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RESEARCH ARTICLE

Speciation, sources, and risk assessment of heavy metals in suburban vegetable garden soil in Xianyang City, Northwest China

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Abstract Intensive anthropogenic activities can lead to soil heavy metal contamination resulting in potential risks to the environment and to human health. To reveal the concentrations, speciation, sources, pollution level, and ecological risk of heavy metals in vegetable garden soil, a total of 136 soil samples were collected from three vegetable production fields in the suburbs of Xianyang City, Northwest China. These samples were analyzed by inductively coupled plasma- atomic emission spectrometry and atomic fluorescence spectrometry. The results showed that the mean concentrations of Cd, Co, Cu, Mn, Pb, Zn, and Hg in vegetable garden soil were higher than the corresponding soil element background values of Shaanxi Province. The heavy metals studied in vegetable garden soil were primarily found in the residual fraction, averaging from 31.26% (Pb) to 90.23% (Cr). Considering the non-residual fractions, the mobility or potential risk was in the order of Pb (68.74%) > Co (60.54%) > Mn (59.28%) > Cd (53.54%) ≫ Ni (23.36%) > Zn (22.73%) > Cu (14.93%) > V (11.81%) > Cr (9.78%). Cr, Mn, Ni, V, and As in the studied soil were related to soil-forming parent materials, while Cu, Hg, Zn, Cd, Co, and Pb were associated with the application of plastic films, fertilizers, and pesticides, as well as traffic emissions and industrial fumes. Cr, Ni, V, and As presented low contamination levels, whereas Co, Cu, Mn, Pb, and Zn levels were moderate, and Cd and Hg were high. Ecological risk was low for Co, Cr, Cu, Mn, Pb, Zn, and As, with high risk observed for Cd and Hg. The overall pollution level and ecological risk of these heavy metals were high.

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1 Introduction

Soil is a major component of the earth's system, and is vital as a nutrient provider for plant growth (Chen et al., 1997; Lu et al., 2007). Thus, the quality of the soil environment plays an important role in crop growth and resident health. However, a large number of toxic and harmful substances (e.g., heavy metals, polycyclic aromatic hydrocarbons, and phthalate esters) are being released into the soil environment due to anthropogenic activities (e.g., industrial and agricultural production, transportation, mining, and smelting). Contaminants accumulated in soil can enter water bodies from precipitation and surface runoff, the atmosphere through volatilization, plants via root adsorption, and the human body through food, as well as ingestion, inhalation, and dermal adsorption of soil dust particles. These soil contaminant pathways pose potential ecological and health risks to the environment and human beings (Sun et al., 2010; Wang et al., 2016). These evidences show that soil has become a sink of contaminants and a transmitter of pollutants into the atmosphere, groundwater, and plants (Chen et al., 1997, 2008; Nicholson et al., 2003; Sun et al., 2010; Yang et al., 2011).

Heavy metals are called the 'chemical time bombs' of soil pollutants (Stigliani et al., 1991); an important environmental issue because of their strong toxicity, non-biodegradability, and bioavailability (Chen et al., 2009). Vegetable production fields are usually located in the suburbs of cities in China and are subjected to anthropogenic pollution, such as atmospheric deposition, waste disposal, and the application of fertilizers and pesticides (Cai et al., 2005; Cui et al., 2015). Heavy metal contamination of suburban soil has increased over the last few decades due to rapid industrialization and urbanization (Cui et al., 2015). Numerous studies on heavy metal contamination in agricultural soil have been conducted in several provinces of China, such as Shandong (Liu et al., 2011; Tian et al., 2016), Jiangsu (Huang et al., 2007; Zhao et al., 2007; Chen et al., 2014; Cui et al., 2015; Tian et al., 2016), Zhejiang (Chen et al., 2008, 2009; Xu et al., 2014; Ye et al., 2015), and Guangdong (Cai et al., 2015). However, these studies have been confined to the rapidly developing regions in east and south China, with limited information from other regions. At the same time, the toxicity, environmental behavior, mobility, and bioavailability of heavy metals in the environmental media depend not only on the total concentrations of heavy metals, but also their physicochemical speciation (Li and Thornton, 2001; Wong et al., 2002; Lu et al., 2003, 2007; Rodríguez et al., 2009; Liu et al., 2013; Li et al., 2015; Sungur et al., 2015; Yang et al., 2015; Botsou et al., 2016).

Xianyang, a medium-sized city with a population of 0.5 million, is located in the center of Shaanxi Province, Northwest China. The urbanization, industrialization, and agricultural modernization of Xianyang progressed rapidly due to China's Western Development Program and Integration of Xi'an (the capital of Shaanxi province) and Xianyang. Vegetable production in Xianyang amounts to one quarter of the total area of vegetable production in Shaanxi Province. Large vegetable production fields are also located in the suburbs located to the south, southwest, and southeast of Xianyang, bordering Xi'an. Potential soil pollution sources include the application of agrochemicals, in addition to those generated by traffic and industries. Due to the limited studies on heavy metal contamination in this area, the focus of this study is to examine the concentrations and speciation of heavy metals in vegetable garden soil, in addition to identifying the environmental sources, and assessing the pollution level and ecological risk.

