Regional accumulation characteristics of cadmium in vegetables: Influencing factors, transfer model and indication of soil threshold content[☆]

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ABSTRACT

A regional investigation in the Youxian prefecture, southern China, was conducted to analyze the impact of environmental factors including soil properties and irrigation in conjunction with the use of fertilizers on the accumulation of Cd in vegetables. The Cd transfer potential from soil to vegetable was provided by the plant uptake factor (PUF), which varied by three orders of magnitude and was described by a Gaussian distribution model. The soil pH, content of soil organic matter (SOM), concentrations of Zn in the soil, pH of irrigation water and nitrogenous fertilizers contributed significantly to the PUF variations. A path model analysis, however, revealed the principal control of the PUF values resulted from the soil pH, soil Zn concentrations and SOM. Transfer functions were developed using the total soil Cd concentrations, soil pH, and SOM. They explained 56% of the variance for all samples irrespective of the vegetable genotypes. The transfer functions predicted the probability of exceeding China food safety standard concentrations for Cd in four major consumable vegetables under different soil conditions. Poor production practices in the study area involved usage of soil with pH values ≤ 5.5 , especially for the cultivation of *Raphanus sativus* L., even with soil Cd concentrations below the China soil quality standard. We found the soil standard Cd concentrations for cultivating vegetables was not strict enough for strongly acidic (pH ≤ 5.5) and SOM-poor (SOM $\leq 10 \text{ g kg}^{-1}$) soils present in southern China. It is thus necessary to address the effect of environmental variables to generate a suitable Cd threshold for cultivated soils.

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1. Introduction

Cadmium (Cd) can cause adverse health effects and is a subject of concern in food production and consumption since the element is easily incorporated in crops such as rice and vegetables (Turner, 1973; McBride, 2002; Adams et al., 2004; Chaney, 2015). The consumption of vegetables is a probable cause of Cd exposure in several world areas including China (Hough et al., 2004; Xu et al., 2013; Norton et al., 2015). In some countries like southern Sweden and Uganda, the consumption of vegetables may contribute from 70% to 80% of the total Cd intake by humans (Nabulo et al., 2010; Swartjes et al., 2013). Low-level concentrations of Cd in

vegetables can also lead to kidney and liver dysfunction (Thomas et al., 2009).

The potential transfer of trace elements from soil to plant is estimated by the dimensionless plant uptake factor (PUF) that refers to the calculated ratio of trace element concentrations in edible plant tissues relative to that of the paired-soil (Chen et al., 2009; Zhang et al., 2011). The concentration of Cd in vegetables is dependent on surrounding environmental factors (Hossain et al., 2010; Augustsson et al., 2015). Significant variables contributing to the Cd intake by vegetables are: the vegetable species, irrigation environment, soil properties (e.g. pH, SOM, cation exchange capacity, texture, etc.), and fertilizer usage (Chaney et al., 1978; Kuboi et al., 1986; Nabulo et al., 2012; Zhao et al., 2015). A large uncertainty appears when use PUF to describe the accumulation of Cd in vegetables in relation to a large range of environmental factors (Chen et al., 2009; Swartjes et al., 2013). Several studies focusing on the relation between Cd concentrations in vegetables and total Cd

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soil concentrations or soil pH, use empirical regression methods. In doing so, other environmental factors such as SOM, irrigation condition, and fertilizer usage, which related to the accumulation of Cd in vegetables are often overlooked (McBride, 2002, 2014; Hough et al., 2004). A large number of national environmental agencies recommend investigating the environmental factors related to the plant Cd uptake (Swartjes et al., 2013; Augustsson et al., 2015), but detailed studies are few.

The established quality standard of soils in several countries is derived from the total concentration or available forms of trace element with or without involving the soil pH (Zhang et al., 2011). This produces an inaccurate assessment of actual risks to the population (Nabulo et al., 2010; Norton et al., 2015). Predictive models factoring in environmental variables on the accumulation of Cd in crops would allow improved soil quality standards that provide a more risk-based approach to the protection of farmlands (Ding et al., 2013). Other studies established a Cd threshold for rice and vegetables using predictive models (Adams et al., 2004; Zhang et al., 2011). However, these were usually carried out on potted or greenhouse vegetables using Cd-enriched soils having a narrow range of compositions (Chen et al., 2009; Augustsson et al., 2015).

