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Review

Spatial Characteristics, Health Risk Assessment and Sustainable Management of Heavy Metals and Metalloids in Soils from Central China

Fei Li [1,2], Ying Cai [1,2] and Jingdong Zhang [1,2,*]

1 Research Center for Environment and Health, Zhongnan University of Economics and Law, Wuhan 430073, China; lifei@zuel.edu.cn (F.L.); 1993cy@zuel.edu.cn (Y.C.)
2 School of Information and Safety Engineering, Zhongnan University of Economics and Law, Wuhan 430073, China
* Correspondence: jdzhang@zuel.edu.cn; Tel.: +86-027-8838-5169

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Abstract: The contents of seven toxic metals (Cu, Cr, Cd, Zn, Pb, Hg and As) in soils from Central China, including Henan Province, Hubei Province and Hunan Province, were collected from published papers from 2007 to 2017. The geoaccumulation index, health risk assessment model and statistics were adopted to study the spatial contamination pattern, to assess the human health risks and to identify the priority control pollutants. The concentrations of soil metals in Central China, especially Cd (1.31 mg/kg), Pb (44.43 mg/kg) and Hg (0.19 mg/kg), surpassed their corresponding background values, and the I_{geo} values of Cd and Hg varied the most, ranging from the unpolluted level to the extremely polluted level. The concentrations of toxic metals were higher in the southern and northern parts of Central China, contrasting to the lowest contents in the middle parts. For non-carcinogenic risk, the hazard index (HI) values for the children in Hubei Province (1.10) and Hunan Province (1.41) exceeded the safe level of one, with higher health risks to children than adults, and the hazard quotient (HQ) values of the three exposure pathways for both children and adults in Central China decreased in the following order: ingestion > dermal contact > inhalation. For carcinogenic risk (CR), the CR values for children in Hubei Province (2.55×10^{-4}), Hunan Province (3.44×10^{-4}) and Henan Province (1.69×10^{-4}), and the CR for adults in Hubei Province (3.67×10^{-5}), Hunan Province (4.92×10^{-5}) and Henan Province (2.45×10^{-5}) exceeded the unacceptable level (10^{-4}) and acceptable level (10^{-6}), respectively. Arsenic (As) appeared to be the main metalloid for both children and adults causing the high carcinogenic risk. For sustainable development in Central China, special attention should be paid to Cd, Hg, Cr, Pb and As, identified as the priority control soil metals. Importance should also be attached to public education, source control, and the remediation of the highly contaminated soils, especially in the areas where it can endanger the groundwater. Furthermore, it is necessary to appropriately adjust the industrial structure and cooperate more to form a complete economic zone.

Keywords: soil contamination; toxic metals; spatial distribution; health risk assessment; sustainable management; Central China

1. Introduction

Toxic metals and their compounds are naturally ubiquitous throughout the soil environment; they are highly toxic and do not easily decompose [1–3]. Heavy metals and metalloids that accumulate in the soil can inhibit soil function, poison plants and contaminate the food chain [4–6]. When heavy metals and metalloids have been transported from the soil to other environmental media, such as groundwater or crops, they can pose a threat to human health as a consequence of inhalation or ingestion through the water supply and food chain [7–13]. In addition, direct oral intake of soil particles by humans, particularly children, also poses a health hazard [14–16]. As natural components of the Earth's crust, heavy metals and metalloids are generally present at low concentrations in natural soils. However, the toxic metal contents in soils has greatly increased through anthropogenic toxic metal inputs, including dumping wastes, waste incineration, vehicle emissions, smelting, smokestack emissions, fertilizer application and sewage sludge production [17–19]. Due to their potential toxic, persistent and irreversible characteristics, toxic metals such as Cd, Cr, As, Hg, Pb, Cu, Zn and Ni, have been listed as priority control pollutants by the United States Environmental Protection Agency (USEPA) and caused more and more attention in many parts of the world [20–22].

In China, soil contamination by toxic metals has been ubiquitous and serious due to the rapid industrial development and fast urban expansion, with 10 million m^2 land and 12 million tons of grains polluted [23–27]. The literature investigating soil metals is growing more and more, and soil contamination has been found in every piece of land in the motherland, like Beijing [28], Shanghai [29], Wuhan [30], Changsha [31]. Most of the existing studies selected a smaller area as the research object, such as a city [32], or even a district [33], and little information is available for whole provinces, not to mention a whole region. Central China is one of the seven geographical divisions in China, including Henan Province, Hubei Province and Hunan Province. Central China has witnessed rapid economic growth, with a gross domestic product (GDP) of RMB 10,370.262 billion in 2016. A representative of the National Development and Reform Commission summed up the key to the rise of the Central China plan as needing to pay attention to solving two problems: the food problem and the issue of environmental protection. It is a pity that environmental problems have not been solved yet, especially soil contamination. In 2006, a sensational soil pollution incident broke out in Hubei Province, with the revelation that over 70% of the area in Heshan Block was polluted and the total amount of earthworks polluted reached about 300,000 cubic meters, with the main pollutants being organic phosphorus and organochlorine pesticides, namely DDT (Clofenotane) and BHC (Benzene hexachloride). In May 2013, it caused a sensation that a large quantity of poisonous rice containing cadmium produced in Hunan was found in Guangdong, China. In recent years, cadmium wheat events occurred frequently in Xinxiang, Henan Province. Therefore, it is of significant importance to systematically study contamination features and the health risks of soil metals in Central China for sustainable green development.

The region of Central China has a long history and strong cultural heritage, and rich mineral and industrial resources, which accounts for the soil metal pollution. To investigate soil pollution status in Central China and to provide important information for soil pollution management, this study aims to (i) collect heavy metal and metalloid concentrations, (ii) survey their spatial distribution, (iii) assess the pollution level of heavy metals and metalloids using a geoaccumulation index, (iv) evaluate the health risks, (v) identify the priority control metals and areas, and (vi) provide sustainable management suggestions to Central China.

2. Materials and Methods

2.1. Study Area

The region of Central China is located in the central and middle reaches of the Yellow River and the middle reaches of the Yangtze River region, which is one of China's seven major geographical divisions, including Henan, Hubei and Hunan Provinces, as shown in Figure 1. It has a population of approximately 222 million, within an area of 560,000 square kilometers, accounting for about 5.9% of the total land area. It belongs to a temperate monsoon climate and subtropical monsoon climate zone and its annual average temperature is 15–21 °C. It has strong history and culture, rich mineral resources, a strong industrial base, and convenient water and land transportation. Central China is a slightly more economically developed area, the heart of China's industry, agriculture and transportation. In 2016, Central China had a GDP of RMB 10,070.262 billion, with Henan, Hubei and Hunan Provinces having a GDP of RMB 4016.001, 3229.791, and 3124.470 billion, respectively. The industrial structure ratio of three kinds of industry—agriculture, industries and services—is 10.7:47.4:41.9 in Henan Province, 10.8:44.5:44.7 in Hubei Province, 11.5:42.2:46.3 in Hunan Province, respectively.

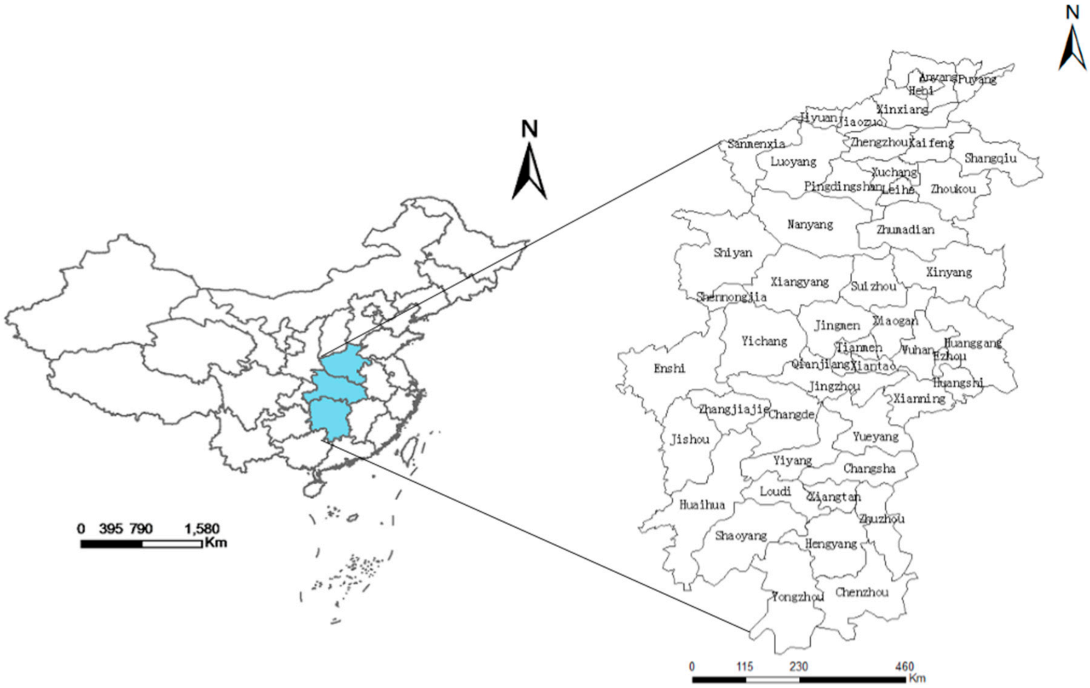


Figure 1. Map of soil sampling sites in Central China.