2 Materials and methods

2.1 Sample collection

Three vegetable production fields were selected for this study in the suburban areas of Xianyang City: 1) Dongzhangcun in the vicinity of the Xibao highway, 2) Caojiazhai along the Xibao highway, and 3) Baxingtan surrounded by highways and railways (Fig. 1). Vegetables have been produced in Dongzhangcun, Caojiazhai, and Baxingtan for 20–40 years, with 37, 65, and 34 soil sampling sites randomly arranged, respectively. Five subsamples (0–25 cm) of topsoil were collected at each sampling site from the four corners and center of a 2 m × 2 m grid, using a stainless steel shovel, and mixed into a composite sample of 1 kg in September and October of 2013. Each sample was stored in a self-sealing plastic bag and taken back to the laboratory. All the collected topsoil samples were air-dried in a cool, dark, and ventilated area at room temperature. The air-dried topsoil samples were crushed and passed through a 1 mm stainless steel sieve to remove stones, plant debris, and other refuse. Half of each sample was ground with a vibration mill to analyze the concentration and speciation of heavy metals. The other half was stored prior to the physicochemical analysis of soil.

2.2 Physiochemical analysis of soil

Soil pH and electrical conductivity (EC) were measured in a 1 : 2.5 (w : v) ratio of soil to distilled water with a pH meter and a conductivity meter (Lu et al., 2007). Soil organic matter (SOM) was measured by loss-on-ignition (LOI). Samples were oven-dried at 105°C, weighted, and then ignited in a muffle furnace at 550°C for 4 h before being re-weighted to determine the loss of organic matter (Irabien and Velasco, 1999). The particle-size composition of clay (< 2 μm), silt (2–50 μm), and sand (50–1000 μm) in soil was determined by a Mastersizer-2000 laser size analyzer (Lu et al., 2009). Magnetic susceptibility (χ) is proportional to the concentration of ferrimagnetic minerals within a sample and sensitive to magnetic grain size (Lu and Bai, 2006). The low and high frequency magnetic susceptibilities (χ_{LF} and χ_{HF}) of soil were measured on 10 g samples with a Bartington MS-2 magnetic susceptibility meter at the frequencies of 0.47 kHz and 4.7 kHz, respectively (Lu et al., 2003; Lu et al., 2009). The frequency-dependent susceptibility (χ_{FD}) indicates the presence of super paramagnetic (SP) grain (Lu and Bai, 2006). The frequency-dependent magnetic susceptibility (χ_{FD}) of soil was calculated by Eq. (1):

$$\chi_{FD} = \frac{\chi_{LF} - \chi_{HF}}{\chi_{LF}} \times 100\%. \tag{1}$$

2.3 Analysis for concentration and speciation of heavy metals

The total concentration and speciation of cobalt (Co), chromium (Cr), copper (Cu), cadmium (Cd), manganese (Mn), nickel (Ni), lead (Pb), vanadium (V), and zinc (Zn) in the soil samples were determined using an Arcos inductively coupled plasma-atomic emission spectrometer (ICP-AES, Spectro Analytical Instruments, Inc., Germany). To find the total concentration of heavy metals, a 0.5 g ground soil sample was placed into a 50-mL polytetrafluorethylene crucible and a small amount of ultrapure water was added to wet it. After adding 5 mL of concentrated hydrogen nitrate (HNO_3), 5 mL of concentrated hydrofluoric acid (HF), and 3 mL of concentrated perchloric acid ($HClO_4$) to each sample, they were heated

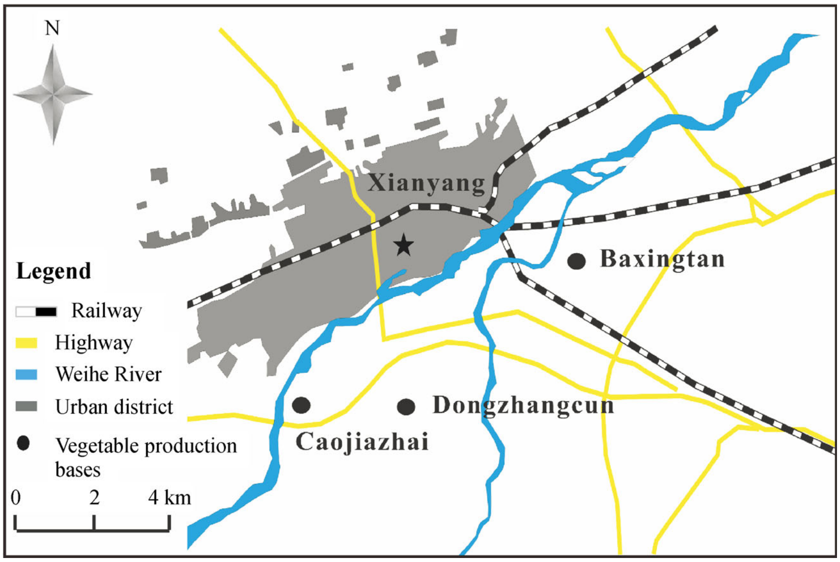

Fig. 1 Location of the study area in the suburb of Xianyang City of China.

until dry. The samples were then cooled, followed by the addition of 3 mL HNO_3, 3 mL HF, and 1 mL $HClO_4$, and reheated until dry. The residuals were dissolved in an HNO_3 solution (v : v = 1 : 1), transferred to a 50-mL volumetric flask, and diluted with additional HNO_3 solution. The modified BCR (European Community Bureau of Reference) sequential extraction procedure, as described in our previous work (Wang et al., 2015), was utilized to fractionate heavy metals into four forms: acid extractable, reducible, oxidizable, and residual (Lu et al., 2007; Janoš et al., 2010; Liu et al., 2013; Botsou et al., 2016). The digested and extracted solutions were analyzed using the ICP-AES under a plasma power of 1350 W, pump speed of 30 rpm, coolant flow of 12.00 L/min, auxiliary flow of 1.20 L/min, nebulizer flow of 0.81 L/min, and carrier gas of highly pure argon. The analytical lines of Co, Cr, Cu, Cd, Mn, Ni, Pb, V, and Zn were selected to be at (230.786, 283.565, 224.700, 228.802, 257.611, 231.604, 168.215, 292.402, and 202.613) nm, respectively.

