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RESEARCH ARTICLE

Occurrence, speciation, and risks of trace metals in soils of greenhouse vegetable production from the vicinity of industrial areas in the Yangtze River Delta, China

Lanqin Yang[1] · Guoming Liu[2] · Lin Di[3] · Xiangyang Wu[1] · Wenhua You[1] · Biao Huang[2]

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Abstract

The effect of industrial activities on trace metals in farmland of rapidly industrializing regions in developing countries has increasingly been a concern to the public. Here, soils were collected from 13 greenhouse vegetable production (GVP) farms or bases near industrial areas in the Yangtze River Delta of China to investigate the occurrence, speciation, and risks of Cr, Cu, Zn, Cd, Ni, and Pb in GVP soil. The results revealed that the main metal elements causing GVP soil pollution were Cd, Zn, Ni, and Cu, of which contamination levels were generally unpolluted to moderately polluted. Zinc pollution was mainly attributed to heavy fertilization, while Cd, Ni, and Cu pollution may be greatly ascribed to industrial effluents and coal combustion. Metal speciation studies showed that most of Cr, Ni, Cu, and Zn was present in residual fraction while more than half of Cd and Pb was present in non-residual fractions. Additionally, pollution of Cd, Cu, Ni, and Zn in GVP soil increased their corresponding mobile fractions. Risk assessment using potential ecological risk index and risk assessment code showed that Cd was the major risk contributor. Specifically, Cd generally posed moderate or considerable ecological risk as well as displayed medium or high mobility risk in GVP soil. Thus, great attention should be paid to the contribution of both industrial discharges and intensive farming to soil pollution by trace metals, especially Cd, because of its high mobility risk.

Keywords Trace metals · Greenhouse vegetable production soil · Industrial areas · BCR procedure · Risk assessment

Introduction

Driven by increasing food demands and economic incentives, greenhouse vegetable production (GVP) has rapidly expanded worldwide, especially in developing countries (Critten and Bailey 2002; Huang and Jin 2008; Hickman 2011; Chang et al. 2013). The statistics in Hickman (2011) showed that GVP accounted for approximately 20% of global total vegetable cultivation area. In China, the GVP area has reached more than 4.1 million hm^2 by the end of 2014 (Tian et al. 2016). The Yangtze River Delta region is one of the major GVP areas in China. For example, by the end of 2014, the GVP area in the Jiangsu Province of the Yangtze River Delta region has reached about 0.76 million hm^2, which accounted for 16.5% of the total arable land in the Jiangsu Province in 2014 (Xia et al. 2017).

However, some previous domestic and abroad studies have reported that accumulation of trace metals, especially Cd, Cu, and Zn, occurred in GVP soil primarily due to heavy fertilization (Gil et al. 2004; Rodriguez Martin et al. 2013; Bai et al. 2015; Zhang et al. 2017). Meanwhile, the Yangtze River Delta is one of the regions in China to experience rapid industrialization in recent several decades. Thus, in this region, many factories were built and scattered around the agricultural lands. Some previous studies conducted in this region have

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✉ Lanqin Yang
yanglanqinujs@ujs.edu.cn

✉ Biao Huang
bhuang@issas.ac.cn

[1] School of the Environment and Safety Engineering, Jiangsu University, 212013 Zhenjiang, People's Republic of China

[2] Key Laboratory of Soil Environment and Pollution Remediation, Institute of Soil Science, Chinese Academy of Sciences, 210008 Nanjing, People's Republic of China

[3] Zhenjiang Station of Farmland Quality Protection, 212009 Zhenjiang, People's Republic of China

reported that irrigation using sewage from the industries, such as electroplating and textile, resulted in soil contamination by Cd, Cr, Cu, Pb, Zn, and Ni (Hang et al. 2009; Zhong et al. 2011). Therefore, the occurrence of trace metals in GVP soil from the vicinity of industrial areas in the Yangtze River Delta requires major concern.

Despite the fact that the total concentration of soil trace metal is a useful indicator for assessing the contamination and potential ecological risk (Zhang et al. 2009; Tian et al. 2017), the mobility and bioavailability of trace metals as well as their eco-toxicity to plants were greatly associated with their chemical forms rather than their total concentrations (Jin et al. 2005; Zhong et al. 2011). The sequential extraction procedures, such as the BCR sequential extraction procedures proposed by the European Community Bureau of Reference (Ure et al. 1993), are widely used to separate trace metals into four forms, including acid-soluble fraction of water-soluble, exchangeable, and carbonate-bound metals, reducible fraction of Fe–Mn oxide-bound metals, oxidizable fraction of organic matter-bound metals, and residual fraction. Among these fractions, the acid-soluble fraction is loosely bound to trace metals and considered as mobile and bioavailable fraction; reducible and oxidizable fractions are potentially bioavailable fractions; residual fraction is defined as non-bioavailable fraction (Devi and Saroha 2014; Nannoni and Protano 2016). Thus, investigating the fractions of trace metals, particularly in contaminated soil, is necessary and would be beneficial for assessing the risks of trace metal release.

Overall, although considerable researches have been conducted for assessment of trace metals in GVP soil (Gil et al. 2004; Rodriguez Martin et al. 2013; Yang et al. 2013; Bai et al. 2015; Xu et al. 2015; Tian et al. 2017; Zhang et al. 2017), most of these studies focused on the investigation of trace metals in peri-urban GVP soil. The study on the occurrence, speciation, and risks of trace metals in GVP soil near industrial areas is still lacking. Therefore, the objectives of the present study were to (a) evaluate the occurrence of Cr, Ni, Cu, Zn, Pb, and Cd in GVP soil collected from the vicinity of industrial areas in the Yangtze River Delta using greenhouse soil quality standards (GSQS) of China (State Environmental Protection Administration of China (SEPAC) 2006) and geo-accumulation index; (b) determine the fractional concentrations and percentages of trace metals in GVP soil using BCR sequential extraction method; and (c) assess potential ecological risk and mobility risk of trace metals in GVP soil. The results of this study may provide some insight into the synergistic effect of industrial activities and intensive farming on trace metals in soil. Additionally, the data of this study can serve as a basis for comparison with trace metals and risks of their release in GVP soil of other rapidly industrializing regions both in China and other developing countries.