2.2. Data Collection and Processing

This study collected published papers on heavy metal and metalloid concentrations in surface soils in Central China from 2007 to 2017, shown in Table 1, through searching "Hubei/Hunan/Henan", "heavy metal", "metals", "soil" and other relevant key words in databases, such as China National Knowledge Infrastructure (CNKI, http://www.cnki.net/), Wanfang Data (http://www.wanfangdata.com.cn/), VIP (http://www.cqvip.com/), Web of Science (http://login.webofknowledge.com/), Science Direct (https://www.elsevier.com/solutions/sciencedirect), and Baidu Scholar (http://xueshu.baidu.com/). According to the principles of literature screening—for example that the literature is not repeated, the literature data is from a field measurement, the sampling location is clear, the number of samples can be verified, and the detection of heavy metal content is accurate—the search collected a total of 85 papers in 35 cities—specifically 11 cities in Henan Province, 16 cities in Hubei Province, and eight cities in Hunan Province—which covered 105,402 effective data. Seven heavy metals and metalloids—Cu, Cr, Cd, Zn, Pb, Hg and As—were collected in this study.

Table 1. The concentrations (mg/kg) of soil metals in different cities in Central China.

Province	City	Cu	Cr	Cd	Zn	Pb	Hg	As	References
Henan Province	Kaifeng	33.91	56.15	1.19	172.90	46.15		6.31	[34–36]
	Luoyang	47.38	66.47	0.65	190.25	51.23			[37,38]
	Jiaozuo	31.57	176.15	0.39	81.43	24.32		17.25	[39–41]
	Zhengzhou	21.82	54.46	0.16	70.55	30.90	0.05	8.74	[42–47]
	Xinxiang	40.28	116.83	12.46	219.93	52.63	0.32		[33,48–55]
	Zhoukou	26.71	56.73	0.05	67.93	29.66			[56,57]
	Shangqiu	22.42	55.26	0.18	66.93	19.51			[58,59]
	Sanmenxia	45.25	118.73	0.25	81.77	21.27	0.16	7.97	[38,60]
	Puyang	36.20	40.60	0.13	118.00	49.20			[61]

Table 1. *Cont.*

Province	City	Cu	Cr	Cd	Zn	Pb	Hg	As	References
	Anyang	22.74	51.96	0.11	55.10	17.27	0.08		[62]
	Nanyang	20.75	54.36	1.12	75.81	29.76		12.68	[63,64]
Hubei Province	Enshi	32.88	93.28	0.69	96.75	31.95	0.12	13.78	[65–70]
	Yichang	26.31	2.39	0.26	0.47	29.16			[70–72]
	Wuhan	28.75	59.02	1.60	87.73	34.00	0.23	12.92	[70,73–79]
	Shiyan	34.73	67.95	0.39	78.72	30.72	0.06	13.78	[64,70,80]
	Tianmen	27.89	67.94	0.16	69.97	23.02	0.11	6.85	[70,81,82]
	Huangshi	126.72	39.11	1.32	95.67	79.12	0.17	33.72	[70,83–89]
	Xiaogan	25.85	78.75	0.25	63.65	46.21			[70,90,91]
	Ezhou	7.67	62.68	0.10	70.05	29.21	0.09	10.50	[70,92]
	Xiangfan	24.76	60.10	0.10	66.48	27.25	0.09	9.96	[70,93]
	Xiantao	29.72	87.30	0.14	77.29	25.82		8.25	[70,94]
	Huanggang	35.06		0.12	70.15	24.15			[70]
	Jingmen	24.28		0.11	55.67	284.00			[70]
	Jingzhou	33.55		0.14	89.29	32.99			[70]
	Qianjiang	32.06		0.11	78.33	24.98			[70]
	Shennongjia	44.36		0.11	69.66	28.63			[70]
	Suizhou	24.46		0.13	63.35	30.10			[70]
	Xianning	24.87		0.11	60.01	31.98			[70]
Hunan Province	Changsha	57.13	56.64	6.26	249.13	91.46	0.48	42.78	[31,95,96]
	Zhuzhou	44.65	115.50	1.31	170.15	81.98	0.25	20.05	[97–100]
	Changde	31.47	26.00	0.49	102.08	29.43	0.45	68.86	[101–103]
	Hengyang	57.63	31.56	1.59	184.62	86.43	0.42	41.50	[100,104,105]
	Xiangtan	37.50	77.43	25.22	863.58	68.81	0.25	61.20	[103,106]
	Chenzhou	36.68		4.33	634.30	467.92		74.44	[100,107,108]
	Loudi	33.26	93.03	0.59	107.24	37.82	0.18	14.96	[109]

The sampling sites of the literature mostly adopted an "S" shape, plum blossom point method with random sampling and sampled the surface soil (0–20 cm) with a multi-point mixture. The samples were naturally air dried in the laboratory, acid digested after sieving, and the contents of toxic metal elements were almost always obtained by the same method, including atomic absorption spectrometer (AAS), atomic fluorescence spectrometer (AFS) and inductively coupled plasma mass spectrometry (ICP-MS). In order to control the unexpected uncertainty in the data collection and processing, the data were mostly detected by the recommended methods in the national standard (GB 15618-1995). As for Cu, Cr, Cd, Zn and Pb, data obtained by AAS and ICP-MS accounted for 66.7% and 30.4% respectively, and as for Hg and As, data obtained by AFS accounted for 65.5%. From the collected literature, the minimum data we collected were greater than the maximum instrument detection limits. The instrument detection limit of AAS and ICP-MS were 0.001 ng/mL (AAS) and 0.04 ng/mL (ICP-MS) for Cu, 0.001 ng/mL (AAS) and 0.04 ng/mL (ICP-MS) for Cr, 0.0003 ng/mL (AAS) and 0.012 ng/mL (ICP-MS) for Cd, 0.0001 ng/mL (AAS) and 0.012 μg/mL (ICP-MS) for Zn, 0.005 ng/mL (AAS) and 0.01 ng/mL (ICP-MS) for Pb, respectively. The instrument detection limits of AFS and ICP-MS were 0.001 ng/mL (AAS) and 0.018 ng/mL (ICP-MS) for Hg, 0.001 ng/mL (AAS) and 0.031 ng/mL (ICP-MS) for As, respectively. The concentration ranges were 7.67–126.72 mg/kg (Cu), 2.39–176.15 mg/kg (Cr), 0.05–25.22 mg/kg (Cd), 0.47–863.58 mg/kg (Zn), 17.27–467.92 mg/kg (Pb), 0.03–41.10 mg/kg (Hg) and 6.31–74.44 mg/kg (As), respectively. The value ranges of pH and SOM were 4.69–8.52 and 2.8–72.3 mg/kg, respectively. It should be taken into account that the number of samples varies in each city, and the values of the toxic metal elements in each study area were calculated by the weighted average method. The corresponding number of toxic metals in the region was taken as the weight value multiplied by the corresponding element concentration of the toxic metal, divided by the sum of the number of samples of the toxic metal element in the region, then the value of the toxic metal element in each study area was obtained [110].