The total concentrations of arsenic (As) and mercury (Hg) in soil samples were detected using a 9700 atomic fluorescence spectrometer (AFS, Beijing Haiguang Instrument Co., Ltd, China). Ground soil (0.1 g) was placed into a 50-mL glass colorimetric tube and a small amount of ultrapure water was added to wet it. After adding 10 mL of a mixed solution of concentrated HCl and HNO_3 (v : v = 1 : 1), each sample was heated for 3 h in a water bath at 100°C. After cooling, each was diluted to 50 mL with ultrapure water. 5 mL of each acid digest was placed in a 10-mL plastic sample cup, followed by the addition of 1 mL of a mixed solution of ascorbic acid (5%) and sulfourea (5%). To determine the concentrations of As and Hg within the acid digests, the AFS was operated under an arsenic hollow cathode lamp current of 50 mA, mercury hollow cathode lamp current of 20 mA, photomultiplier negative high-voltage of 260 V, carrier gas (argon) flow of 400 mL/min, shielding gas (argon) flow of 900 mL/min, atomizer temperature of 200°C, atomizer height of 8.0 mm, read time of 10 s, delay time of 1 s, read mode of peak area, and injection volume of 1.5 mL.

Glassware and plasticware used in the experiments were first soaked in a 5% HNO_3 solution for 24 h, and then rinsed with tap water, deionized water, and ultrapure water, respectively. The HCl, HNO_3, and $HClO_4$ were guaranteed reagents, while other chemicals, including ascorbic acid, sulfourea, and HF, were analytical reagents. Standard stock solutions of As and Hg were provided by the National Steel and Iron Analytical Center of China. A mixed standard stock solution of heavy metals was purchased from Inorganic Ventures (USA). Chinese soil standard reference substances (GSS-1 and GSS-2) were used to check the accuracy of analytical methods. Blank experiments were also conducted and 10% of the soil samples were analyzed in duplicate. For total concentration of heavy metals, the recovery defined as the ratio of measured value to standard value ranged from 84% to 114%. The recovery in the speciation of heavy metals was defined as the ratio of the sum of the four forms to the total concentration of each heavy metal, varying between 80% and 119%. The relative standard deviation (RSD) of the results for the duplicate experiments was below 8%.

2.4 Source identification

Principal component analysis (PCA) and cluster analysis (CA) were performed to identify the relationships and possible sources of heavy metals in vegetable garden soil, using SPSS version 19.0 for Windows. PCA extracts a few independent factors (principal components, PCs). Each PC may represent a specific pollution source. CA classifies the observations into two or more mutually exclusive unknown groups (clusters, Cs) based on a combination of internal variables. The members of each group can have the same or similar pollution source. CA is often coupled with PCA to crosscheck (Chen et al., 2008; Lu et al., 2010).

2.5 Pollution assessment

Single pollution index (*SPI*) and integrated pollution index (*IPI*) were used in this study to assess the pollution level of heavy metals in vegetable garden soil (Chen et al., 2005). The *SPI* is defined as the ratio of heavy metal concentration in the present study to the background concentration of heavy metal in soil of Shaanxi Province (China National Environmental Monitoring Center, 1990). *SPI* levels are classified as low at $SPI \leqslant 1$, moderate $1 < SPI \leqslant 3$, and high at $SPI > 3$. The *IPI* is defined as the mean of the *SPI* values for individual heavy metals in a sample. *IPI* levels are classified as low, moderate, or high at $IPI \leqslant 1$, $1 < IPI \leqslant 2$, and $IPI > 2$, respectively (Chen et al., 2005).

2.6 Ecological risk assessment

This study used the potential ecological risk index (*RI*) to evaluate the ecological risk of heavy metals in vegetable garden soil (Håkanson et al., 1980). *RI* is calculated as:

$$RI = \sum E_i = \sum T_i \times \frac{C_i}{B_i}, \quad (2)$$

where, C_i is the measured concentration of heavy metal i, B_i is the corresponding background concentration of heavy metal i in soil of Shaanxi Province (China National Environmental Monitoring Center, 1990), T_i is the toxicity coefficient of heavy metal i, E_i is the single ecological risk index of metal i, and *RI* is the total ecological risk index. T_i of Zn, Cr, Cu, Pb, Ni, As, Cd, and Hg is 1, 2, 5, 5, 5, 10, 30, and 40, respectively (Håkanson et al., 1980; Xu et al., 2008). The E_i and *RI* values were classified with a system described by Ma et al. (2011), i.e., $E_i < 40$ indicates low risk, $40 \leqslant E_i < 80$ moderate, $80 \leqslant E_i < 160$ higher, $160 \leqslant E_i < 320$ high, and $E_i \geqslant 320$ severe; $RI < 138$ indicates low risk, $138 \leqslant RI < 276$ moderate, $276 \leqslant RI < 552$ high, and $RI \geqslant 552$ severe.