Materials and methods

Study area

Ten farms and three greenhouse vegetable bases, located in six typical industrial towns or sub-districts of the Yangtze River Delta, were selected as a case study area (Fig. 1). These towns or sub-district included Xiashu Town, Xinfeng Town, Xinba Town, Economic development zone of Yangzhong City, and Sanmao and Xinglong Sub-districts. The GVP area in these farms and vegetable bases varied within 0.27 and 5.57 hm^2 (Table S1). The durations of GVP generally ranged from 2 to 10 years. Around the study areas, there were a lot of industries, including electric power, building material, bearing production, textile, electronics, electroplating, plastics, metal product, electrical engineering, and hardware manufacturing (Fig. 1 and Table S1). Topographically, the study sites in Xiashu Town and Xinfeng Town are gently undulating, and soils are mainly comprised of Argosols developed from loess parent material (Huang and Pan 2017). The other study sites, however, are plains, and soils are mainly composed of Anthrosols derived from calcareous deposits of the Yangtze River (Huang and Pan 2017).

Sample collection and treatment

A total of 31 topsoil samples (0–20 cm) were sampled from greenhouse vegetable plots using a stainless steel auger throughout the study farms and bases (Fig. 1 and Table S1). Meanwhile, 19 topsoil samples were collected from open fields near greenhouses (Table S2). Each soil sample was comprised of five sub-samples. All the samples were put in polyethylene sampling bags and brought back to the lab for further treatment and analysis. After air drying at room temperature, each soil sample was divided into three parts. The part ground to pass a 10-mesh sieve was for determining pH and cation exchange capacity (CEC); the part ground to pass a 100-mesh sieve was for measuring organic matter (OM) and total metal contents; and the other part ground to pass a 200-mesh sieve was for speciation analysis of trace metals.

Sample analysis

Soil pH was determined in a 1:2.5 soil–water suspension using a glass electrode pH meter (Model PHS-3C, Shanghai Precision and Scientific Instrument Co. Ltd.). Soil OM content was determined following the $K_2Cr_2O_7$–H_2SO_4 digestion method (Nelson and Sommers 1982). Soil CEC was detected by an EDTA ammonium acetate method (The National Agro-Tech Extension and Service Center (NATESC) 2006). Soil total Cr, Cd, Ni, Cu, Pb, and Zn contents were measured using

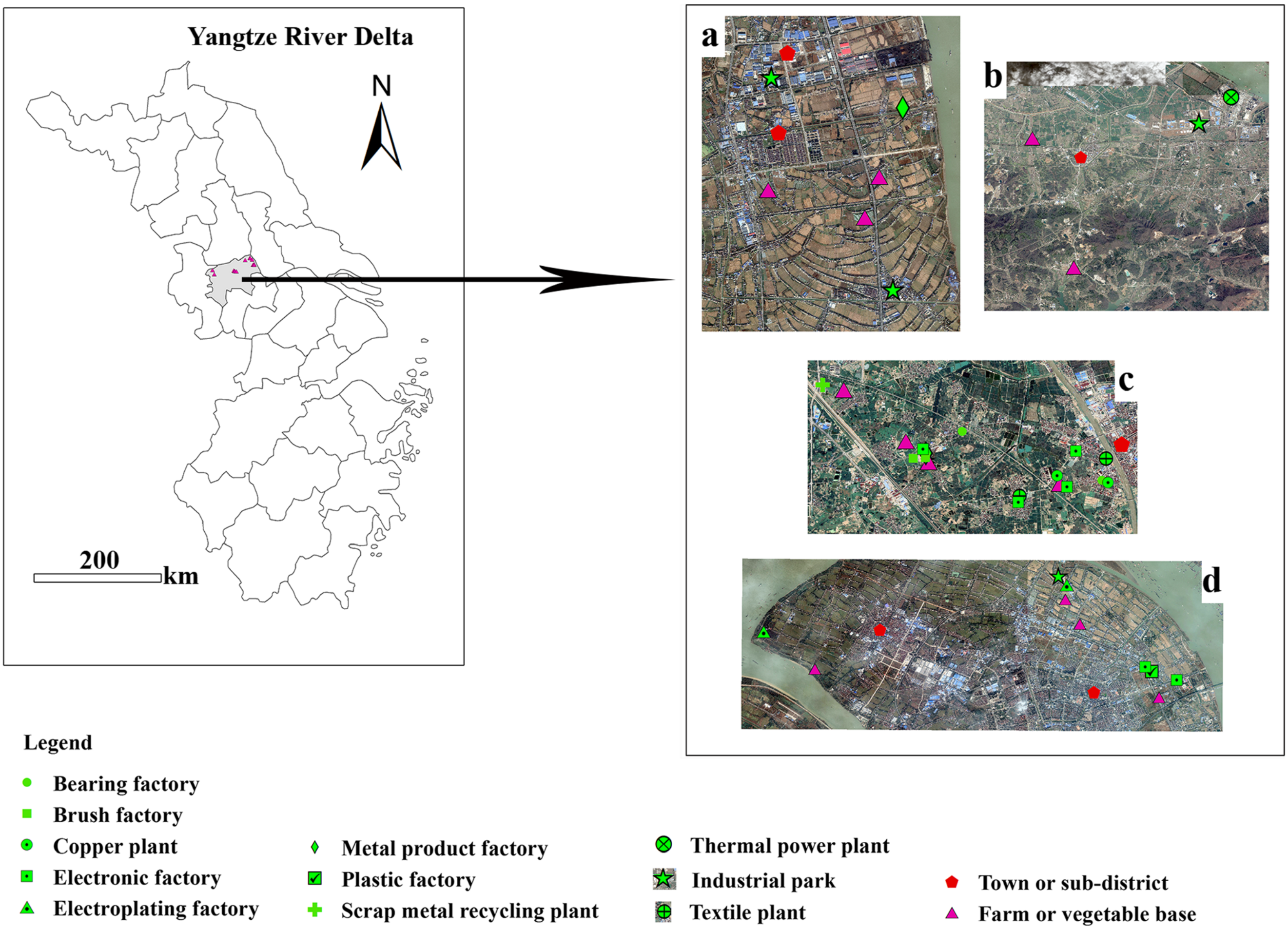


Fig. 1 Location of the selected farms and vegetable bases near industrial areas in six towns or sub-districts, including Xinglong Sub-district and Economic development zone of Yangzhong City (**a**), Xiashu Town (**b**), Xinfeng Town (**c**), and Xinba Town and Sanmao Sub-district (**d**) in the Yangtze River Delta, China

inductively coupled plasma mass spectrometry (ICP-MS) after digestion with $HClO_4$, HNO_3, and HF. Fractions of trace metals in soil were extracted using BCR sequential extraction method (Ure et al. 1993; Rauret et al. 1999) and determined by ICP-MS. Specifically, the fractions were defined as follows: (1) acid-soluble fraction (F1, 1.0 g of soil sample extracted by 40 ml 0.11 M acetic acid for 16 h at 22 ± 5 °C); (2) reducible fraction (F2, the residue from the former step extracted by 40 ml 0.5 M hydroxylammonium chloride for 16 h at 22 ± 5 °C); (3) oxidizable fraction (F3, the residue from the former step digested with 10 ml + 10 ml 30% hydrogen peroxide (pH 2-3) at room temperature for 1 h and at 85 ± 2 °C in a water bath for 2 h, respectively, after evaporation to about 1 ml extracted by 50 ml 1.0 M ammonium acetate, pH = 2.0 for 16 h at 22 ± 5 °C); (4) residual fraction (F4, determined by subtracting the three above-mentioned fractions from the corresponding total metal concentrations).