2.3. Geoaccumulation Index

The geoaccumulation index (I_{geo}) was put forward by Muller in the late 1960s [111], and is a geochemical criterion to evaluate pollution level in soils or sediments. It can be calculated by Equation (1).

$$I_{geo} = \log_2\left(\frac{Cn}{1.5Bn}\right) \tag{1}$$

where Cn is the measured concentration of the heavy metals and metalloids in soil (mg/kg), Bn is the geochemical background value of the corresponding heavy metals and metalloids in soil (mg/kg), and the coefficient 1.5 is used to detect very small anthropogenic influences [112]. In this study, Bn refers to the background concentration of the heavy metals and metalloids in the soils of Hubei, Hunan and Henan Province [113–115]. According to Muller [111], the geoaccumulation index consists of seven classes. The corresponding relationships between I_{geo} and the pollution level are given as follows: unpolluted ($I_{geo} \leq 0$), unpolluted to moderately polluted ($0 < I_{geo} \leq 1$), moderately polluted ($1 < I_{geo} \leq 2$), moderately to heavily polluted ($2 < I_{geo} \leq 3$), heavily polluted ($3 < I_{geo} \leq 4$), heavily to extremely polluted ($4 < I_{geo} \leq 5$), or extremely polluted ($I_{geo} > 5$).

2.4. Health Risk Assessment

Generally, an individual is exposed to soil metals through three main pathways—ingestion, inhalation and dermal contact—so the exposure scenarios we made are the most stringent settings including all the three exposures. This may need to be adapted to local conditions, because there may be a risk factor for only one or two pathways of exposure. The methodology used in this study to calculate the exposure risks of adults or children to soil metals is based on those developed by the United States Environmental Protection Agency for health risk assessment [116,117] and the Dutch National Institute of Public Health Agency [118]. The corresponding dose received through each of the three pathways was evaluated by Equations (2)–(4) [116,117,119].

$$ADI_{ing} = \frac{C \times IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{2}$$

$$ADI_{inh} = \frac{C \times InhR \times EF \times ED}{PEF \times BW \times AT} \tag{3}$$

$$ADI_{dermal} = \frac{C \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{4}$$

where ADI_{ing}, ADI_{inh} and ADI_{dermal} are the average daily intake from soil ingestion, inhalation and dermal contact, respectively (mg/kg·day); C is the concentration of the metal element in the soil (mg/kg); IngR and InhR are the ingestion and inhalation rate of soil, respectively (mg/day, m^3/day); EF is the exposure frequency (day/year); ED is the exposure duration (year); BW is the average bodyweight of the exposed individual (kg); AT is the averaged contact time (day); PEF is the particle emission factor (m^3/kg); SA is the exposed skin surface area (cm^2); AF is the adherence factor (mg/m^2·day); and ABS is the dermal absorption factor (unitless). Detailed information of the probabilistic parameters is provided in Table 2, and all the parameters refer to the literature [36–38,40], and to decrease the corresponding parameter uncertainty, the local parameters BW, SA, EXF, ED, AF, and AT included were preferentially adopted [120].

Table 2. Distribution of parameters used to evaluate exposure risks of soil metals in Central China.

Parameter	Symbol	Units	Distribution
Soil ingestion rate	IngR	mg/day	200 [a]; 100 [b]
Soil inhalation rate	InhR	m^3/day	7.6 [a]; 20 [b]
Exposure frequency	EF	day/year	350
Exposure duration	ED	year	6 [a]; 24 [b]

Table 2. *Cont.*

Parameter	Symbol	Units	Distribution
Body weight	BW	kg	15.9 [a]; 56.8 [b]
Exposure skin	SA	cm^2	2448 [a]; 5075 [b]
Adherence factor	AF	$mg/m^2 \cdot day$	0.2 [a]; 0.07 [b]
Dermal absorption factor	ABS	unitless	0.001
Particle emission factor	PEF	m^3/kg	1.36×10^9
Average contact time	AT	day	ED × 365 (non-carcinogens) 74 × 65 (carcinogens)

[a] Children; [b] Adults.

The doses calculated for each element and exposure pathway were subsequently divided by the toxicity threshold value, which is referred to as the reference dose (RfD, mg/kg·d) of a specific chemical to yield a non-carcinogenic hazard quotient (HQ), and hazard index (HI) is presented as the sum of HQ for each exposure pathway to a certain toxic metal, whereas carcinogenic risk (CR) is multiplied by the corresponding slope factor (SF, kg·d/mg) to produce a level of cancer risk [121]. They were calculated by Equations (5) and (6):

$$HI = \sum HQ_i = \sum \frac{ADI_i}{RfD_i} \tag{5}$$

$$CR = \sum ADI_i \times SF_i \tag{6}$$

where RfD_i is the corresponding reference dose for each toxic metal and for exposure pathway i; SF_i is the corresponding carcinogenic slope factor for each toxic metal and for exposure pathway I; non-carcinogenic risk is accepted when the HI value is below one; and the degree of risk increases as HI increases [117,120]. For carcinogenic risk, the exposure doses at each exposure pathway were multiplied by the SF to produce a level of cancer risk. CR represents the probability of an individual developing any type of cancer from lifetime exposure to carcinogenic hazards. The acceptable risk level for regulatory purposes is 1×10^{-6}, i.e., one over one million of the population [120]. The RfD and SF values of the studied metals are shown in Table 3, which were taken from the literature [121–125].

Table 3. The reference dose and slope factor of metals.

Elements	RfD/(mg/kg·d)			SF/(kg·d/mg)		
	ing	inh	dermal	ing	inh	dermal
Cu	4.00×10^{-2}	4.02×10^{-2}	1.20×10^{-2}	-	-	-
Cr	3.00×10^{-3}	2.86×10^{-5}	6.00×10^{-5}	-	42	-
Cd	1.00×10^{-3}	1.00×10^{-3}	1.00×10^{-5}	-	6.3	-
Zn	3.00×10^{-1}	3.00×10^{-1}	6.00×10^{-2}	-	-	-
Pb	3.50×10^{-3}	3.52×10^{-3}	5.25×10^{-4}	-	-	-
Hg	3.00×10^{-4}	3.00×10^{-4}	2.40×10^{-5}	-	-	-
As	3.00×10^{-4}	1.23×10^{-4}	3.00×10^{-4}	1.5	15.1	3.66

Although the seven metals in this study have chronic non-carcinogenic health risks, only three metals (As, Cd and Cr) have a carcinogenic risk [126]. Among them, the carcinogen of As for all three exposure pathways was calculated in the model, whereas the carcinogenic risks for Cd and Cr were considered only through inhalation. The aggregate risk was calculated by summing the individual cancer risks across all exposure pathways [119,121].

2.5. Statistics Methods

To explore the relationship between the contents of soil metals, a correlation analysis (CA) was performed with the software package SPSS version 17.0. CA refers to the analysis of two or more variable elements with dependencies to measure the relative degree of correlation between the two variables. To make the calculated data visualization spatially, the rendering method was employed by ArcGIS 9.3. According to the concentration range, the concentrations are divided into different levels, and the different levels are rendered in corresponding colors in order to distinguish between different concentration areas. The rendering method was used to spatially analyze the distribution and induce the health risk of soil metals.

3. Results and Discussion

3.1. Overview of Concentrations of Soil Metals in Central China

Basic descriptive statistics were derived to provide a summary of the concentration of soil metals in Central China, shown in Tables 1 and 4. The mean concentrations of all toxic metals exceeded their corresponding background values (BVs), especially for Cd, Pb and Hg, which were about 13.48, 1.71 and 2.92 times greater than their BVs, respectively. According to the current Chinese Environmental Quality Standard for Soils (GB 15618-1995), Class I was developed to protect the natural ecology of the region and to maintain the natural background of the soil quality, Class II was developed to protect agricultural production and corresponding human health, and Class III was developed to ensure the normal growth of agriculture, forestry and plants [127]. The results showed that the mean concentrations of all soil metals were lower than their corresponding class II values, except Cd, which was 2.18 times greater than its corresponding class II value. Specifically, the mean concentrations of all toxic metals in three provinces surpassed their corresponding BVs, except Cr in Hubei Province and As in Henan Province. The mean concentrations of all soil metals in the three provinces were within their corresponding class II values, except Cd, which accounted for the whole situation in Central China.