3 Results and discussion

3.1 Physiochemical properties of vegetable garden soil

The descriptive statistics of physiochemical parameters of the vegetable garden soil from the suburbs of Xianyang City, Northwest China are given in Table 1. As also shown

Table 1 Physiochemical properties of vegetable garden soil from the suburbs of Xianyang City

Items	Min	Max	Mean	SD	CV
pH	6.98	8.25	7.60	0.31	0.04
EC/($\mu S \cdot cm^{-1}$)	3.4	1902.0	459.1	312.80	0.68
LOI/%	3.53	8.00	4.76	0.64	0.13
Clay/%	3.11	9.78	6.29	1.48	0.24
Slit/%	39.76	73.64	61.06	7.25	0.12
Sand/%	16.59	57.13	32.65	8.58	0.26
χ_{LF} /($10^{-8} m^3 \cdot kg^{-1}$)	53.3	139.9	88.2	13.07	0.15
χ_{HF} /($10^{-8} m^3 \cdot kg^{-1}$)	51.3	137.5	83.2	12.17	0.15
χ_{FD} /%	1.29	9.80	5.64	1.86	0.33

SD: standard deviation; CV: coefficient of variation.

in Table 1, the pH values of the vegetable garden soil ranged from 6.98 to 8.25 with a mean of 7.60, indicating a slightly alkaline soil. The EC was in the range of 3.4–1902.0 μS/cm with an average of 459.1 μS/cm, presenting a large variation. The contents of SOM varied from 3.53% to 8.00% with a mean of 4.76%. The particle-size distribution of the vegetable garden soil was 6.29% clay, 61.06% silt, and 32.65% sand. The low-frequency magnetic susceptibility (χ_{LF}) ranged from (53.3 to 139.9) $\times 10^{-8} m^3 \cdot kg^{-1}$ with a mean of 88.2 $\times 10^{-8} m^3 \cdot kg^{-1}$. The high-frequency magnetic susceptibility (χ_{HF}) varied from (51.3 to 137.5) $\times 10^{-8} m^3 \cdot kg^{-1}$ with a mean of 83.2 $\times 10^{-8} m^3 \cdot kg^{-1}$. The magnetic susceptibility values were lower than those in agricultural soil around the Pb-Zn smelting plant of Baoji and indicated relatively low levels of ferromagnetic mineral (Lu and Bai, 2006; Wang et al., 2015). The frequency dependent susceptibility (χ_{FD}) ranged from 1.29% to 9.80% with an average of 5.64%, indicating the presence of super paramagnetic particles (Lu and Bai, 2006).

3.2 Concentration of heavy metals in vegetable garden soil

As shown in Table 2, all analyzed heavy metals were detected in the vegetable garden soil samples taken from the suburbs of Xianyang City, Northwest China. The average concentrations of Cd, Co, Cr, Cu, Mn, Ni, Pb, V, Zn, As, and Hg were (0.78, 17.10, 55.36, 25.52, 611.07, 25.33, 39.44, 66.16, 190.54, 8.35, and 0.13) mg/kg, respectively. Compared to the Chinese Environmental Quality Standard for Soil (State Environmental Protection Administration of China, 1995), the concentrations of Cd and Zn in 64 and 26 soil samples, respectively, exceeded the critical values of grade two, indicating safe levels for agricultural production and human health. The mean concentrations of Cr, Ni, V, and As were below the corresponding soil element background values of Shaanxi Province (China National Environmental Monitoring

Table 2 Concentrations of heavy metals in vegetable garden soil from the suburbs of Xianyang City (mg/kg)

Elements		Cd	Co	Cr	Cu	Mn	Ni	Pb	V	Zn	As	Hg
All sampling sites (n = 136)	Min	0.20	10.45	31.30	15.65	461.35	16.00	24.25	39.50	40.35	3.48	0.01
	Max	2.45	30.80	70.30	49.50	833.90	33.00	132.55	84.00	525.75	11.89	0.54
	Mean	0.78	17.10	55.36	25.52	611.07	25.33	39.44	66.16	190.54	8.35	0.13
	SD	0.50	3.23	8.12	5.69	89.06	4.13	16.49	9.72	111.36	1.41	0.10
	CV	0.65	0.19	0.15	0.22	0.15	0.16	0.42	0.15	0.58	0.17	0.77
Baxingtan (n = 34)	Min	0.80	10.45	31.30	16.45	461.35	16.00	35.10	46.25	48.85	5.47	0.06
	Max	2.45	25.35	65.65	49.50	608.80	26.45	132.55	61.00	364.25	9.54	0.50
	Mean	1.23	17.45	48.72	26.58	530.22	19.63	51.39	52.14	163.26	6.83	0.16
	SD	0.36	3.44	7.15	9.39	37.81	2.44	19.86	3.72	85.02	1.02	0.09
	CV	0.29	0.20	0.15	0.35	0.07	0.12	0.39	0.07	0.52	0.15	0.55
Caojiaozhai (n = 65)	Min	0.20	11.10	34.15	15.65	491.70	18.90	24.25	39.50	40.35	3.48	0.01
	Max	1.65	30.80	70.30	37.35	684.25	33.00	43.00	77.35	456.70	11.29	0.33
	Mean	0.72	16.97	54.75	23.19	582.82	26.10	32.31	68.25	179.09	8.90	0.09
	SD	0.50	3.69	6.99	2.81	33.31	2.24	4.79	5.27	107.20	1.16	0.08
	CV	0.70	0.22	0.13	0.12	0.06	0.09	0.15	0.08	0.60	0.13	0.89
Dongzhangcun (n = 37)	Min	0.20	13.95	52.20	24.55	568.40	26.00	26.80	67.90	74.25	6.40	0.01
	Max	1.50	21.90	70.05	33.05	833.90	32.95	102.05	84.00	525.75	11.89	0.54
	Mean	0.47	17.00	62.53	28.62	735.00	29.20	40.97	75.35	235.71	8.78	0.17
	SD	0.31	1.95	4.31	2.45	55.34	1.62	19.70	3.43	128.10	1.10	0.12
	CV	0.66	0.11	0.07	0.09	0.08	0.06	0.48	0.05	0.54	0.13	0.69
SEBVS		0.094	10.6	62.5	21.4	557	28.8	21.4	66.9	69.4	11.1	0.030
CEQSS	pH < 6.5	0.30		150	50		40	250		200	40	0.30
	6.5 < pH < 7.5	0.30		200	100		50	300		250	30	0.50
	pH > 7.5	0.60		250	100		60	350		300	25	1.0