For quality control, analytical reagent blanks were performed with each batch of experiments. Additionally, soil certified reference materials (GSS-3) obtained from the China National Center for Standard Reference Materials were analyzed for assessing the accuracy of detecting total metal contents in soil. The results suggested that the detected values of Cr, Ni, Cd, Cu, Pb, and Zn in the reference materials met the accuracy requirements by comparing with the certified values. For checking the reliability of sequential extraction procedure, the residual metal concentrations in one fifth of all the soil samples were digested using HF, $HClO_4$, and HNO_3. The ratios of the sum of each metal concentration in individual fractions to the total metal concentration in soil ranged from 88.6 to 115.2%, suggesting that the results of sequential extractions were reliable. Furthermore, for evaluating the precision of the total metal concentration determination and the sequential extraction, analytical duplicates, which made up 10% of the entire set of samples, were also performed. The relative standard deviations for all the metals determined in the duplicate samples ranged from 0.009 to 18.5%, indicating that the results were acceptable.

Data processing

Geo-accumulation index

The geo-accumulation index originally introduced by Muller (1969) has been used as a common criterion for assessing trace metal pollution levels in sediments and agricultural soils (Howari and Banat 2001; Bhuiyan et al. 2010), which can be defined as follows:

$$I_{\mathrm{geo}} = \log_2\left(\frac{C_{\mathrm{n}}}{1.5B_{\mathrm{n}}}\right)$$

where C_{n} is the trace metal content in soil and B_{n} is the background value. The constant 1.5 is the background matrix correction factor associated with lithogenic effects (Wei and Yang 2010). Pollution levels on the basis of the I_{geo} can be classified as (I) unpolluted (≤ 0), (II) unpolluted to moderately polluted (0–1), (III) moderately polluted (1–2), (IV) moderately to heavily polluted (2–3), (V) heavily polluted (3–4), (VI) heavily to extremely polluted (4–5), and (VII) extremely polluted (> 5).

Potential ecological risk index

The potential ecological risk index (RI) proposed by Hakanson (1980) has been widely used for assessing the potential ecological risks of trace metals in sediments and soils (Bo et al. 2015; Saeedi and Jamshidi-Zanjani 2015; Tian et al. 2017), which can be calculated as follows:

$$C_{\mathrm{f}} = \frac{C_{\mathrm{n}}}{B_{\mathrm{n}}}$$

$$Er^i = Tr^i \times C_{\mathrm{f}}^i$$

$$RI = \sum_{i=1}^{n} Er^i$$

where C_{f}, C_{n}, and B_{n} denote the contamination factor for a given element, the trace metal content in soil, and the background value, respectively. Tr^i is the toxic response factor for a given trace metal element. The proposed values of Tr for Cd, Cu, Pb, Zn, Cr, and Ni are 30, 5, 5, 1, 2, and 5 (Hakanson 1980; Tian et al. 2017). Er^i represents the potential ecological risk factor for a given element. The Er^i and RI can be classified as (I) low potential ecological risk ($Er^i < 40$ and $RI < 150$), (II) moderate potential ecological risk ($40 \leq Er^i < 80$ and $150 \leq RI < 300$), (III) considerable potential ecological risk ($80 \leq Er^i <$ 160 and $300 \leq \mathrm{RI} < 600$), (IV) high potential ecological risk ($160 \leq Er^i < 320$), and (V) very high potential ecological risk ($Er^i \geq 320$ and $RI \geq 600$).

Risk assessment code

In order to evaluate the mobility and bioavailability of trace metals in soil, risk assessment code (RAC) was used in this study, which is defined as follows (Perin et al. 1985):

$$RAC_i = 100 \times \frac{C_{ex,i} + C_{carb,i}}{C_{t,i}}$$

where $C_{ex,i}$ is the concentration of metal i in exchangeable fraction, $C_{carb,i}$ is the concentration of metal i in carbonate fraction, and $C_{t,i}$ is the total concentration of metal i. In this study, the metal concentrations in acid-soluble fraction (F1) included the concentrations in exchangeable and carbonate fractions. The risk level classifications of the RAC are presented as follows: (I) no risk ($\mathrm{RAC} < 1$), (II) low risk ($1 \leq \mathrm{RAC} < 10$), (III) medium risk ($10 \leq \mathrm{RAC} < 30$), (IV) high risk ($30 \leq \mathrm{RAC} < 50$), and (V) very high risk ($\mathrm{RAC} \geq 50$).

However, RAC is not an aggregated index and does not take into account the toxicity of metals in assessing the risk. Thus, to consider both toxicity and aggregation of the polymetallic risk in an individual index, the modified risk assessment code (mRAC) was introduced and applied in this study as well. The mRAC can be calculated as follows (Saeedi and Jamshidi-Zanjani 2015):

$$mRAC = \frac{\sum_{i=1}^{n} Tr^i RAC_i}{\sum_{i=1}^{n} Tr^i}$$

where Tr^i is the toxic response factor, which has been mentioned in the "Potential ecological risk index" section. The risk level classifications of the mRAC are listed as follows: (I) no potential adverse effect ($\mathrm{mRAC} < 1$), (II) low potential adverse effect ($1 \leq \mathrm{mRAC} < 10$), (III) medium potential adverse effect ($10 \leq \mathrm{mRAC} < 30$), (IV) high potential adverse effect ($30 \leq \mathrm{mRAC} < 50$), and (V) very high potential adverse effect ($\mathrm{mRAC} \geq 50$).

