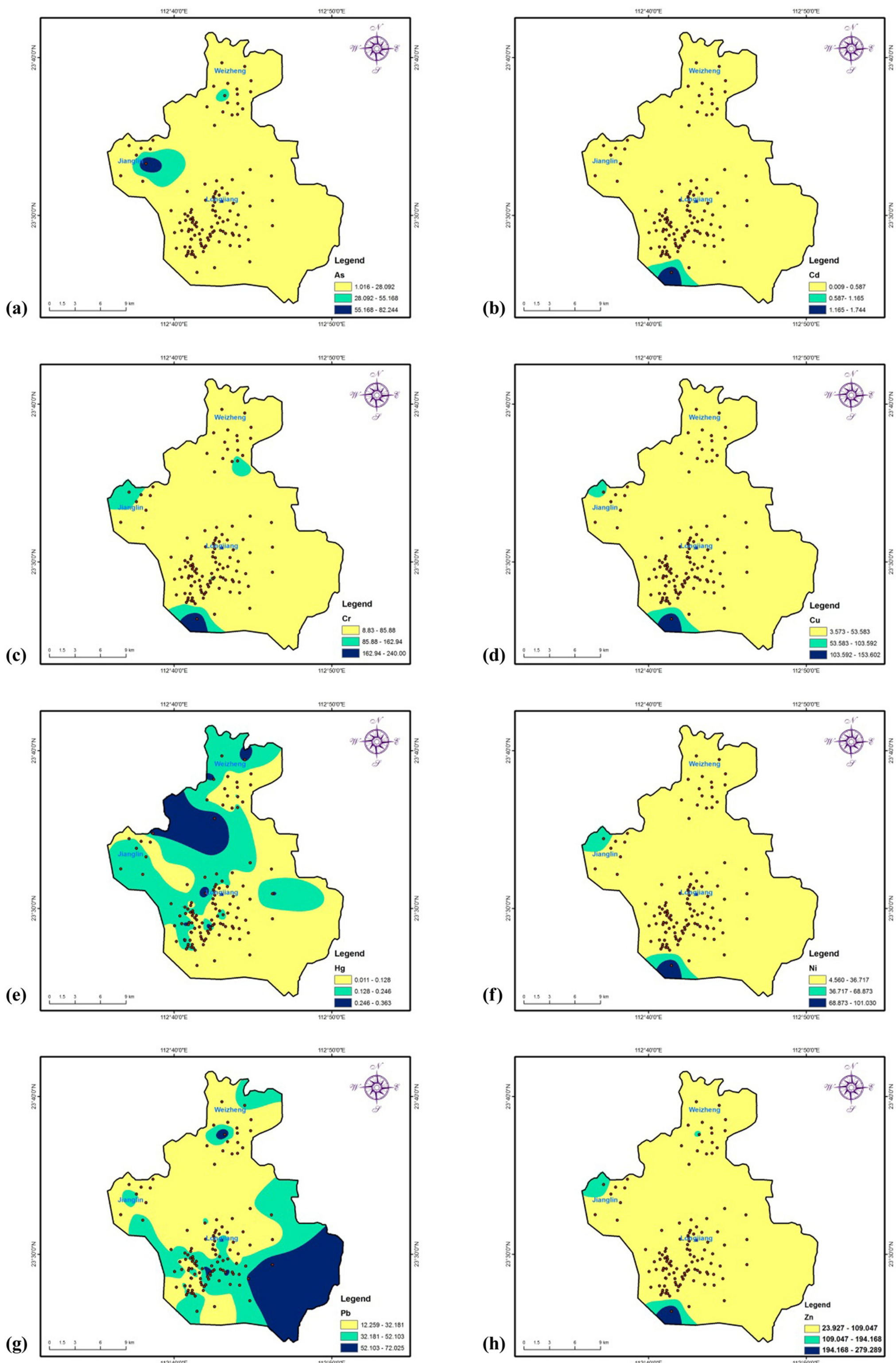


**Figure 2:** Spatial distribution of As (a), Cd (b), Cr (c), Cu (d), Hg (e), Ni (f), Pb (g), and Zn (h) in agricultural soils from the study area.

the retention phenomenon [58,59]. Mercury and arsenic belong to Group II and their concentrations are not significantly related to other toxic metals ($p > 0.05$). Lead belongs to Group III.