SD: standard deviation; CV: coefficient of variation; SEBVS: Soil element background values of Shaanxi (China National Environmental Monitoring Center, 1990); CEQSS: Chinese environmental quality standard for soil (State Environmental Protection Administration of China, 1995).

Center, 1990). Low coefficients of variation (CV) values indicated natural inputs of Cr, Ni, V, and As (Lu et al., 2010; Wang et al., 2014). Even though the average concentrations of Co, Cu, and Mn were slightly higher (1.2–1.6 times) than the corresponding soil element background values of Shaanxi Province (China National Environmental Monitoring Center, 1990), the CV values were relatively low, suggesting mixed sources of natural and anthropogenic inputs of Co, Cu, and Mn (Lu et al., 2010; Wang et al., 2014). The concentrations of Cd, Pb, Zn, and Hg were higher (1.8–8.3 times) than the corresponding soil element background values of Shaanxi Province (China National Environmental Monitoring Center, 1990) with relatively larger CV values, implying anthropogenic inputs of Cd, Pb, Zn, and Hg (Lu et al., 2010; Wang et al., 2014). Elevated heavy metals in the present study were probably related to the relatively high pH, SOM, and slit in the vegetable garden soil.

The concentration comparisons of heavy metals in the vegetable garden soil from the suburbs of Xianyang City, Northwest China and in vegetable garden/agricultural soils from other regions are given in Table 3. Table 3 also shows that the average concentrations of Cd, Co, Mn, and Zn in the present study were higher than those in the vegetable garden or agricultural soils from other areas of China. The mean concentrations of Cd and Mn were lower in our study than those in the agricultural soil from Turkey, with the average value of Co lower than that in the agricultural soil from Greece. These findings indicate that metal content levels were higher in the studied vegetable garden soil. The mean concentrations of other heavy metals (except for V, which had no comparable data) were in the same ranges as reported for different regions, indicating moderate metal content levels in the studied soil. Research by Divrikli et al. (2003) found the highest concentration of heavy metals (i.e., Cd, Fe, Co, Mn, Bi, Pb, Ni, Zn, and Cu) in spinach relative to lettuce and cabbage, even though they easily accumulate in the roots of lettuce and cabbage. This poses a potential health risk to the people of China given both spinach and cabbage are common dietary foods. The concentrations of heavy metals varied across the three vegetable production fields (Table 2) with mean concen-

Table 3 Concentration comparison of heavy metals in vegetable garden/agricultural soil from different areas

Regions	Soil types	Cd	Co	Cr	Cu	Mn	Ni	Pb	Zn	As	Hg	References
Xianyang, Shaanxi, China	Vegetable soil	0.78	17.1	55.36	25.52	611.07	25.33	39.44	190.54	8.35	0.13	In this study
Shandong, China	Vegetable soil	0.15	NA	41.78	29.28	NA	28.00	16.18	82.26	9.02	0.09	Liu et al. (2011)
Hangzhou, Zhejiang, China	Vegetable soil	0.26	9.25	62.23	36.57	415.27	20.04	46.15	116.07	6.25	0.69	Chen et al. (2008)
Hangzhou, Zhejiang, China	Vegetable soil	0.193	9.10	NA	21.34	393.65	20.28	NA	81.11	NA	0.187	Chen et al. (2009)
Hangzhou, Zhejiang, China	Vegetable soil	0.39	NA	81.4	NA	NA	NA	30.3	NA	7.57	0.42	Ye et al. (2015)
Jiaxing, Zhejiang, China	Vegetable soil	0.26	NA	82.7	NA	NA	NA	29.5	NA	7.89	0.42	Ye et al. (2015)
Ningbo, Zhejiang, China	Vegetable soil	0.34	NA	82.90	NA	NA	NA	30.10	NA	7.60	0.42	Ye et al. (2015)
Shaoxing, Zhejiang, China	Vegetable soil	0.34	NA	82.60	NA	NA	NA	33.60	NA	7.36	0.38	Ye et al. (2015)
Taizhou, Zhejiang, China	Vegetable soil	0.21	NA	82.80	NA	NA	NA	27.40	NA	6.39	0.30	Ye et al. (2015)
Wuxi, Jiangsu, China	Vegetable soil	0.143	NA	58.6	40.4	NA	NA	46.7	112.9	14.3	0.161	Zhao et al. (2007)
Baguazhou Island, Nanjing, Jiangsu, China	Vegetable soil	0.314	NA	133	41.0	NA	58.0	31.8	114	NA	NA	Cui et al. (2015)
Nanjing, Jiangsu, China	Conventional greenhouse soil	0.19	NA	NA	35.67	NA	NA	37.87	97.05	9.58	0.27	Chen et al. (2014)
	Organic greenhouse soil	0.27	NA	NA	37.13	NA	NA	26.5	94.07	6.21	0.08	Chen et al. (2014)
Shouguang, Shandong, China	Solar greenhouse soil	0.196	NA	67.13	27.86	NA	28.93	20.06	115.79	6.88	0.044	Tian et al. (2016)
Dongtai, Jiangsu, China	Plastic greenhouse soil	0.161	NA	49.33	14.87	NA	21.3	17.58	56.63	5.42	0.031	Tian et al. (2016)
Yangzhou, Jiangsu, China	Agricultural soil	0.3	NA	77.2	33.9	NA	38.5	35.7	98.1	10.2	0.2	Huang et al. (2007)
Jiaxing, Zhejiang, China	Agricultural soil	0.221	NA	87.8	32.4	NA	36.4	33.9	94.9	8.55	0.199	Xu et al. (2014)
Dehui, Changchun, Jilin, China	Agricultural soil	NA	NA	49.7	18.9	NA	20.8	35.4	58.9	NA	NA	Sun et al. (2013)
Shunde, Guangzhou, China	Agricultural soil	0.60	16.76	78.87	NA	NA	33.45	NA	NA	16.08	0.38	Cai et al. (2015)
Shunyi, Beijing, China	Agricultural soil	0.136	NA	NA	22.4	NA	NA	20.4	69.8	7.85	0.073	Lu et al. (2012)
Taiyuan, Shanxi, China	Agricultural soil	0.25	NA	74.10	32.11	NA	29.74	27.87	90.76	10.7	0.09	Liu et al. (2015)
Canakkale, Turkey	Agricultural soil	1.48	9.08	43.38	22.50	629.8	61.11	19.66	41.24	NA	NA	Sungur et al. (2015)
Argolida basin, Greece	Agricultural soil	NA	21.99	83.12	74.68	1020.5	146.8	19.74	74.88	6.95	NA	Kelepertzis (2014)