Statistical analysis

Statistical analysis was performed by SPSS 19.0. The significant differences ($p \leq 0.05$) of soil pH, OM, CEC, total trace metal concentrations, and metal speciation in GVP soil and OF soil were determined by independent-sample t tests. Correlation analysis was used to determine the relationships between the concentrations or percentages of metals in acid-soluble fraction and other soil parameters, including soil pH, OM, CEC, and total metal concentrations.

Results

GVP soil pH, OM, CEC, and trace metal concentrations

As shown in Table 1, soil pH in greenhouse vegetable plots was significantly lower than that in open fields regardless of soil types in the study regions. In contrast, GVP soil CEC was found significantly higher than that in open field (OF) soil in the Argosols area. However, OM and trace metal concentrations of GVP and OF soils were similar in the study regions except for significantly higher Zn concentrations in GVP soil of the Anthrosols area. Compared with GSQS (State Environmental Protection Administration of China (SEPAC) 2006), Cr, Pb, and Zn concentrations in all GVP soil samples were within the maximum permissible limits, while 61.3, 3.2, and 9.7% of the samples were contaminated by Cd, Cu, and Ni, respectively.

Table 1 Soil pH, organic matter (OM), cation exchange capacity (CEC), and trace metal concentrations in greenhouse vegetable production and open field soil at different soil orders of the Yangtze River Delta, China

Parameters	pH	OM (g kg^{-1})	CEC (cmol kg^{-1})	Cd (mg kg^{-1})	Cu	Zn	Pb	Cr	Ni
Argosols									
Greenhouse soil (n = 13)									
Mean	4.88b[E]	22.50a	19.55a	0.20a	33.9a	102.4a	29.2a	64.8a	32.1a
SD[A]	0.77	6.66	3.79	0.10	18.6	33.7	6.5	10.7	11.4
Min	3.65	11.41	15.35	0.061	19.3	66.6	18.7	50.5	21.1
Max	6.05	33.96	28.71	0.42	92.8	181.7	38.6	88.3	64.1
Open field soil (n = 10)									
Mean	6.00a	21.71a	15.96b	0.20a	30.9a	85.9a	26.6a	58.5a	26.7a
SD	1.19	7.25	2.49	0.07	18.7	33.2	4.7	9.1	4.8
Min	3.96	13.36	13.03	0.11	19.4	52.4	17.7	47.9	21.6
Max	7.86	33.60	21.63	0.32	82.6	170.0	36.3	75.2	35.0
Background values[B]				0.094	29.0	69.0	24.5	84.0	38.0
Anthrosols									
Greenhouse soil (n = 18)									
Mean	7.13b	39.82a	16.50a	0.38a	40.7a	121.2a	32.2a	83.4a	39.9a
SD	0.23	27.45	4.03	0.06	6.7	14.5	2.2	9.2	5.1
Min	6.72	23.72	11.05	0.30	31.6	102.1	28.5	64.6	30.9
Max	7.59	147.67	30.85	0.48	50.9	156.7	35.8	98.4	48.4
Open field soil (n = 9)									
Mean	7.65a	35.13a	16.57a	0.35a	36.5a	108.0b	31.9a	79.1a	38.0a
SD	0.13	10.17	5.88	0.05	4.7	16.3	1.9	5.1	2.7
Min	7.48	24.39	10.30	0.28	31.8	92.8	29.9	74.6	34.8
Max	7.89	50.98	28.99	0.43	44.6	138.1	34.6	88.3	43.3
Background values[C]				0.20	32.1	79.4	26.7	80.9	33.4
GSQS[D]	< 6.5			0.30	50	200	50	150	40
	6.5–7.5			0.30	100	250	50	200	50
	> 7.5			0.40	100	300	50	250	60
% of samples above GSQS									
Argosols (n = 13)				15.4	7.7	0	0	0	23.1
Anthrosols (n = 18)				94.4	0	0	0	0	0
All (n = 31)				61.3	3.2	0	0	0	9.7

[A] Standard deviations

[B] Liao et al. (2011)

[C] Huang et al. (2006)

[D] SEPAC (2006)

[E] Numbers followed by the same lowercase letter in each column at each soil order represent no significant difference at $p \leq 0.05$

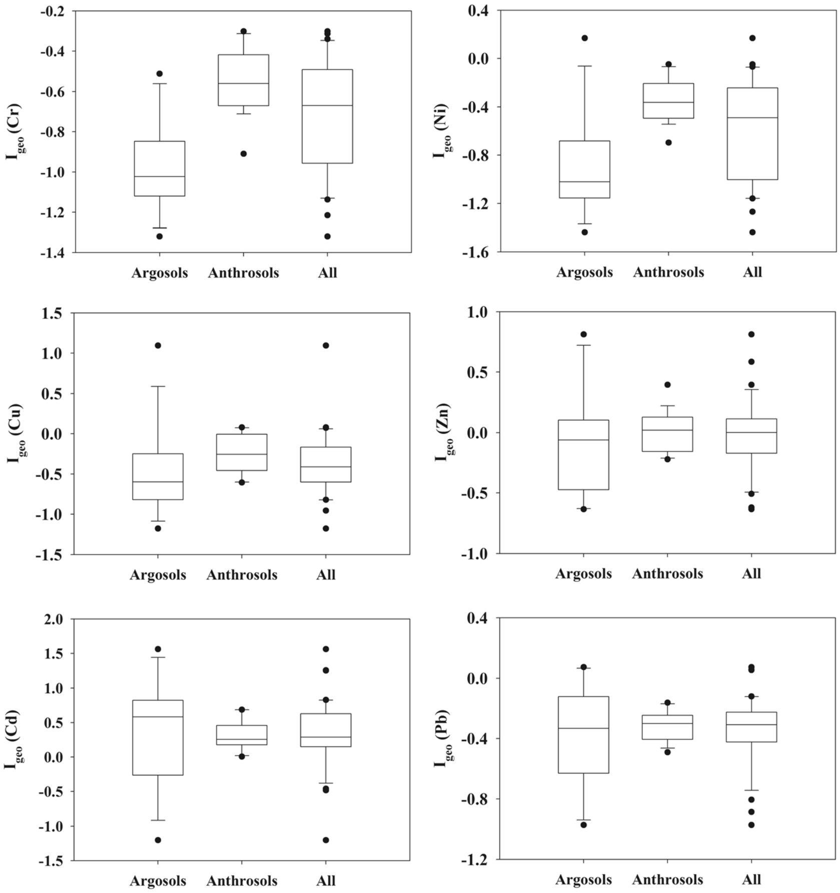
Geo-accumulation index of GVP soil trace metal concentrations