**Table 2:** Pearson correlation matrix between soil properties and toxic metals.

| | As | Cd | Cr | Cu | Hg | Ni | Pb | Zn | Mn | $Al_2O_3$ | $Fe_2O_3$ | pH | OM |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| As | 1 | | | | | | | | | | | | |
| Cd | 0.064 | 1 | | | | | | | | | | | |
| Cr | 0.253** | 0.587** | 1 | | | | | | | | | | |
| Cu | 0.122 | 0.840** | 0.793** | 1 | | | | | | | | | |
| Hg | 0.081 | 0.014 | 0.145 | 0.021 | 1 | | | | | | | | |
| Ni | 0.155 | 0.724** | 0.872** | 0.934** | 0.051 | 1 | | | | | | | |
| Pb | 0.059 | 0.088 | 0.076 | 0.183* | 0.028 | 0.187* | 1 | | | | | | |
| Zn | 0.128 | 0.780** | 0.741** | 0.893** | 0.040 | 0.888** | 0.391** | 1 | | | | | |
| Mn | −0.003 | 0.212* | 0.159 | 0.322** | −0.120 | 0.335** | 0.397** | 0.407** | 1 | | | | |
| $Al_2O_3$ | −0.162 | −0.040 | −0.177 | −0.053 | −0.006 | −0.067 | 0.430** | 0.115 | −0.099 | 1 | | | |
| $Fe_2O_3$ | 0.132 | 0.036 | 0.149 | 0.153 | −0.004 | 0.223* | 0.271** | 0.291** | 0.039 | 0.648** | 1 | | |
| pH | 0.058 | 0.014 | 0.153 | 0.059 | 0.137 | 0.090 | −0.001 | 0.071 | 0.058 | −0.017 | −0.129 | 1 | |
| OM | 0.072 | 0.189* | 0.210* | 0.183* | 0.240** | 0.204* | 0.047 | 0.197* | 0.110 | −0.046 | 0.011 | 0.059 | 1 |

*Correlation is significant at 0.05 levels (two-tailed).
**Correlation is significant at 0.01 levels (two-tailed).

PCA was applied to identify and analyze the potential sources of eight toxic metals. The rotated component matrix of PCA is presented in Table 3. The PCA results agree with the results of the cluster analysis. Figure 3 shows the loadings of the three principal components extracted for the surface soils. The Kaiser–Meyer–Olkin (KMO) and Bartlett's values are 0.78 and 831.25 (df = 28, Sig < 0.01), respectively, indicating that PCA may be suitable to reduce data size. Three principal components with eigenvalues >1 account for 80.41% of the total variance. In the interpretation of PCA patterns, Nowak [60] considered the factor loadings greater than 0.71 are excellent.

Factor 1 includes Cu, Ni, Zn, Cd, and Cr, with high positive loadings of >0.8, and account for 52.41% of the total variance. The spatial variations of Cu, Ni, Zn, Cd, and Cr are consistent in the study area. EF values of Cu, Ni, Zn, and Cr indicate that the spatial distributions of these metals are mainly influenced by natural sources. EF values indicate that Cd is clearly affected by anthropogenic input. Cadmium is grouped with Cu, Zn, Ni, and Cr, which indicates input of Cd both from natural and anthropogenic sources [61]. EF values of Cu, Zn, Ni, and Cr in several sample sites are over 1.5, which is likely contributed by point pollution sources. Therefore, Cu, Ni, Zn, Cd, and Cr are not only controlled by parental material but also by anthropogenic sources such as the discharge of industrial and domestic waste water [62]. Factor 2 includes Hg and As, with high positive loadings of >0.7, and accounts for 14.31% of the total variance. The concentrations of Hg and As are not significantly related to other toxic metals ($p$ > 0.05). The mean concentration of Hg and As are higher than their background values in the three districts. High concentrations of Hg and As are found in the farmlands in Weizheng and Jianglin. The EF values of Hg show that 39.5% of the sample sites are greater than 1.5, suggesting an anthropogenic source. This is probably contributed by agricultural nonpoint sources such as fertilizer and Hg-containing pesticides, which are frequently used in the rural area. The long-term application of fertilizers (especially phosphate ones) in agriculture can increase the soil As content [63]. Thus, Factor 2 can be attributed to anthropogenic agricultural input [64,65]. Factor 3 includes only Pb, with high positive loading of 0.989, and accounts for 13.70% of the total variance. Different from other metals, the mean Pb concentrations in the three districts are lower than its background value. Lead has lower coefficients of variation in surface soil in Sihui.