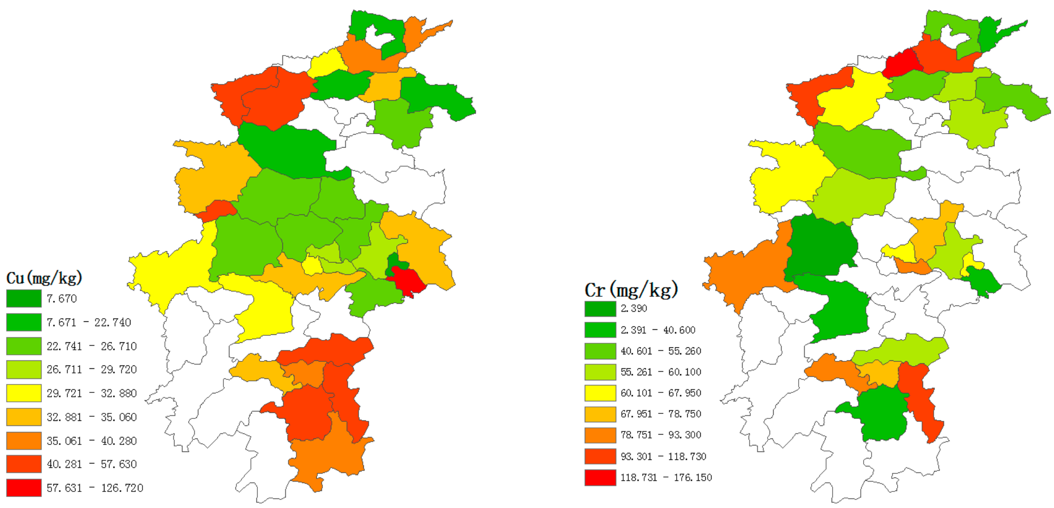
Table 4. Descriptive statistics of soil metal concentrations (mg/kg) in Central China.

		Cu	Cr	Cd	Zn	Pb	Hg	As
Central China	n	16,565	14,506	16,974	16,376	17,108	11,784	12,089
	WA	34.72	83.08	1.31	112.93	44.43	0.19	16.23
	SD	18.63	33.79	4.52	160.10	81.40	40.28	20.42
	CV (%)	1.51	1.72	6.88	3.35	3.85	18.09	3.93
Henan Province	n	6983	6561	7094	6934	7011	6603	6895
	WA	28.30	68.59	2.07	108.16	34.29	0.07	9.29
	SD	10.10	33.31	7.73	275.71	135.80	61.48	21.73
	CV (%)	3.38	7.86	17.95	11.93	14.97	27.80	6.30
Hubei Province	n	6667	5090	6842	6375	6878	3790	3789
	WA	37.09	80.69	0.80	87.82	39.91	0.12	14.00
	SD	24.21	24.62	0.44	21.00	60.37	0.05	7.93
	CV (%)	4.15	3.98	7.50	1.76	7.42	6.09	7.22
Hunan Province	n	2915	2855	3038	3067	3219	1391	1405
	WA	35.13	91.25	1.47	138.14	53.53	0.26	18.87
	SD	9.23	38.26	3.35	54.05	12.59	0.10	3.62
	CV (%)	2.64	4.14	19.88	4.26	3.18	16.07	5.59
BV_{China} [a]		22.6	61	0.097	74.2	26	0.065	11.2
BV_{Henan} [b]		20	63.2	0.065	62.5	22.3	0.025	9.8
BV_{Hubei} [c]		30.7	86	0.17	83.6	26.7	0.08	12.3
BV_{Hunan} [d]		25	68	0.07	96	30	0.07	13.41
Class I [e]		35	90	0.2	100	35	0.15	15
Class II [e]		100	250	0.6	300	350	1.0	25
Class III [e]		400	300	1.0	500	500	1.5	40

[a] CNEMC (1990) [113]; [b] Soil background value of Henan Province, China [114]; [c] Soil background value of Hubei Province, China [113]; [d] Soil background value of Hunan Province, China [115]; [e] Environmental quality standard secondary grade for soils, soil limitations to ensure agricultural production and human health (NEPAC 1995); n the number of samples, WA weighted average, SD standard deviation, CV coefficient of variation.

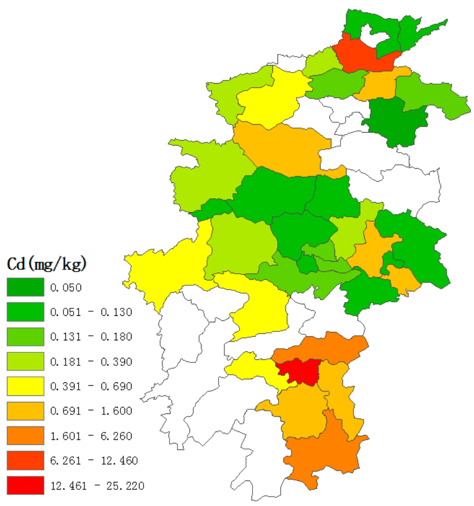
3.2. Spatial Distribution Patterns of Toxic Metals in Central China

Owing to the fact that the spatial distribution of metals is a useful way to identify hotspot areas with high metal concentrations [27,128], the corresponding distribution maps were produced and are shown in Figure 2. The percentage of high Cu concentrations decreased in the order of Hunan Province > Henan Province > Hubei Province, which was similar to Cd, Zn, Pb and Hg. The high-value zones of Cd, Zn, Pb and Hg appeared in Xiangtan city, Chenzhou city and Changsha city in Hunan Province, while that of Cu appeared in Daye city in Hubei Province. The concentration of Cr as a whole was the highest in Henan Province, followed by Hunan and Hubei Province, while the concentration of As was much higher in Hunan Province than that in Hubei and Henan Province. On the whole, the concentration of toxic metals was higher in Hunan and Henan Province, the southern and northern parts of Central China, contrasting the lowest contents in the middle parts. The analysis suggested that Huangshi city, which is rich in minerals and one of the six major copper production bases in China, accounted for the high concentration of Cu. A large number of chromium salt chemical plants in Henan Province led to the high concentration of Cr. Hunan Province, known as the town of nonferrous metal and the land of non-metal, accounted for the high background value of Zn and Pb. At the same time, the economic layout of heavy chemical industry made the toxic metal pollution more severe, with the percentage of toxic metal concentration of Hg, Cd and As in the sewage in the Xiangjiang river basin was 54.5%, 37% and 14.1%, respectively, in 2007. It has been found that areas with high levels of toxic metals are generally rich in mineral resources and in industry structure. Specifically, Huangshi city, with high-value for Cu, is famous as China's old industrial base, called the "ancient capital of bronze". As for Jiaozuo city, with a high-value for Cr, due to the regional division of labor in the state's macro-industrial layout and the advantages of abundant mineral resources in the region, in Jiaozuo, industry has formed a heavy-duty structural layout with prominent industrial positions for a long time. As for Chenzhou city, with high values for Zn, Pb and As, it is a famous non-ferrous metal town in the world. Now it has discovered about 110 kinds of mineral, which increased its heavy industry. As for Changsha city, with high values for Hg, it has a wide range of minerals, especially non-metallic minerals. Furthermore, Changsha is a medium-sized integrated city, with developed mining and transportation, which contributed to the high level of Hg.

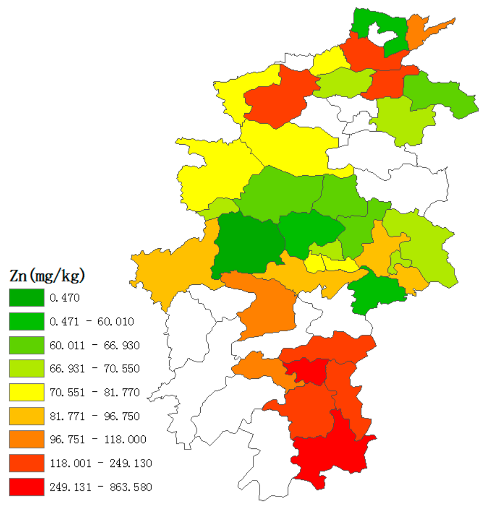


(**a**) (**b**)