As can be seen from Fig. 2, the I_{geo} values of Cr, Ni, Cu, Zn, Cd, and Pb in GVP soil of the Argosols area ranged from − 1.32 to − 0.51, − 1.44 to 0.17, − 1.17 to 1.09, − 0.64 to 0.81, − 1.20 to 1.56, and − 0.97 to 0.07, respectively. Similarly, for Cr, Ni, Cu, Zn, Cd, and Pb, their I_{geo} values in GVP soil of the Anthrosols area varied from − 0.91 to − 0.30, − 0.70 to − 0.05, − 0.61 to 0.08, − 0.22 to 0.40, 0.01 to 0.69, and − 0.49 to − 0.16, respectively. According to the I_{geo} classification, there was no Cr pollution in all the GVP soils because the I_{geo} values of Cr were all below 0. In contrast, 87.1 and 51.6% of all the GVP soil samples had Cd and Zn I_{geo} values ranging from 0.01 to 1.56 and 0.00 to 0.81, respectively. Thus, above half of the GVP soils were unpolluted to moderately polluted by Cd and Zn. Similarly, 16.1, 6.5, and 3.2% of all the GVP soil samples had Cu, Pb, and Ni I_{geo} values varying within 0.02 and 1.09. This indicates that GVP soils at some sampling sites were slightly polluted by Cu, Pb, and Ni.

Fractional mass concentrations and percentages of trace metals in GVP soil

Trace metal concentrations in each fraction were similar in GVP and OF soils of the Argosols area (Table 2). However, there were significant differences between metal speciation in GVP and OF soil of the Anthrosols area. For example, acid-soluble Ni, Cd, and Pb concentrations as well as oxidizable Zn concentrations were significantly lower in GVP soil than those in OF soil in the Anthrosols study area. In contrast, acid-soluble Zn concentrations, reducible Ni and Cd concentrations, oxidizable Cd and Pb concentrations, as well as residual Cu and Zn concentrations were significantly higher in GVP soil than those in OF soil in the Anthrosols study area. Overall, irrespective of soil types in the study areas, trace metal concentrations in F1 + F2 + F3 showed no significant differences between GVP and OF soils (Table S3). Furthermore, Cr, Ni, Cu, and Zn in GVP soil had much higher concentrations in residual fraction than those in the other fractions in both Argosols and Anthrosols areas. As for Cd and Pb speciation in GVP soil, the residual Cd and Pb concentrations were the highest among their four fractions in the Argosols area while

Fig. 2 Geoaccumulation indices (I_{geo}) of trace metals in greenhouse vegetable production soil at different soil orders



reducible Cd and Pb concentrations were the highest in the Anthrosols area (Table 2).

In addition, the percentages of trace metals in each fraction were also similar in GVP and OF soils in the Argosols area. Nevertheless, the percentages of metal fractions showed significant differences between GVP and OF soils in the Anthrosols area. For example, percentages of acid-soluble Ni, Cd, and Pb, reducible Cu as well as percentages of oxidizable Zn were significantly lower in GVP soil than those in OF soil in the Anthrosols study area. In contrast, percentages of acid-soluble Zn, reducible Cd, oxidizable Cd and Pb, as well as percentages of residual Cu and Cd were significantly higher in GVP soil than those in OF soil in the Anthrosols study area (Table 3). In summary, the percentages of residual Cr, Ni, Cu, and Zn were generally above 50% while more than half of Cd and Pb were present in non-residual fractions (Table 3 and Table S3). Additionally, acid-soluble Cd fraction generally accounted for more than 20% of the total Cd concentrations in GVP soil (Table 3). The percentages of acid-soluble Cd fraction were much higher than those of acid-soluble Cr, Ni, Cu, Zn, and Pb fraction.

Potential ecological risks of trace metals in GVP soil

The Er^i values for Cr, Ni, Cu, Zn, and Pb in GVP soil were all below 40, indicating that low potential ecological risk was posed by Cr, Ni, Cu, Zn, and Pb in GVP soil of both the Argosols and Anthrosols areas (Table 4). Cadmium, however,

Table 2 Fractional mass concentrations (mg kg^{-1}) of trace metals in greenhouse vegetable production and open field soil

Trace metals	Argosols		Anthrosols	
	Greenhouse soil (SD^A)	Open field soil (SD)	Greenhouse soil (SD)	Open field soil (SD)
F1				
Cr	0.037 (0.013)a^B	0.038 (0.012)a	0.044 (0.007)a	0.047 (0.007)a
Ni	1.66 (1.77)a	0.99 (0.16)a	0.74 (0.18)b	0.89 (0.06)a
Cu	1.64 (4.15)a	1.37 (3.17)a	0.34 (0.13)a	0.36 (0.09)a
Zn	8.67 (11.48)a	8.29 (11.42)a	7.04 (3.63)a	4.12 (2.76)b
Cd	0.055 (0.027)a	0.064 (0.026)a	0.11 (0.02)b	0.16 (0.03)a
Pb	0.20 (0.27)a	0.19 (0.24)a	0.090 (0.046)b	0.23 (0.10)a
F2				
Cr	1.72 (0.54)a	1.60 (0.39)a	2.30 (0.30)a	2.38 (0.21)a
Ni	3.01 (2.65)a	2.64 (1.36)a	4.29 (0.43)a	3.99 (0.18)b
Cu	8.33 (7.72)a	8.16 (6.77)a	8.29 (1.96)a	9.23 (1.09)a
Zn	15.94 (6.08)a	14.90 (7.28)a	27.62 (6.04)a	25.27 (8.11)a
Cd	0.062 (0.041)a	0.066 (0.039)a	0.15 (0.04)a	0.11 (0.02)b
Pb	13.16 (4.15)a	12.36 (3.22)a	16.09 (1.26)a	16.46 (2.14)a
F3				
Cr	3.99 (0.82)a	3.63 (0.56)a	4.55 (0.64)a	4.14 (0.52)a
Ni	3.02 (2.44)a	2.11 (0.59)a	3.67 (0.34)a	3.44 (0.23)a
Cu	2.03 (2.44)a	1.61 (2.13)a	2.54 (1.46)a	1.94 (0.55)a
Zn	4.91 (4.91)a	2.81 (3.82)a	3.77 (1.36)b	5.72 (1.24)a
Cd	0.011 (0.008)a	0.010 (0.005)a	0.021 (0.006)a	0.014 (0.003)b
Pb	1.69 (0.49)a	1.50 (0.29)a	2.00 (0.41)a	1.58 (0.13)b
F4				
Cr	59.10 (9.84)a	53.23 (8.70)a	76.52 (9.08)a	72.56 (4.42)a
Ni	24.37 (5.30)a	20.93 (3.46)a	31.19 (5.11)a	29.68 (2.45)a
Cu	21.92 (5.34)a	19.81 (6.98)a	29.56 (6.01)a	24.92 (3.16)b
Zn	72.90 (21.08)a	59.85 (13.61)a	82.75 (12.61)a	72.85 (5.17)b
Cd	0.074 (0.030)a	0.064 (0.011)a	0.094 (0.034)a	0.070 (0.016)a
Pb	14.19 (2.31)a	12.53 (1.39)a	14.01 (1.18)a	13.62 (0.72)a