NA: no available data.

trations following the order of: Dongzhangcun > Caojiazhai > Baxingtan for Cr, Mn, Ni, V and Zn; Baxingtan > Dongzhangcun > Caojiazhai for Co and Pb; Dongzhangcun > Baxingtan > Caojiazhai for Cu and Hg; Baxingtan > Caojiazhai > Dongzhangcun for Cd; and Caojiazhai > Dongzhangcun > Baoxingtan for As. The differences in the distribution patterns of heavy metals were likely associated with the differences in the application of agricultural production materials, such as agricultural plastic films and fertilizers. Traffic patterns for the distance traveled between the vegetable fields and the urban areas are also likely to have an effect.

3.3 Speciation of heavy metals in vegetable garden soil

Figure 2 shows the speciation of heavy metals in the vegetable garden soil from the suburbs of Xianyang City, Northwest China and their predominance in the residual fraction. This partitioning pattern indicates that a significant fraction of heavy metals, averaging from 31.26% (Pb) to 90.23% (Cr), is relatively immobile under normal environmental conditions. The present findings are consistent with the results reported by Botsou et al. (2016).

For the non-residual fractions, Cd was dominated by the acid extractable and reducible forms, averaging 27.96% and 23.09% of total concentration; Co, Cu, Mn, Ni, V, and Zn were controlled by the reducible form which comprised 44.52%, 9.66%, 37.47%, 11.18%, 7.22%, and 17.17% of total concentrations, respectively; Cr and Pb were associated with the oxidizable form, accounting for 7.01% and 41.08% of total concentrations, respectively. Heavy metals in the acid-soluble forms are generally considered readily and potentially mobile. The reducible and oxidizable forms are relatively stable under normal soil conditions. Heavy metals in the residual fraction are entrapped within the crystal structure and the minerals, representing the least mobile fraction. Therefore, the sum of the first three fractions may reflect the mobility and potential risk of heavy metals. As shown in Fig. 2, the order of mobility or potential risk of heavy metals was Pb (68.74%) > Co (60.54%) > Mn (59.28%) > Cd (53.54%) >> Ni (23.36%) > Zn (22.73%) > Cu (14.93%) > V (11.81%) > Cr (9.78%). Among these heavy metals, Cd, Co, Mn, and Pb had a relatively stronger mobility or higher potential risk.

The ratio of the acid extractable fraction to total concentration can be used as an assessment index to evaluate the availability of heavy metals in soil or sediment (Botsou et al., 2016). The soil with < 1% acid extractable fraction within total concentration is considered as non-risk for the environment, 1%–10% is low risk, 11%–30% medium risk, 31%–50% high, and over 51% an extreme risk. In the present study, the percentage of the acid extractable forms to total concentrations for Cr and V in the studied soil was only 0.60% and 0.40%, respectively, implying a non-risk for the environment. The ratios of the acid extractable fractions to total concentrations for Cu, Ni, and Zn were 2.09%, 3.93%, and 4.83%, respectively, indicating a low risk for the environment. Alternatively, the values for Cd, Co, Mn, and Pb were 27.96%, 13.37%, 19.24%, and 15.37%, respectively, suggesting a medium risk for the environment.