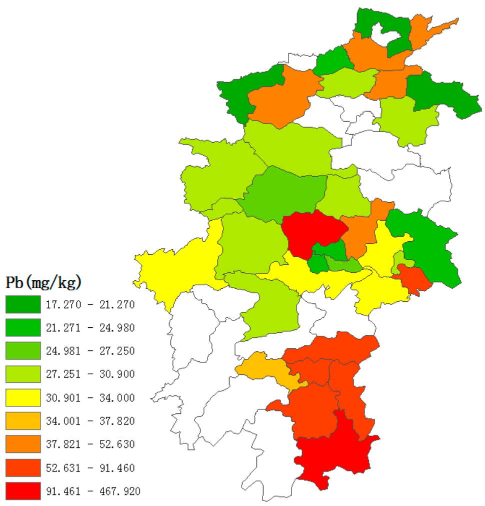
Figure 2. *Cont.*



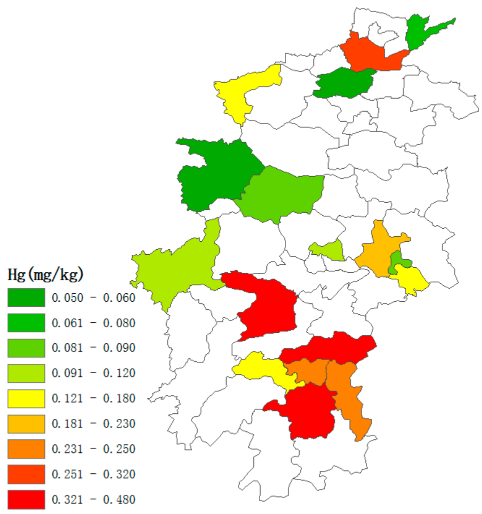
(**c**)



(**d**)

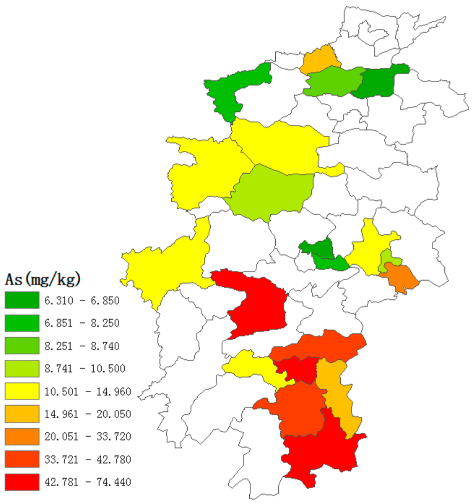


(**e**)



(**f**)

Figure 2. *Cont*.



(g)

Figure 2. Spatial distribution of Cu (**a**), Cr (**b**), Cd (**c**), Zn (**d**), Pb (**e**), Hg (**f**) and As (**g**) in soils from Central China.

3.3. Geoaccumulation Indexes of Metals

The boxplots of the geoaccumulation index (I_{geo}) values and the percentages of class distribution for the pollution assessment of heavy metals and metalloids are presented in Figure 3 and Table S1. The results showed that the I_{geo} values of Cd and Hg varied the most, ranging from the unpolluted level to the extremely polluted level. In particular, more than half of the I_{geo} values of Cd (62.2%) and Hg (52.6%) were higher than zero, suggested that soils should be unpolluted by Cd and Hg in Central China, similar to the pollution situation in the whole of China [111,128]. Specifically, more than 80% of the sites of each metal, except Cd, were unpolluted in Hubei Province, contrasting to 60% in Hunan Province. Only 11.8% sites of Cd and 5.9% sites of Pb fell into class three, which means moderate to heavily pollution. Soil in Henan Province was heavily polluted by Cd, Zn, Pb and As, especially Cd. Even 50.0% of the sites with Cd and 28.6% of the sites of Hg were extremely polluted, which accounted for the whole pollution situation in Central China. Although the concentrations of toxic metals were higher in Hunan Province than those in Henan Province, the I_{geo} values of metals were the opposite, because the background values in Hunan Province are higher than those in Henan Province.

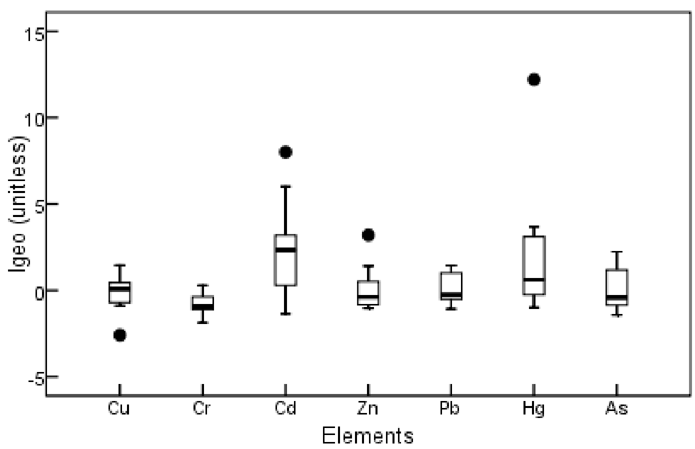


Figure 3. Boxplots of the geoaccumulation index for soil metals in Central China.

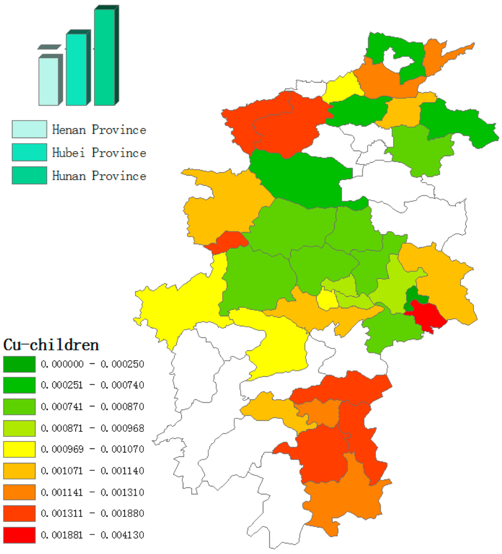
3.4. Human Health Assessment of Children and Adults in Central China

After investigating the spatial distribution and geoaccumulation indexes of the studied metals, a health risk assessment for children and adults from exposure to soil metals in Central China through possible exposure pathways was calculated using Equations (2)–(6), and the results are shown in Tables 5 and 6 and Figures 4 and 5.

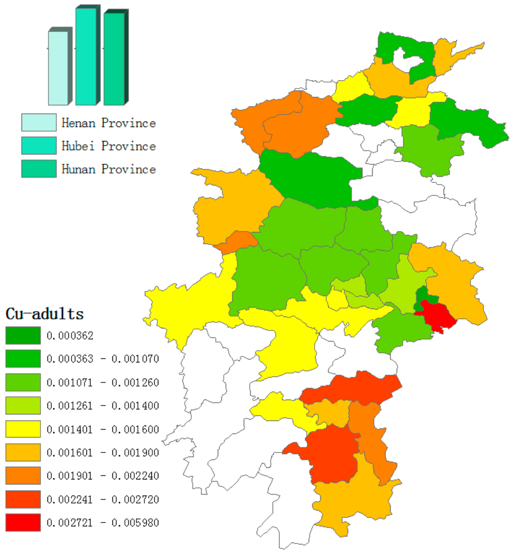
Table 5. Hazard quotients (unitless) of soil metals in Central China for children and adults.