[A] Standard deviations

[B] Numbers followed by the same lowercase letter in each row at each soil order represent no significant difference at $p \leq 0.05$

Table 3 Fractional mass percentages (%) of trace metals in greenhouse vegetable production and open field soil

Trace metals	Argosols		Anthrosols	
	Greenhouse soil (SD[A])	Open field soil (SD)	Greenhouse soil (SD)	Open field soil (SD)
F1				
Cr	0.060 (0.025)a[B]	0.067 (0.024)a	0.054 (0.011)a	0.059 (0.010)a
Ni	4.66 (2.21)a	3.81 (0.86)a	1.89 (0.50)b	2.34 (0.18)a
Cu	2.78 (4.31)a	2.62 (3.56)a	0.85 (0.35)a	0.99 (0.21)a
Zn	7.57 (6.37)a	7.82 (6.42)a	5.72 (2.50)a	3.60 (1.81)b
Cd	27.31 (7.29)a	30.87 (6.26)a	30.61 (6.54)b	44.46 (3.78)a
Pb	0.62 (0.70)a	0.67 (0.71)a	0.28 (0.16)b	0.72 (0.34)a
F2				
Cr	2.61 (0.49)a	2.71 (0.36)a	2.79 (0.44)a	3.01 (0.13)a
Ni	8.38 (3.53)a	9.64 (4.17)a	10.99 (2.08)a	10.51 (0.41)a
Cu	22.42 (5.09)a	24.84 (3.72)a	20.62 (4.64)b	25.36 (1.19)a
Zn	15.71 (4.02)a	16.81 (5.04)a	22.84 (4.18)a	22.95 (3.73)a
Cd	28.05 (8.36)a	30.01 (9.14)a	39.31 (5.57)a	31.74 (3.54)b
Pb	44.06 (6.13)a	45.87 (5.03)a	49.98 (2.14)a	51.46 (3.92)a
F3				
Cr	6.21 (1.23)a	6.29 (1.16)a	5.51 (1.05)a	5.22 (0.39)a
Ni	8.62 (2.84)a	7.84 (0.94)a	9.28 (1.11)a	9.07 (0.59)a
Cu	5.12 (2.69)a	4.23 (1.90)a	6.35 (3.49)a	5.25 (0.84)a
Zn	4.21 (2.79)a	2.55 (2.25)a	3.16 (1.23)b	5.27 (0.40)a
Cd	5.26 (1.40)a	4.63 (1.05)a	5.44 (1.20)a	4.03 (0.82)b
Pb	5.80 (0.97)a	5.72 (1.03)a	6.21 (1.27)a	4.96 (0.42)b
F4				
Cr	91.12 (1.54)a	90.93 (1.07)a	91.65 (1.24)a	91.72 (0.47)a
Ni	78.33 (7.73)a	78.72 (4.69)a	77.84 (3.08)a	78.07 (1.00)a
Cu	69.67 (11.03)a	68.31 (8.26)a	72.18 (3.79)a	68.39 (1.34)b
Zn	72.51 (10.18)a	72.82 (11.01)a	68.29 (6.59)a	68.19 (5.69)a
Cd	39.38 (10.01)a	34.49 (13.09)a	24.64 (6.54)a	19.77 (3.82)b
Pb	49.52 (6.19)a	47.74 (4.56)a	43.52 (1.85)a	42.85 (3.72)a

[A] Standard deviations

[B] Numbers followed by the same lowercase letter in each row at each soil order represent no significant difference at $p \leq 0.05$

posed moderate potential ecological risk at all the GVP soil sampling sites in the Anthrosols area. Similarly, 61.5 and 15.4% of the GVP soil samples in the Argosols area had moderate and considerable potential ecological risk posed by Cd, respectively. In conclusion, over 90% of all the GVP soil samples in the study areas had moderate or considerable ecological risk posed by Cd.

Based on the classifications of potential ecological risks by RI values, there were 92.3 and 7.7% of GVP soils in the Argosols area with low and moderate risks, respectively, while all the GVP soils of the Anthrosols area were classified to have low ecological risk (Table 4). In conclusion, the RI values for more than 95% of all the GVP soil samples in the study areas were below 150 and indicated low ecological risk to the environment at these sampling sites.

Mobility risks of trace metals in GVP soil

The mean RAC values for Cr, Ni, Cu, Zn, Cd, and Pb in GVP soil in the Argosols area were 0.06, 4.66, 2.78, 7.57, 27.31, and 0.62, while those in the Anthrosols area were 0.05, 1.89, 0.85, 5.72, 30.61, and 0.28, respectively, indicating that RAC values for Cd were much higher than those for the other metals (Table 5). According to the classifications of potential metal release risk by RAC values, Cr displayed no mobility risk with RAC values in all GVP soils lower than 1. Similarly, the RAC values for Pb, Ni, Cu, and Zn at most of the GVP soil sampling sites in the study areas were below 10, suggesting no risk or low risk of Pb, Ni, Cu, and Zn release at these sites. However, the mobility risk level of Cd in GVP soils of the study areas was generally medium or high, since the RAC values for Cd varied within 10 and 50. Considering the risk

Table 4 Potential ecological risk levels of trace metals in greenhouse vegetable production soils. Numbers in parentheses are the standard deviations