3.4 Possible sources of heavy metals

PCA with the varimax rotation of Kaiser Standardization (the test values of KMO and Bartlett were 0.78 and 951.93, respectively) extracted three principal components (PC1, PC2, and PC3) with eigenvalues > 1 from the heavy metals studied, explaining 67.14% of total variance (Table 4). PC1 had high loading for Cr, Mn, Ni, V, and As, accounting for 37.69% of total variance. PC2 was loaded by Cu, Hg, and Zn, responsible for 15.33% of total variance. PC3 was composed of Cd, Co, and Pb, explaining 14.15% of total variance. CA separated the heavy metals studied into three clusters (C1, C2, and C3)

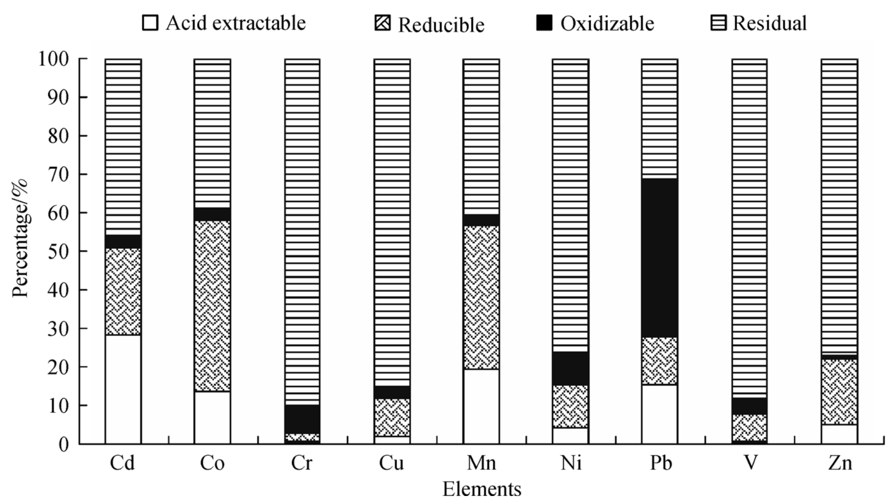

Fig. 2 Speciation of heavy metals in the vegetable garden soil.

Table 4 Results of principal component analysis of heavy metals

Elements	Factor			Community
	1	2	3	
Cd	−0.39	0.06	**0.72**	0.67
Co	0.08	−0.14	**0.73**	0.55
Cr	**0.83**	0.34	−0.07	0.81
Cu	0.34	**0.78**	0.17	0.74
Mn	**0.82**	0.24	0.01	0.74
Ni	**0.96**	0.07	−0.14	0.95
Pb	−0.22	0.50	**0.56**	0.61
V	**0.95**	−0.03	−0.13	0.91
Zn	0.35	**0.57**	0.29	0.21
As	**0.71**	−0.10	−0.09	0.53
Hg	0.01	**0.79**	−0.20	0.66
Eigenvalue	4.14	1.69	1.56	
Variance/%	37.67	15.32	14.15	
Cumulative variance/%	37.67	52.99	67.14	

Factor loading values of > 0.50 are shown in bold.

(Fig. 3). C1 consisted of Ni, V, Cr, Mn, and As; C2 of Cd, Pb, and Co; and C3 of Cu, Hg, and Zn. Meanwhile, C2 and C3 formed a new cluster at a higher level. The results of CA were consistent with PCA.

The concentrations of heavy metals in PC1 and C1 were lower than the local soil element background values (except for Mn) where the CV was relatively smaller. Therefore, PC1 and C1 represented soil-forming parent materials. The concentrations of heavy metals in PC2/C3 and PC3/C2 were higher than the soil element background values with relatively large CV values, thus PC2/C3 and PC3/C2 were mainly related to anthropogenic activities. On one hand, the agricultural plastic films, pesticides, and fertilizers were widely applied for vegetable production based on our field investigations. These agrochemicals contain heavy metals, such as Cu, Pb, Zn, Cd, As, and Hg (Nicholson et al., 2003; Chen et al., 2008; Liu et al., 2011). For example, Cd and Pb heat stabilizers are applied to agricultural plastic films during production. Pesticides may also contain heavy metals, such as Cu, Pb, and Hg (Yaylalı-Abanuz, 2011). Inorganic phosphate fertilizer may contain Cd because the raw material phosphorite is rich in Cd (Williams and David, 1973; Yaylalı-Abanuz, 2011). Livestock manure contains Zn and Cu derived from feed additives (Nicholson et al., 2003; Yaylalı-Abanuz, 2011). Alternatively, these heavy metals are possibly related to traffic emissions and industrial fumes given the close proximity of vegetable production fields to urban areas and highways. Marin et al. (1997) found a significant correlation between the concentrations of soil heavy metals (e.g., Cr, Cu, Zn, Pb, Co, Bi, and Ni) and traffic intensity. Therefore, PC2/C3 and PC3/C2 denoted the agrochemical application, along with traffic and industrial emissions.

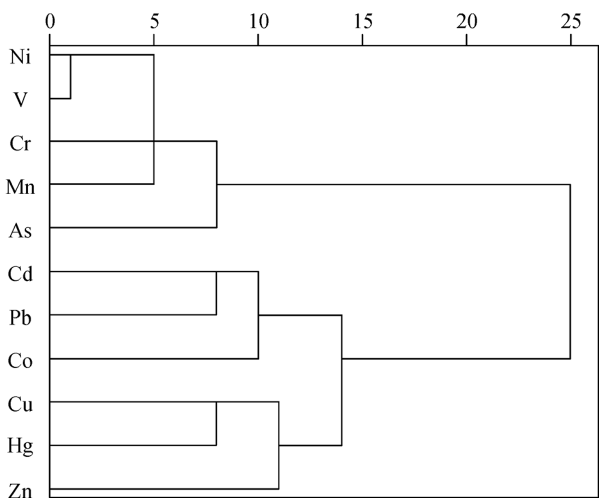

Fig. 3 Results of cluster analysis of heavy metals.