Elements	Province	Children				Adults			
		HQing	HQinh	HQdermal	HQ	HQing	HQinh	HQdermal	HQ
Cu	Henan	8.53×10^{-3}	2.37×10^{-7}	6.96×10^{-5}	8.60×10^{-3}	1.19×10^{-3}	1.75×10^{-7}	1.41×10^{-5}	1.21×10^{-3}
	Hubei	1.12×10^{-2}	3.11×10^{-7}	9.13×10^{-5}	1.13×10^{-2}	1.57×10^{-3}	2.29×10^{-7}	1.85×10^{-5}	1.58×10^{-3}
	Hunan	1.06×10^{-2}	2.95×10^{-7}	8.64×10^{-5}	1.07×10^{-2}	1.48×10^{-3}	2.17×10^{-7}	1.76×10^{-5}	1.50×10^{-3}
Cr	Henan	2.76×10^{-1}	8.08×10^{-4}	3.38×10^{-2}	3.10×10^{-1}	3.86×10^{-2}	5.95×10^{-4}	6.86×10^{-3}	4.61×10^{-2}
	Hubei	3.24×10^{-1}	9.51×10^{-4}	3.97×10^{-2}	3.65×10^{-1}	4.54×10^{-2}	7.00×10^{-4}	8.07×10^{-3}	5.42×10^{-2}
	Hunan	3.67×10^{-1}	1.08×10^{-3}	4.49×10^{-2}	4.13×10^{-1}	5.13×10^{-2}	7.92×10^{-4}	9.12×10^{-3}	6.13×10^{-2}
Cd	Henan	2.50×10^{-2}	6.97×10^{-7}	6.11×10^{-3}	3.11×10^{-2}	3.49×10^{-3}	5.14×10^{-7}	1.24×10^{-3}	4.73×10^{-3}
	Hubei	9.60×10^{-3}	2.68×10^{-7}	2.35×10^{-3}	1.20×10^{-2}	1.34×10^{-3}	1.98×10^{-7}	4.78×10^{-4}	1.82×10^{-3}
	Hunan	1.78×10^{-2}	4.97×10^{-7}	4.35×10^{-3}	2.21×10^{-2}	2.49×10^{-3}	3.66×10^{-7}	8.84×10^{-4}	3.37×10^{-3}
Zn	Henan	4.35×10^{-3}	1.22×10^{-7}	5.32×10^{-5}	4.40×10^{-3}	6.09×10^{-4}	8.95×10^{-8}	1.08×10^{-5}	6.20×10^{-4}
	Hubei	3.53×10^{-3}	9.87×10^{-8}	4.32×10^{-5}	3.57×10^{-3}	4.94×10^{-4}	7.27×10^{-8}	8.78×10^{-6}	5.03×10^{-4}
	Hunan	5.55×10^{-3}	1.55×10^{-7}	6.80×10^{-5}	5.62×10^{-3}	7.77×10^{-4}	1.14×10^{-7}	1.38×10^{-5}	7.91×10^{-4}
Pb	Henan	1.18×10^{-1}	3.28×10^{-6}	1.93×10^{-3}	1.20×10^{-1}	1.65×10^{-2}	2.42×10^{-6}	3.92×10^{-4}	1.69×10^{-2}
	Hubei	1.38×10^{-1}	3.82×10^{-6}	2.24×10^{-3}	1.40×10^{-1}	1.92×10^{-2}	2.81×10^{-6}	4.56×10^{-4}	1.97×10^{-2}
	Hunan	1.84×10^{-1}	5.13×10^{-6}	3.01×10^{-3}	1.87×10^{-1}	2.58×10^{-2}	3.78×10^{-6}	6.11×10^{-4}	2.64×10^{-2}
Hg	Henan	2.63×10^{-3}	7.35×10^{-8}	8.05×10^{-5}	2.71×10^{-3}	3.68×10^{-4}	5.42×10^{-8}	1.64×10^{-5}	3.85×10^{-4}
	Hubei	4.81×10^{-3}	1.34×10^{-7}	1.47×10^{-4}	4.96×10^{-3}	6.73×10^{-4}	9.90×10^{-8}	2.99×10^{-5}	7.03×10^{-4}
	Hunan	1.05×10^{-2}	2.92×10^{-7}	3.20×10^{-4}	1.08×10^{-2}	1.46×10^{-3}	2.15×10^{-7}	6.50×10^{-5}	1.53×10^{-3}
As	Henan	3.73×10^{-1}	2.55×10^{-5}	9.14×10^{-4}	3.74×10^{-1}	5.23×10^{-2}	1.87×10^{-5}	1.86×10^{-4}	5.25×10^{-2}
	Hubei	5.63×10^{-1}	3.84×10^{-5}	1.38×10^{-3}	5.64×10^{-1}	7.88×10^{-2}	2.83×10^{-5}	2.80×10^{-4}	7.91×10^{-2}
	Hunan	7.59×10^{-1}	5.17×10^{-5}	1.86×10^{-3}	7.61×10^{-1}	1.06×10^{-1}	3.81×10^{-5}	3.77×10^{-4}	1.07×10^{-1}

For non-carcinogenic risk, the total HIs of studied metals for children in Central China were 1.10 (Hubei), 1.41 (Hunan), 0.852 (Henan), respectively, and the total HIs for adults were 0.158 (Hubei), 0.202 (Hunan), 0.122 (Henan), respectively. It was obviously that the children in Hubei and Hunan Province faced non-carcinogenic risks, in contrast with the children in Henan and the adults in Central China. The HQ of each toxic metal for children and adults in Central China decreased in the order of As > Cr > Pb > Cd > Cu/Hg/Zn. Cr, Pb and As caused relatively higher non-carcinogenic risks to possible receptors than the other four toxic metals. For instance, the HQ of As (0.564), Cr (0.365) and Pb (0.14) accounted for 51.31%, 33.19%, and 12.71% of the entire HI value for children in Hubei, respectively. HIs for the studied metals in Central China were higher for children than for adults [128]. In particular, the HQs for children through ingestion were averaged at 7.14 times higher than those for adults, with dermal contact 4.92 times higher and inhalation 1.36 times higher [27]. The HQ of the three exposure pathways for both children and adults in Central China decreased in the following order: ingestion > dermal contact > inhalation, which was similar to other reports [121,129,130].



(**a**)



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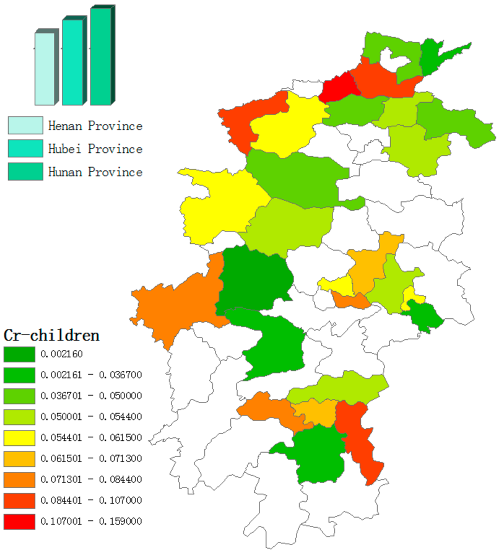

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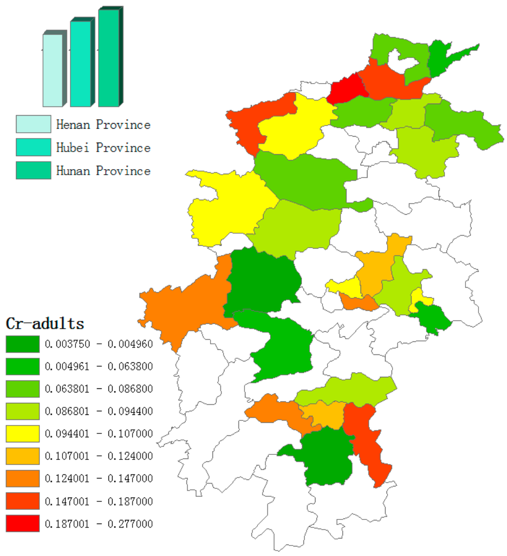
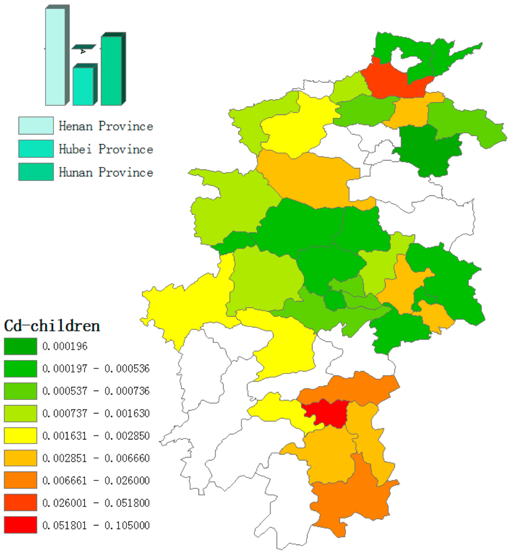
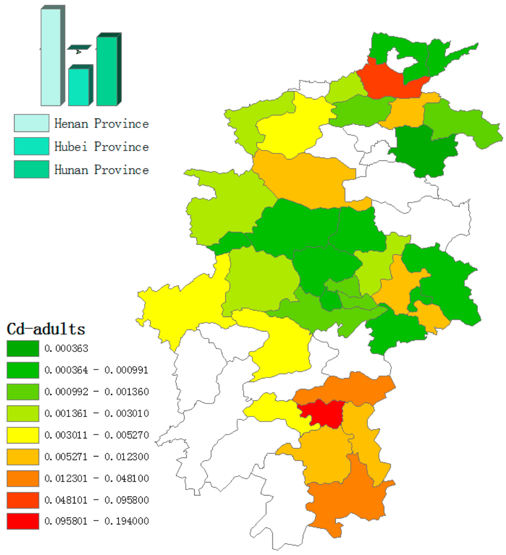


(**d**)

Figure 4. *Cont.*



(e)