The prefecture of Youxian located in southern China was chosen as the case study area to analyze the effect of diverse environmental factors on the accumulation of Cd in vegetables. The investigation relied on a large scale agricultural survey accompanied by paired sampling of soil-vegetable. The specific objectives of our research are: 1) to characterize the transfer of Cd to the edible segments of vegetables under a wide range of environmental conditions; 2) to qualify the main influencing factors and provide regression-based predictive models for the Cd accumulation in vegetables under different soil compositions; and 3) to establish a Cd threshold for cultivated soils producing vegetables that is based upon the Chinese regulations for food consumption.

2. Methods and materials

2.1. Study area and agricultural survey

The Youxian prefecture (113.32 °E long., 27.01 °N lat.) is located in the Hunan province of southern China and is a major regional crop producer (Fig. 1). The prefecture comprises 23 townships covering an area of 2648 km² and contains a population of 0.8

million. The region is affected by the subtropical monsoon and presents a mild and moist climate, with a moderately cold winter and a humid summer. The prefecture of Youxian is known nationwide as the “Cd-laced rice” area (Wang et al., 2016). The contamination of the agricultural products by Cd is of great concern to the government and population, but the process by which Cd pollutes various types of vegetables is unknown.

The fertilizer usage contributed to the Cd accumulation in cropland soils and produced soil acidification (Guo et al., 2010; Zhao et al., 2015). Nitrogenous, phosphorus, potassic, and compound fertilizers are commonly spread on vegetable fields within the study area. The quantity of applied fertilizers from different sampled areas was obtained from a household survey performed in 23 townships. The annual average fertilizer use from different townships was selected for the multivariate statistical analysis. Detailed information on the fertilizer usage is listed in Table S11.

2.2. Field sampling

A total of 791 matched soil and vegetable and 585 water samples were collected throughout the Youxian prefecture (Fig. 1). All vegetables were harvested from open field cultures. We investigated and recorded the growth status and types of vegetables from each site. Then 5 to 10 sub-samples extracted from the edible parts of the vegetable were collected and stored in sealed polyethylene bags. We sampled 51 vegetable species from 25 genera belonging to 12 families (Table S12). The sampled vegetables comprised the Chinese cabbage (*Beassica pekinensis* L.), bok choy (*Brassica rapa* var. chinensis), radish (*Raphanus sativus* L.), and lettuce (*Lactuca sativa* L.) which constitute the four major consumed vegetables of the study area, and the sample proportion for these four species were 10.7%, 14.3%, 11.1%, and 7.1%, respectively.

Five soil samples were collected from the topsoil at 0–20 cm depth from where each sampled vegetable grew. Samples of irrigation water were gathered at the surface from different irrigation areas associated with vegetable cropping fields. The water samples were poured into 50 ml acid-washed polyethylene bottles and stored in a refrigerated box until analysis.

2.3. Analytical procedures

The irrigation water pH was measured in the field. Water samples were first filtered through a 0.22 µm filter membrane (Millipore), and then kept at –20 °C for Cd concentration determination. Soil samples were air-dried, sieved through a 2 mm nylon mesh, and then milled for further analysis. The soil pH was determined using a 1:2.5 soil-to-water ratio. The soil organic matter, cation-exchange capacity, and clay contents were measured following routine analytical methods for soil analysis detailed in Bao (2000). Vegetable samples were washed with tap water, then rinsed 4–5 times with deionized water, dried in an oven at 60 °C and finally milled for chemical analysis. The vegetable samples were digested in a concentrated HNO₃–HClO₄ solution, whereas soils were digested in a mixture of HCl–HNO₃–HF–HClO₄ (Wang et al., 2016).

The Cd and Zn concentrations were determined by Graphite Furnace Atomic Absorption Spectroscopy (GFAAS, Germany). Quality assurance and control procedures involved the inclusion of standard reference materials in each analyzed batch; namely GSB-5 (cabbage), GSB-25 (carrot) and GSB-4 (soybean) for vegetables, GSS-5 for soil, and GSB07-1185-2000 for water. The recovery ratio of standards ranged from 82.4% to 104.7%, 93.9%–107.9%, and 85.1%–118.0% for cabbage, carrot, and soybean, respectively, and from 92.1% to 105.6% for soil, and 81.3%–116.5% for water.

Fig. 1. Location of the study area and the sampling sites.