**Table 3:** Rotated component matrix of toxic metals in agricultural soils

| | Component | | |
|---|---|---|---|
| | Factor 1 | Factor 2 | Factor 3 |
| Cu | 0.970 | 0.019 | 0.079 |
| Ni | 0.955 | 0.094 | 0.083 |
| Zn | 0.910 | 0.037 | 0.316 |
| Cd | 0.867 | −0.065 | −0.008 |
| Cr | 0.857 | 0.274 | −0.047 |
| Hg | −0.011 | 0.749 | −0.010 |
| As | 0.113 | 0.702 | 0.045 |
| Pb | 0.108 | 0.034 | 0.989 |
| % of variance | 52.4 | 14.3 | 13.7 |
| % of cumulative | 52.4 | 66.7 | 80.4 |

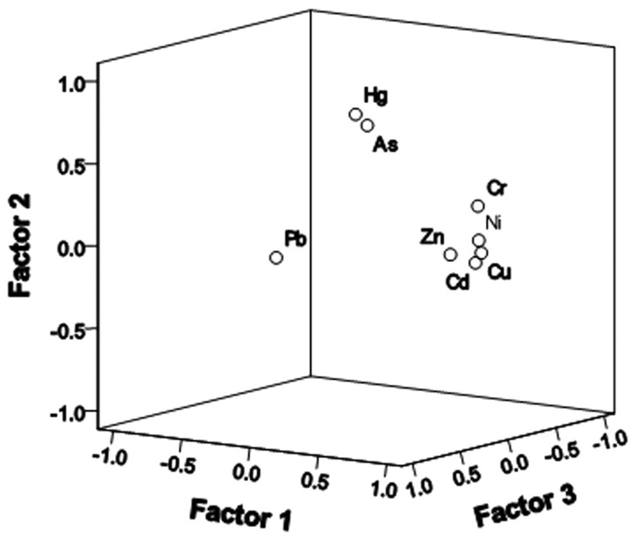


Figure 3: Principle component analysis loading plot of eight toxic metals in agricultural soils from the study area.

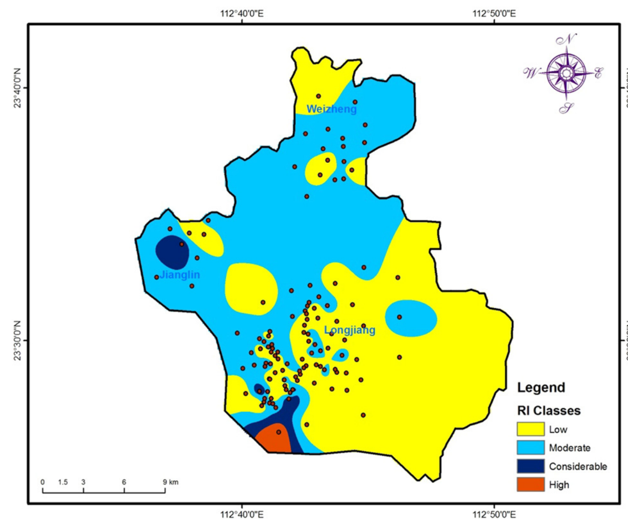


Figure 4: Spatial distribution pattern of risk index.

Lead shows positive correlations with Mn, $Al_2O_3$, and $Fe_2O_3$ ($p < 0.01$). Mn, Al, and Fe are major rock-forming elements, and are commonly considered to come from rocks and regolith [66]. Hence, the distribution of Pb is likely controlled by natural lithogenic sources.

## 3.4 Potential ecological risk of soil toxic metal pollution

Evaluation for the toxic metal concentration in the surface soil by the potential ecological risk index (PERI) is presented in Table S9. Figure 4 shows the occurrence and distribution of the toxic metal PERI in the study area, which decreases in the order of Cd > Hg > As > Cu > Ni > Pb > Cr > Zn. Cadmium and Hg mostly exhibit moderate and considerable potential ecological risk in the three districts. The other toxic metals likely pose much lower levels of risk (mean risk factor < 40), indicating low ecological risk. The average PERI values, calculated as the sum of the mean risk factors of the toxic metals, are 185.02, 187.32, and 161.05 in Weizheng, Jianglin, and Longjiang, respectively, indicating an overall moderate ecological risk posed by the toxic metals. The PERI of the surface soils decreases in the order of Jianglin > Weizheng > Longjiang. 23.5%, 25.0%, and 54.3% of the sample sites exhibit low level of risk in Weizheng, Jianglin, and Longjiang, respectively, while 76.5, 62.5, and 41.5% of the sample sites show moderate risk. Only the areas in the center of Jianglin and southwest of Longjiang show considerable and high potential risk. The PERI of high NPC incidence area is not higher than that of low NPC incidence area. The RI values, EF ratios, as well as variation coefficient all suggest that there were Cd and Hg pollution. The mean PERI coefficients of Cd and Hg in all districts are within the range of 40–80, which belongs to moderate risk. As, Cr, Cu, Ni, Zn, and Pb in three districts are all below 40, indicating low ecological risk.