3.5 Contamination levels from heavy metals

The assessment results of contamination levels from heavy metals in vegetable garden soil are provided in Table 5. As shown, the average values of the single pollution index (*SPI*) for Cr, Ni, V, and As were less than 1, indicating overall low pollution levels. However, the *SPI* values reached moderate pollution levels in 25 soil samples for Cr, 26 for Ni, 79 for V, and 4 for As. The mean SPI values for Co, Cu, Mn, Pb, and Zn ranged from 1 to 3, also suggesting overall moderate pollution. The pollution levels were high in 11 and 48 soil samples for Pb and Zn, respectively. High concentrations of Cd and Hg and lower soil background values resulted in increased pollution levels (China National Environmental Monitoring Center, 1990). The values of the integrated pollution index (*IPI*)

Table 5 Pollution indices of heavy metals in the study area for 136 vegetable garden soil samples

Indices	Elements	Min	Max	Mean	SD	CV
SPI	Cd	2.13	26.06	8.28	5.37	0.65
	Co	0.99	2.91	1.61	0.30	0.19
	Cr	0.50	1.12	0.89	0.13	0.15
	Cu	0.73	2.31	1.19	0.27	0.22
	Mn	0.83	1.50	1.10	0.16	0.15
	Ni	0.56	1.15	0.88	0.14	0.16
	Pb	1.13	6.19	1.84	0.77	0.42
	V	0.59	1.26	0.99	0.15	0.15
	Zn	0.58	7.58	2.75	1.60	0.58
	As	0.31	1.07	0.75	0.13	0.17
	Hg	0.21	18.06	4.41	3.40	0.77
IPI		1.14	4.21	2.24	0.62	0.28

SD: standard deviation; CV: coefficient of variation.

ranged from 1.14 to 4.21, representing moderate to high pollution levels. Pollution levels of heavy metals were moderate in 52 samples and high in 84 samples, indicating an overall high pollution level.

3.6 Ecological risk of heavy metals

As shown in Table 6, the values of a single ecological risk index (E_i) for Co, Cr, Cu, Mn, Pb, Zn, and As were below 40 in all the soil samples, suggesting a low ecological risk. The E_i of Hg varied between 8.20 and 722.53, representing a low to severe ecological risk. Meanwhile, the ecological risk of Hg was low, moderate, higher, high, and severe in 19, 19, 35, 44, and 19 soil samples, respectively, with an overall high ecological risk. The E_i of Cd ranged from 63.83 to 781.91, indicating a moderate to severe ecological risk. The ecological risk of Cd was moderate, higher, high, and severe in 15, 54, 15, and 42 soil samples, respectively, with an overall high ecological risk. The higher ecological risk of Hg and Cd was primarily related to the higher toxicity coefficient (T_i) and lower soil background values (China National Environmental Monitoring Center, 1990).

The total ecological risk index (RI) of the heavy metals ranged from 124.07 to 1169.57, with a mean of 465.32, implying that there was high overall ecological risk in the vegetable garden soil.

Table 6 Ecological risk indices of heavy metals in the study area with 136 vegetable garden soil samples

Indices	Elements	Min	Max	Mean	SD	CV
E_i	Cd	63.83	781.91	248.28	161.14	0.65
	Co	4.93	14.53	8.06	1.52	0.19
	Cr	1.00	2.25	1.77	0.26	0.15
	Cu	3.66	11.57	5.96	1.33	0.22
	Mn	0.83	1.50	1.10	0.16	0.15
	Ni	2.78	5.73	4.40	0.72	0.16
	Pb	5.67	30.97	9.21	3.85	0.42
	Zn	0.58	7.58	2.75	1.60	0.58
	As	3.13	10.71	7.52	1.27	0.17
	Hg	8.20	722.53	176.26	135.80	0.77
RI		124.07	1169.57	465.32	205.37	0.44

SD: standard deviation; CV: coefficient of variation.

4 Conclusions

Soil contamination from heavy metals is an environmental concern of significant importance. In the present study, the soil for growing vegetables in the suburbs of Xianyang City, Northwest China contained high concentrations of heavy metals, especially Cd, Pb, Zn, and Hg, exceeding the corresponding soil element background values of Shaanxi Province. The heavy metals studied in vegetable garden soil were primarily found in the residual fraction, averaging from 31.26% (Pb) to 90.23% (Cr). The mobility or potential risk of the non-residual fractions was in the order of Pb (68.74%) > Co (60.54%) > Mn (59.28%) > Cd (53.54%) >> Ni (23.36%) > Zn (22.73%) > Cu (14.93%) > V (11.81%) > Cr (9.78%). The ratio of the acid extractable fraction to total concentration indicated that Cu, Ni, and Zn were low risk for the environment, whereas Cd, Co, Mn, and Pb presented a medium risk. Cr, Mn, Ni, V, and As originated from soil-forming parent materials, while Cu, Hg, Zn, Cd, Co, and Pb were related to the application of plastic films and fertilizers as well as traffic emissions and industrial fumes. Soil contamination was reported as low from Cr, Ni, V, and As, moderate from Co, Cu, Mn, Pb, and Zn, and high from Cd and Hg. The overall pollution level of heavy metals was high. Co, Cr, Cu, Mn, Pb, Zn, and As posed a low ecological risk, while the risk from Cd and Hg was severe. The total ecological risk from heavy metals was high.

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