2.4. Path analysis model

The direct cause-and-effect relationships between environmental factors and vegetables accumulating Cd from soil are difficult to determine because environmental factors are often correlated (Basta et al., 1993; Ding et al., 2013). By partitioning the correlation coefficient into direct and indirect effects, the path model (PA) can be used to determine the causal relationship in the agronomic studies (Shipley, 2009). In this study, a PA was constructed to analysis the relationship between plant uptake factor (PUF) and soil pH, SOM, soil Zn, irrigation water pH, and nitrogenous fertilizer usage (N-fertilizer) (Fig. 2; the selection of analyzed environmental variables are shown in Section 3.2). Direct effect of environmental variables (D_{ij}) on PUF values is represented by single arrows, while coefficients of correlations between environmental factors are illustrated by double-headed arrows (Fig. 2). The direct effects of environmental variables on PUF are termed path coefficients and derived from multiple regression of environmental variables on PUF (Basta et al., 1993). Indirect effects are determined from the product of a simple correlation coefficient between environmental factors and the path coefficients (Ding et al., 2013). The relations between the correlation coefficient, direct effect and indirect effect are described in the following function:

$$r_{i6} = D_i + \sum_{j=1, j \neq i}^5 r_{ij} \times D_j \quad i = 1, 2, \dots, 5 \quad (1)$$

where subscript designations are: (1) soil pH, (2) SOM, (3) soil Zn, (4) water pH, (5) N-fertilizer, and (6) PUF (Fig. 2); r_{i6} is the correlation coefficient between i_{th} environmental variable and PUF, D_i is direct effect of i_{th} environmental variable on PUF, and $r_{ij} \times D_j$ is the indirect effect of environmental variable on PUF.

Since the multivariate normality in assuming a large sample size may not hold (Shipley, 2009), bootstrap simulations were applied to explain the direct and indirect effects during the PA analysis (Cheung and Lau, 2008). In total, 1000 operations were performed to obtain stable results. The effectiveness of the PA is evaluated by the mean root-square error and the variance (R^2) (Ding et al., 2013).

2.5. Statistical analysis

Spearman correlation (two-tailed) and step-wise multiple regression analyses were performed using the Matlab 14.0a software. Path model analysis and bootstrap simulations were achieved via the Amos 17.0 software. Except for the soil and water pH values, other data were log-transformed prior to analysis to avoid their low

Table 1

Descriptive statistics of Cd concentrations in water, soil and vegetable samples as well as major environmental variables.

Variables	Min	Max	Mean	SD	Median
Soil pH	3.4	8.0	5.3	0.8	5.1
Water pH	4.5	7.5	6.3	0.8	6.7
Clay (%)	43.1	81.3	66.8	11.5	72.6
Soil organic matter (g kg ⁻¹)	5.3	93.4	21.6	12.2	18.0
CEC (cmol [±] kg ⁻¹)	3.8	19.5	9.7	3.8	9.1
Water Cd (ug L ⁻¹)	0.001	1.50	0.13	0.13	0.09
Soil Zn (mg kg ⁻¹)	23.2	248.7	112.7	34.6	111.4
Soil total Cd (mg kg ⁻¹)	0.01	3.00	0.47	0.31	0.42
Vegetables Cd (mg DW kg ⁻¹)	0.003	1.81	0.06	0.07	0.05
<i>B. pekinensis</i> Cd (mg DW kg ⁻¹)	0.01	0.12	0.05	0.03	0.05
<i>B. rape</i> Cd (mg DW kg ⁻¹)	0.01	0.11	0.05	0.02	0.05
<i>R. sativus</i> Cd (mg DW kg ⁻¹)	0.01	0.15	0.06	0.03	0.05
<i>L. sativa</i> Cd (mg DW kg ⁻¹)	0.02	0.27	0.07	0.05	0.06
Plant uptake factor (PUF)	0.01	2.1	0.15	0.13	0.12

levels of significance (Shapiro-Wilk determination, $p < 0.05$) and non-normal distributions.

3. Results and discussion

3.1. Cadmium concentrations in soils and vegetables

Table 1 summarizes the concentration of Cd in irrigation water, soil and vegetable samples accompanied by major environmental factors. The Cd content of irrigation water averaged $0.13 \mu\text{g L}^{-1}$, well within the values proposed in the national water guideline (SEPA, 2002). The average Cd soil concentration is 0.47 mg kg^{-1} , with 79.1% of soil samples exceeding the proposed Cd limit for agricultural soils in China (e.g. 0.25 mg kg^{-1} , SEPA, 2008). The vegetables Cd concentrations ranged from 0.003 to 1.81 mg kg^{-1} on a dry weight (DW) basis, averaging 0.06 mg kg^{-1} . Vegetables of the Umbelliferae family display significant higher accumulation of Cd ($p < 0.05$) compared to that of other vegetable families. The Cd concentrations in edible vegetable parts did not vary significantly between the four major groups.