Potential ecological risks	Er^i (SD[A])						RI (SD)
	Cr	Ni	Cu	Zn	Cd	Pb	
Argosols (*n* = 13)	1.54 (0.25)	4.22 (1.50)	5.85 (3.21)	1.48 (0.49)	64.58 (31.64)	5.97 (1.32)	83.64 (34.70)
% of low risk	100	100	100	100	23.08	100	92.31
% of moderate risk	0	0	0	0	61.54	0	7.69
% of considerable risk	0	0	0	0	15.38	0	0
% of high risk	0	0	0	0	0	0	–
% of very high risk	0	0	0	0	0	0	0
Anthrosols (*n* = 18)	2.06 (0.23)	5.97 (0.76)	6.34 (1.05)	1.53 (0.18)	56.26 (8.34)	6.03 (0.41)	78.19 (9.94)
% of low risk	100	100	100	100	0	100	100
% of moderate risk	0	0	0	0	100	0	0
% of considerable risk	0	0	0	0	0	0	0
% of high risk	0	0	0	0	0	0	–
% of very high risk	0	0	0	0	0	0	0
All (*n* = 31)	1.84 (0.35)	5.24 (1.41)	6.14 (2.19)	1.51 (0.34)	59.75 (21.38)	6.00 (0.89)	80.48 (23.35)
% of low risk	100	100	100	100	9.68	100	96.77
% of moderate risk	0	0	0	0	83.87	0	3.23
% of considerable risk	0	0	0	0	6.45	0	0
% of high risk	0	0	0	0	0	0	–
% of very high risk	0	0	0	0	0	0	0

[A] Standard deviations

Table 5 Risk assessment code (RAC) and modified risk assessment code (mRAC) values of trace metals in greenhouse vegetable production soils

Mobility risks	RAC (SD[A])						mRAC (SD)
	Cr	Ni	Cu	Zn	Cd	Pb	
Argosols (*n* = 13)	0.06 (0.02)	4.66 (2.21)	2.78 (4.31)	7.57 (6.37)	27.31 (7.29)	0.62 (0.70)	18.07 (5.10)
% of no risk	100	0	23.08	0	0	84.62	0
% of low risk	0	92.31	69.23	84.62	0	15.38	7.69
% of medium risk	0	7.69	7.69	15.38	84.62	0	92.31
% of high risk	0	0	0	0	15.38	0	0
% of very high risk	0	0	0	0	0	0	0
Anthrosols (*n* = 18)	0.05 (0.01)	1.89 (0.50)	0.85 (0.35)	5.72 (2.50)	30.61 (6.54)	0.28 (0.16)	19.57 (4.15)
% of no risk	100	5.56	55.56	0	0	100	0
% of low risk	0	94.44	44.44	88.89	0	0	5.56
% of medium risk	0	0	0	11.11	50	0	94.44
% of high risk	0	0	0	0	50	0	0
% of very high risk	0	0	0	0	0	0	0
All (*n* = 31)	0.06 (0.02)	3.05 (2.01)	1.66 (2.91)	6.49 (4.54)	29.23 (6.94)	0.43 (0.49)	18.94 (4.56)
% of no risk	100	3.23	41.94	0	0	93.55	0
% of low risk	0	93.55	54.83	87.10	0	6.45	6.45
% of medium risk	0	3.23	3.23	12.90	64.52	0	93.55
% of high risk	0	0	0	0	35.48	0	0
% of very high risk	0	0	0	0	0	0	0

[A] Standard deviations

from the six trace metals evaluated by the mRAC index as a whole, more than 90% of the GVP soil samples in the study areas had medium potential adverse effect because the mRAC values in these soil samples generally varied within 10 and 30.

Discussion

Effect of anthropogenic activities on trace metal occurrence in GVP soil

Considering the occurrence of trace metals in GVP soils evaluated by the GSQS (State Environmental Protection Administration of China (SEPAC) 2006) and geo-accumulation index (Table 1 and Fig. 2), it is easily found that anthropogenic activities contributed greatly to Cd and Zn pollution at more than half of the GVP soil sampling sites and Cu, Ni, and Pb pollution at some GVP soil sampling sites.

The results revealed that Zn concentrations in all the GVP soils ($n = 31$) were significantly higher than those in OF soils ($n = 19$) ($P = 0.034$). Thus, zinc pollution in GVP soils was mainly associated with greenhouse farming. From the household survey, we found that application rates of chicken manure and commercial organic fertilizers during GVP in the study area have reached 59.5 and 31.5 t ha^{-1} year^{-1}, respectively. Given that these organic fertilizers always contain high Zn concentrations (Yang et al. 2013, 2016), it can be deduced that Zn pollution in GVP soils of this study was predominately related to heavy application of organic fertilizers. This is consistent with the previous studies (Yang et al. 2013; Bai et al. 2015; Tian et al. 2016).

Despite that the study areas had about ten-year GVP history (Table S1), pollution of Cd, Cu, Ni, and Pb cannot be explained by intensive application of organic fertilizers because concentrations of these metals showed no significant differences between GVP and OF soils (Table 1). In view of locations of sampling sites (Fig. 1), industrial activities are likely to be the main sources of trace metals, especially Cd, Cu, and Ni, because 61.3, 3.2, and 9.7% of all the GVP soil samples had Cd, Cu, and Ni concentrations exceeding GSQS (State Environmental Protection Administration of China (SEPAC) 2006) (Table 1), respectively.

Specifically, the GVP soil sampling sites with Cd concentrations above GSQS were mainly located in Xiashu Town, Xinba Town, Economic development zone of Yangzhong City, and Sanmao and Xinglong Sub-districts (Fig. 1). These towns and sub-districts, except for Xiashu Town, were all located in Yangzhong City. Given that the sampling sites in Xiashu Town were near a thermal power plant, it can be deduced that Cd pollution of GVP soil in Xiashu Town might be primarily ascribed to atmospheric Cd emissions caused by coal combustion from the thermal power plant (Tian et al. 2012a; Shao et al. 2013). Similarly, there were considerable factories around GVP soil sampling sites in Yangzhong (Fig. 1 and Table S1). These factories included those of electroplating, plastics manufacturing, metal product, and textile, which have been identified as possible anthropogenic source of Cd pollution in soil in previous studies (Adriano 2001; Lin et al. 2002; Kirkham 2006; Manzoor et al. 2006). It can be deduced that Cd pollution of GVP soil in the study area of Yangzhong is likely to have mainly resulted from irrigation using sewage from these factories. Notably, some research have reported that industrial sewage irrigation from electroplating affected Cd in soil greatly in Yangzhong City especially before electroplating plants were required to move into industrial parks (Dong et al. 2014), which indicates that the electroplating industry in Yangzhong was an important source of Cd pollution in GVP soil. In contrast, among all the GVP soils, only one sampling site located adjacent to a copper plant in Xinfeng Town (Fig. 1) had Cu concentration exceeding GSQS. So, discharges from this copper plant are likely to be the main source of Cu pollution in GVP soil at this site. Additionally, the GVP soil sampling sites with Ni concentrations above GSQS were mainly located in Xiashu Town and Xinfeng Town (Fig. 1). Likewise, the sampling sites in Xiashu Town were in the vicinity of a thermal power plant. So, nickel pollution of GVP soil in Xiashu Town might be closely related to atmospheric Ni emissions caused by coal combustion (Tian et al. 2012b). Whereas Ni pollution in Xinfeng Town was greatly attributed to discharges from a scrap metal recycling plant since the sampling site was adjacent to this plant (Ayres 1997).