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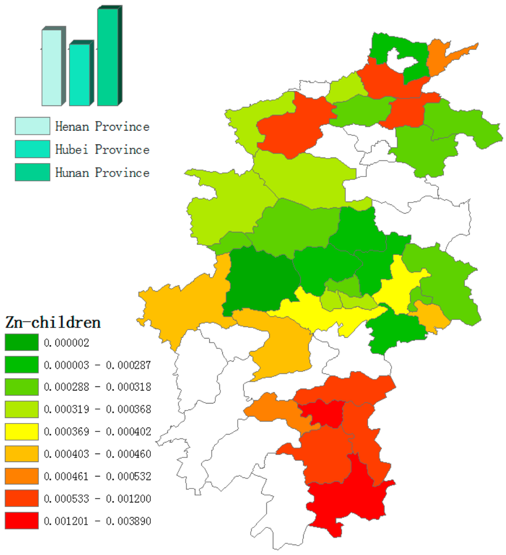

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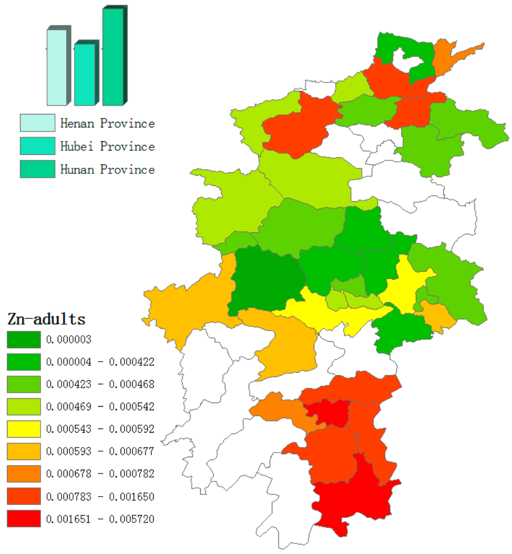

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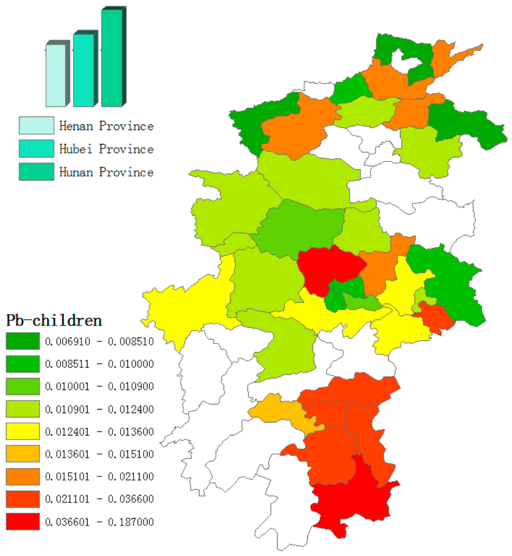
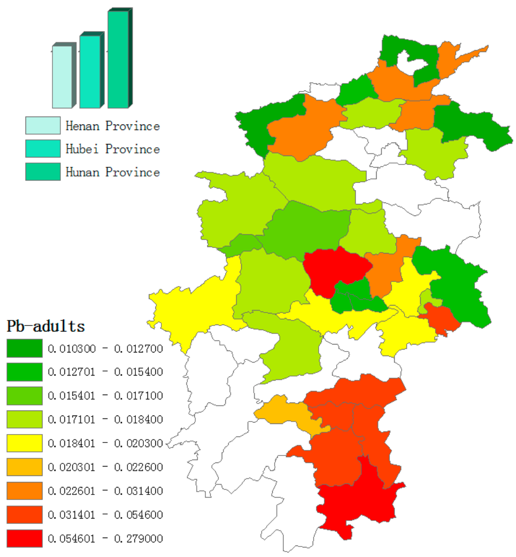
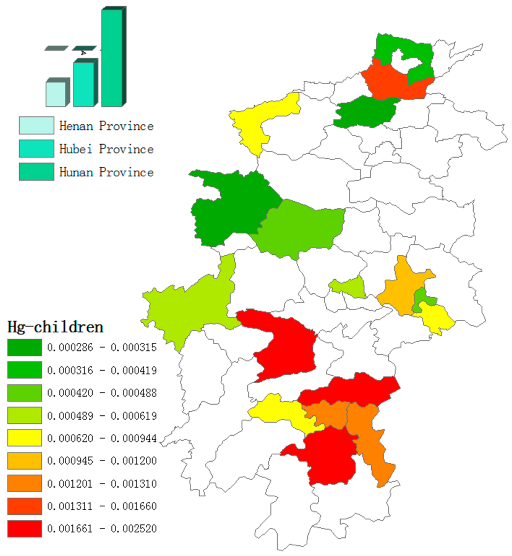
Figure 4. *Cont.*



(i)



(j)



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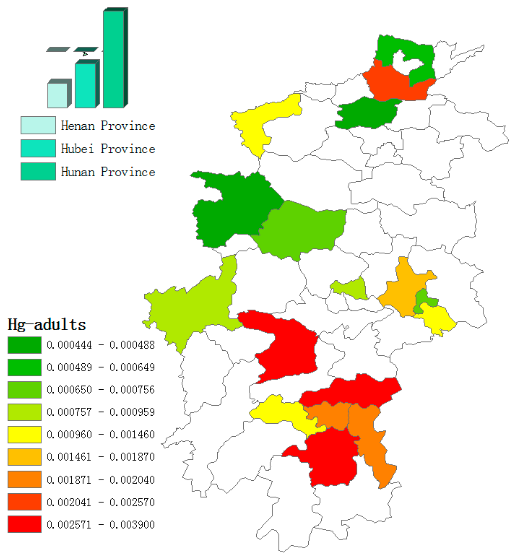

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Figure 4. *Cont.*

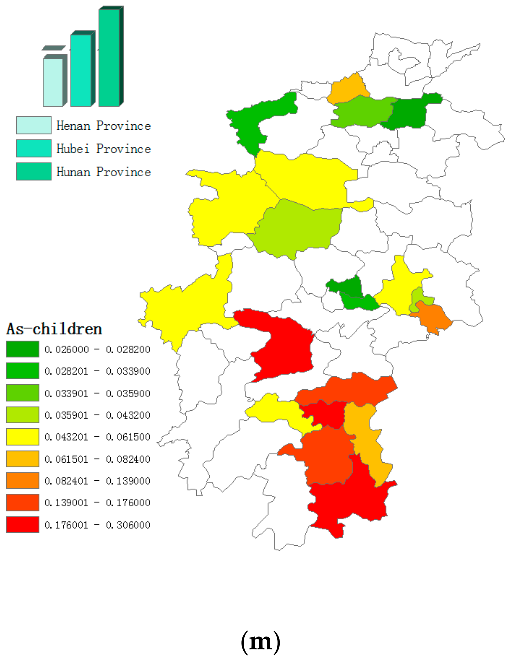

Figure 4. Non-carcinogenic hazard indices of Cu, Cr, Cd, Zn, Pb, Hg and As for children (**a**,**c**,**e**,**g**,**i**,**k**,**m**) and adults (**b**,**d**,**f**,**h**,**j**,**l**,**n**) in soils from Central China.

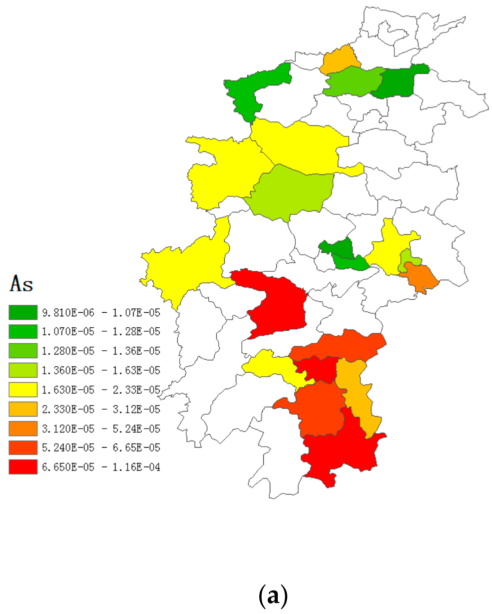

Figure 5. Carcinogenic risk of As for children (**a**) and adults (**b**) in soils from Central China.