## 3.5 Health risk assessment for exposure to soil toxic metals

Noncarcinogenic risks of soil toxic metals for adults in the study area are shown in Figure 5 and Table S10. The HIs of As were highest in Weizheng, Jianglin, and Longjiang with ratios to the total noncarcinogenic HI of 58.0%, 56.5%, and 53.9%, respectively. The HIs of Cr were the second highest in three regions. Among three exposure pathways, ingestion contributed to over 75% of the total HIs value and dermal contact contributed to over 20% in three regions. The effect of inhalation posed little risk. The importance order of three exposure pathways was consistent with the results of health risk assessment of previous researches [2,67]. The mean HI value of Longjiang was lower than those of Weizheng and Jianglin. The mean and maximum HQ values of all soil toxic metals and HI values of three regions were below the threshold value of 1, which indicated that there were no adverse noncarcinogenic health effects of

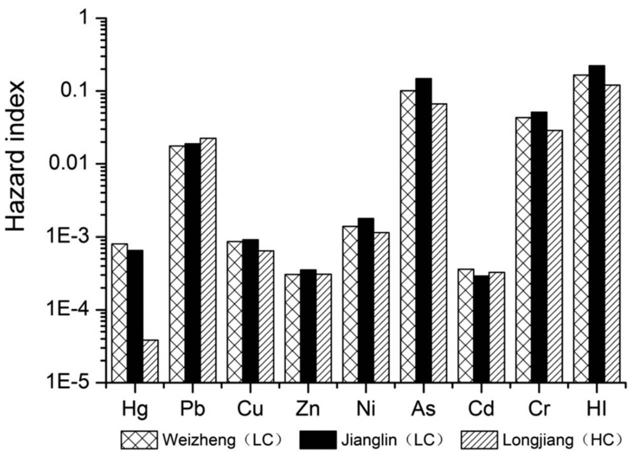


**Figure 5:** Hazard index of toxic metals for adults in the study area.

soil toxic metals to adults in agricultural soil of the study area.

The carcinogenic risks of As, Cd, and Cr of soil toxic metals in three regions were calculated (Table S11). The mean values of total carcinogenic risks ($CR_T$) were in the following order: Jianglin > Weizheng > Longjiang. The lifelong carcinogenic risks for three exposure pathways of toxic metals decreased in the following sequence: ingestion > dermal contact > inhalation. All the mean values of air inhalation in three regions were lower than $10^{-6}$, which indicated no risk. The mean values of carcinogenic risks of As and Cr via ingestion and dermal contact and $CR_T$ in the study area were within the warning range, from $10^{-6}$ to $10^{-4}$. Although the present level was tolerable, necessary policies and remediation were needed to increased carcinogenic risks of As and Cr. Notably, in this study, human risk assessment of heavy metals exposure may be overestimated. In fact, an increasing number of scholars believe that the relative bioavailability (RBA) of heavy metals is also an important factor that needs to be considered in assessments of human health risks, apart from the total concentrations of heavy metals in soils, and risk assessments based on RBA are more reliable than those only based on concentration [3,38,68].

## 4 Conclusion

The average Cd contents in agricultural soils are over two times higher than the background in both high and low NPC incidence areas of Sihui. The soil As–Cr contents in Longjiang are significantly lower than those in Weizheng and Jianglin, whereas the soil Pb content in Longjiang is significantly higher than Weizheng. No significant differences in Cd, Cu, Hg, Ni, and Zn contents are observed among the three districts. The $PI_N$ of soils from three districts decreased in the following order: Jianglin > Weizheng > Longjiang.

Multivariate analyses show that Cd, Cu, Ni, Zn, and Cr were not only controlled by the parent material but also by anthropogenic sources. Mercury and arsenic can be attributed to agricultural activities, while Pb was mainly controlled by natural lithogenic source.

The potential ecological risk of high NPC incidence area was not higher than that of low-incidence area. The mean HI value of Longjiang was lower than those of Weizheng and Jianglin. There were no adverse non-carcinogenic health effects of soil toxic metals to adults in the study areas. Carcinogenic risks of As and Cr via ingestion and dermal contact and $CR_T$ were within the warning range, from $10^{-6}$ to $10^{-4}$. Hence, toxic metals in the soil were not a major geochemical carcinogenic factor of high NPC incidence in Sihui. But necessary policies and remediation were needed to control the increased carcinogenic risks of As and Cr.

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