In conclusion, industrial activities exacerbated pollution of trace metals, especially Cd, Cu, and Ni in GVP soil near industrial areas in the Yangtze River Delta, which deserves great concern from perspective of sustainable GVP.

Trace metal speciation in GVP soil

In summary, the results revealed that the concentrations and percentages of Cr, Ni, Cu, and Zn in residual fraction were generally higher than those in non-residual fractions (F1 + F2 + F3) while the concentrations and percentages of Cd and Pb in residual fraction were lower than those in non-residual fractions in GVP soils (Tables 2, 3, and S3).

Among the metal fractions, the acid-soluble metals are the weakest bonded metals in soils and considered to be labile, more easily bioavailable, and highly toxic (Gao and Chen 2012; Moore et al. 2015). The result that the percentages of acid-soluble Cd fraction were much higher than those of acid-soluble Cr, Ni, Cu, Zn, and Pb fraction (Table 3) indicates that Cd had relatively higher mobility in GVP soil. In the Anthrosols area, GVP soils generally had lower concentrations and percentages of acid-soluble Ni, Cd, and Pb, in contrast with OF soils (Tables 2 and 3). This may be predominantly associated with soil parent material and soil acidification caused by GVP. Considering that the soil in the study Anthrosols area is derived from calcareous deposits of the Yangtze River, it can be speculated that carbonate-bound Ni, Cd, and Pb might account for most of acid-soluble Ni, Cd, and Pb in soil of the study Anthrosols area. Furthermore, based on the significantly positive correlation between soil pH and carbonate-bound metals reported in previous study (Cui et al. 2013; Houben and Sonnet 2015), it can be concluded that acidification in GVP soil of the study Anthrosols area (Table 1) might lead to the decrease of carbonate-bound Ni, Cd, and Pb and subsequent decrease of acid-soluble Ni, Cd, and Pb. Also, this result can explain why concentrations and percentages of acid-soluble Ni, Cd, and Pb were significantly and positively correlated with soil pH in the Anthrosols area (Tables S4 and S5). Unlike Ni, Cd, and Pb, concentrations and percentages of acid-soluble Zn in GVP soil were significantly higher than those in OF soil in the Anthrosols area (Tables 2 and 3). This was greatly attributed to accumulation of Zn in the GVP soil because soil total Zn concentration, significantly and positively correlated with soil acid-soluble Zn, was also found to be a major factor controlling soil acid-soluble Zn (Tables S4 and S5).

As a whole, industrial discharge and intensive fertilization not only resulted in pollution of Cd, Zn, Cu, Ni, and Pb but also increased concentrations of mobile Cd, Zn, Cu, Ni, and Pb since concentrations of acid-soluble Cd, Zn, Cu, Ni, and Pb in all the soil samples were significantly and positively correlated with their respective total concentrations in soil (Table S4). This indicates that both intensive farming and industrial activities increased the bioavailability of Cd, Zn, Cu, Ni, and Pb in GVP soil, which might increase subsequent health risks via the food chain.

Risk assessment of trace metals in GVP soil

Although most of the GVP soils had low ecological risk based on assessment by RI, the single potential ecological risk factor (Er^i) values indicated that Cd posed moderate or considerable ecological risk at most of the GVP soil sampling sites (Table 4). Similarly, the mRAC values revealed that most of the GVP soil samples in the study areas had medium potential adverse effect (Table 5). Furthermore, among the study trace metals, Cd, which displayed medium or high mobility risk at all the GVP soil sampling sites, was also found to be the major mobility risk contributor. Given that the ingestion of Cd will lead to deleterious effects on human health (Jarup 2003), medium or high risk of Cd release in GVP soil should be of great concern.

In order to alleviate these environmental risks, the reduction of industrial discharges and organic fertilizer application rates is necessary. Also, remediation technologies for the phytoavoidation of trace metals, particularly Cd, such as the immobilization of Cd in GVP soils by soil amendments (e.g., limestone, biochar, and hydroxyapatite) (Cui et al. 2013; Zhou et al. 2014; Zia-ur-Rehman et al. 2017) and the selection of low metal accumulation vegetable species for cultivation (Yang et al. 2016) should be considered, which can abate mobility risk of Cd through reducing Cd movement into the food chain.

Conclusions

The results showed that the concentrations (mg kg^{-1}) ranged from 0.061 to 0.48 for Cd, 19.3 to 92.8 for Cu, 66.6 to 181.7 for Zn, 18.7 to 38.6 for Pb, 50.5 to 98.4 for Cr, and 21.1 to 64.1 for Ni in the GVP soil in the study area. The geo-accumulation index revealed that the main trace metal elements causing GVP soil pollution were Cd and Zn. Pollution levels of Cd and Zn were generally unpolluted to moderately polluted. Compared with GSQS, more than 60% of the GVP soils in the study areas were contaminated by Cd. In contrast, only some GVP soils were contaminated by Ni and Cu. Cadmium, Cu, and Ni pollution are likely to have mainly resulted from industrial discharges, while Zn pollution was primarily related to heavy fertilization during GVP. More than half of Cu, Zn, Ni, and Cr (68.3–91.7%) were associated with residual fraction, while more than half of Cd and Pb (50.5–75.4%) were related to non-residual fractions. Pollution of Cd, Zn, Cu, and Ni has resulted in the increase of their concentrations in mobile fractions. This suggests that pollution caused by both intensive farming and industrial activities increased the bioavailability of Cd, Zn, Cu, and Ni in GVP soil, which might increase subsequent health risks via the food chain.

The potential ecological risk index and risk assessment code showed that Cd was the major risk contributor and generally posed moderate or considerable ecological risk as well as displayed medium or high mobility risk at the GVP soil sampling sites. In order to abate trace metal pollution and their environmental risks, the reduction of industrial discharges and fertilizer application and the adoption of remediation technologies for the immobilization of trace metals in GVP soils should be considered.

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