For carcinogenic risk, the average carcinogenic risk values for children in Central China were 2.55×10^{-4} (Hubei), 3.44×10^{-4} (Hunan), 1.69×10^{-4} (Henan), respectively, and the average carcinogenic risk values for adults were 3.67×10^{-5} (Hubei), 4.92×10^{-5} (Hunan), 2.45×10^{-5} (Henan), respectively, which mean children and adults faced unacceptable and acceptable carcinogenic risk in Central China, respectively, shown in Table 6 and Figure 5. The results show that As appeared to be the main metal for both children and adults causing the high carcinogenic risk, followed by Cr, and the carcinogenic risks of Cd and Cr were generally within the internationally accepted precautionary criterion (1.0×10^{-6}), which was similar to other reports [101]. The carcinogenic risk of As for both children and adults in Central China decreased in the following order: ingestion > dermal contact > inhalation [130], which was similar to the non-carcinogenic risk. Therefore, based on the results of the geoaccumulation indexes and health risks for the studied metals, the carcinogenic risk levels of toxic metals in Central China were unacceptable, and As should be regarded as a priority control pollutant, especially for children.

Table 6. Carcinogenic risk (unitless) of soil metals in Central China.

Elements	Province	Children				Adults			
		CRing	CRinh	CRdermal	CR	CRing	CRinh	CRdermal	CR
Cr	Henan	—	9.71×10^{-8}	—	9.71×10^{-8}	—	7.15×10^{-7}	—	7.15×10^{-7}
	Hubei	—	1.14×10^{-7}	—	1.14×10^{-7}	—	8.41×10^{-7}	—	8.41×10^{-7}
	Hunan	—	1.29×10^{-7}	—	1.29×10^{-7}	—	9.51×10^{-7}	—	9.51×10^{-7}
Cd	Henan	—	4.39×10^{-9}	—	4.39×10^{-9}	—	3.24×10^{-9}	—	3.24×10^{-9}
	Hubei	—	1.69×10^{-9}	—	1.69×10^{-9}	—	1.25×10^{-9}	—	1.25×10^{-9}
	Hunan	—	3.13×10^{-9}	—	3.13×10^{-9}	—	2.30×10^{-9}	—	2.30×10^{-9}
As	Henan	1.68×10^{-4}	4.73×10^{-8}	1.00×10^{-6}	1.69×10^{-4}	2.35×10^{-5}	3.48×10^{-8}	2.04×10^{-7}	2.38×10^{-5}
	Hubei	2.53×10^{-4}	7.13×10^{-8}	1.51×10^{-6}	2.55×10^{-4}	3.55×10^{-5}	5.25×10^{-8}	3.07×10^{-7}	3.58×10^{-5}
	Hunan	3.41×10^{-4}	9.60×10^{-8}	2.04×10^{-6}	3.44×10^{-4}	4.78×10^{-5}	7.07×10^{-8}	4.14×10^{-7}	4.83×10^{-5}

3.5. Correlation Analysis

There is a certain correlation between the content of soil metals due to the similarity of geochemical conditions and the coexistence of pollutant metal elements. If the correlation between the elements is significant, it means that the elements are generally homologous or complex pollution [131–133]. Pearson's correlation coefficients of soil metals in Central China were performed, and the results are shown in Table 7. The element pairs Zn-Cd, Zn-Pb, Zn-As and As-Pb had a significantly positive correlation at $p < 0.01$ significance level, and the element pair As-Cd had a significantly positive correlation at $p < 0.05$ significance level, which means there was high possibility that the Zn, Cd, As and Pb came from a common source [126,128]. The abundant mineral resources and rapid development of optoelectronic, metallurgical and alloy manufacturing, have had a great impact on the contents of Cd, Zn, As and Pb [134–136]. Hg is a kind of volatile heavy metal, and it has been found that the released amount of Hg from the combustion industry accounts for about 60% of the total amount of Hg in the air and it becomes a soil pollutant after atmospheric settlement [137]; this is responsible for the high concentration in Central China. Cr has the lowest pollution level of heavy metals in the soil, 1.36 times the background value, generally considered by the impact of geochemistry, which is similar to other reports [80,138,139].

Table 7. Pearson correlation matrix for toxic metal concentrations.

	Cu	Cr	Cd	Zn	Pb	Hg	As
Cu	1						
Cr	−0.073	1					
Cd	0.130	0.124	1				
Zn	0.146	0.102	0.840 **	1			
Pb	0.112	−0.091	0.153	0.515 **	1		
Hg	−0.064	0.121	−0.088	−0.059	−0.062	1	
As	0.227	−0.324	0.535 *	0.716 **	0.639 **	−0.119	1

* Correlation is significant at the 0.05 level (two-tailed); ** correlation is significant at the 0.01 level (two-tailed).

3.6. Sustainable Management

Based on the above analysis of the spatial characteristics and the health risk assessment, there are four suggestions for Central China's sustainable development. First of all, the priority control soil metals need to be given enough attention. According to comparisons with the environmental quality standard for soils in Central China, the mean concentration of Cd was 2.18 times higher than its corresponding class II value, which is hazardous to agricultural production and corresponding human health. Thus, Cd should be selected as a priority control heavy metal. Based on the pollution assessment using the geoaccumulation index, the soils in Central China have been polluted by Cd and Hg, which identify Cd and Hg. Ultimately, people are most exposed to Cr, Pb and As, among all the investigated toxic metals. In conclusion, Cd, Hg, Cr, Pb and As are selected as the priority control soil metals in Central China, to which special attention should be paid in order to target the lowest threats to human health.

Second, importance should be attached to sources control. As for Henan Province, more importance should be attached to Cr in Jiaozuo city and Cd in Xinxiang city. In Hubei Province, more attention should be paid to Cu in Huangshi city, and Pb in Jingmen city. In Hunan Province, more attention should be paid to Zn, Pb and As in Chenzhou city, Cd and Zn in Xiangtan city, Hg in Changsha city and As in Changde city. At the same time, we recommend that decision-makers pay attention to the remediation of the highly-contaminated soil, especially in the areas where it could endanger the groundwater. For example, Huangshi city and Changsha city should be concerned, not only due to the high values of Cu and Hg, but also because the Yangtze River and the Xiang River flow there.

Third, the secondary industry ratio in central China is too high at present, causing serious pollution. Therefore, it is necessary to appropriately adjust the industrial structure to increase the tertiary service industry, promote entrepreneurship and employment, and reasonably optimize urban planning. Furthermore, the three provinces should engage in more cooperation to form a complete economic zone.

Moreover, publicity and education for sustainable development should be given due attention, to popularize a series of issues such as environmental pollution, health risks and sustainable development with the media by means of television, radio, the Internet and so on, so that the public will participate more actively.

4. Conclusions

In this study, data on soil metals in Central China were collected, with a comprehensive description and a systematic evaluation performed. According to the pollution assessment, it is apparent that the concentration of soil metals in Central China, especially Cd of 1.31 mg/kg and Hg of 0.19 mg/kg, surpass their corresponding background values, indicating anthropogenic input. The concentrations of toxic metals were higher in Hunan and Henan Province, the southern and northern parts of Central China, contrasting with the lowest contents in the middle parts. For non-carcinogenic risk, the children in Hubei Province (HI = 1.10) and Hunan Province (HI = 1.41) faced non-carcinogenic risks, with higher health risks to children than adults. The exposure pathway that resulted in the highest levels of exposure risk for children and adults was ingestion, followed by dermal contact and inhalation. For carcinogenic risk, children and adults faced unacceptable and acceptable carcinogenic risks in Central China, respectively, and arsenic (As) appeared to be the main metal for both children and adults causing the high carcinogenic risk. For sustainable development in Central China, special attention should be paid to Cd, Hg, Cr, Pb and As, which were identified as the priority control soil metals. Importance should also be attached to public education, source control, and remediation of highly-contaminated soil, especially in the areas where it can endanger the groundwater. Furthermore, it is necessary to appropriately adjust the industrial structure and cooperate more to form a complete economic zone.

Supplementary Materials: The following are available online at www.mdpi.com/2071-1050/10/1/91/s1, Table S1: Percentages (unitless) of class distribution for pollution assessment of soil metals in Central China using geoaccumulation index.

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Author Contributions: Fei Li organized this study, conducted the study design, and drafted the manuscript. Ying Cai contributed to the study design, prepared datasets, performed the statistical analysis, and drafted the manuscript. Jingdong Zhang contributed to study design, interpretation of analysis, and revision of the manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

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