According to a National Nutrition Survey in the Hunan province (SEPA, 2013), the daily average vegetable consumption of people living in the Hunan Province can be as high as 357.3 g d^{-1} , next only to the average consumption by the population of the Hubei Province (SEPA, 2013). Several reports indicated the cultivation of rice and tobacco in Cd-contaminated soils leads to dangerous Cd exposures for people living in affected areas (Thomas et al., 2009; Wang et al., 2016). A conservative Cd concentration of

Fig. 2. Schematic description of the path model analysis considering the influence of environmental variables on PUF values. The subscript designations are: (1) soil pH, (2) soil Zn, (4) water pH, (5) N-fertilizer, and (6) PUF. The correlation coefficients (r_{ij}) between each component pair are illustrated by double-headed arrows. The direct effect of environmental variables (D_{i6}) on PUF values is defined by single arrows.

Fig. 3. Correlations between Cd concentrations in soils and vegetables from: (a) all samples, (b) uncontaminated sites and, (c) highly contaminated sites.

0.05 mg kg⁻¹ in vegetables, suggested by the State Health Administration of China (SHAC, 1994), was adopted as a toxicological reference to the present study. Therefore, 48.3% of the collected vegetable samples exhibited Cd concentrations surpassing the maximum safe value. Excessive Cd values are recorded for 42.3%, 43.4%, 60.2%, and 58.9% of *B. pekinensis*, *B. rapa*, *R. sativus*, and *L. sativa*, respectively. Vegetables belonging to the Umbelliferae, Liliaceae, and Brassicaceae families showed high accumulations of Cd, whereas vegetables from the Rutaceae, Solanaceae, Leguminosae, Cucurbitaceae families are safe, in agreement with the investigations of Kuboi et al. (1986), Xu et al. (2013), and Norton et al. (2015).

The vegetables Cd concentrations do not correlate with the paired-soil Cd contents (Fig. 3a), an observation recorded by other studies (Chen et al., 2009; Zhang et al., 2011). The poor correlation may be attributed to the highly Cd contamination in soil (McBride et al., 2014). Fig. 3b–c presents the samples collected from highly contaminated areas, showing soil Cd contents > 0.75 mg kg⁻¹ (three-fold greater than the soil Cd threshold), compared to uncontaminated sites (Cd content in soils < 0.25 mg kg⁻¹). The concentration of Cd in vegetables cultivated in highly contaminated areas are higher ($p < 0.001$) to that of samples from clean sites. Therefore, vegetables growing in contaminated agricultural soils tend to accumulate a high level of Cd. However, there is no significant relation between samples from these two groups (Fig. 3b–c), suggesting a very difficult risk management.

The PUF values varied from 0.01 to 2.1, with a mean value of 0.15 (Table 1), indicating a low level of Cd transfer from soil to vegetables. The probabilistic distribution of PUFs in logarithmic form was determined by the statistical method described in Chen et al. (2009). Fig. 4a confirms a Gaussian distribution matches the PUF distribution. The range of PUF values is approximately three orders

of magnitude. *R. sativus* and *L. sativa* are two of the four major eaten vegetables showing a propensity to accumulate Cd from the paired soil (Fig. 4a). Chen et al. (2009) and Zhang et al. (2011) used a Gaussian distribution function to match the Cd PUF variations for vegetables cultivated in California and vegetable data extracted from the literature. In general, the reported PUF values and distribution patterns (Chen et al., 2009; Zhang et al., 2011; Swartjes et al., 2013) were comparable to that of the PUFs calculated in the present study.

3.2. Correlation with environmental factors

Soil and vegetable crop Cd concentrations may reflect the state of contamination of an entire agricultural system (Swartjes et al., 2013). The PUF may correlate with the vegetable species, soil properties, types of irrigation and fertilizer use (Zhang et al., 2011; Augustsson et al., 2015). Table 2 summarizes the significant Spearman correlation coefficients between PUF values and other important environmental factors. The soil properties control in large part the accumulation of Cd in vegetables. For instance, the soil pH ($r = -0.667^{**}$), soil organic matter content (SOM, $r = -0.428^{**}$), and soil Zn concentrations ($r = -0.412^{**}$) display significant correlations with PUF values. The pH of irrigation water ($r = -0.099^{*}$) were negatively correlated with the PUF values. In contrast, the nitrogen fertilizer usage showed a major influence on PUF values with $r = 0.189^{*}$.

A path model (PA) was employed to better define the correlations between the PUF values and independent variables such as: the soil pH, SOM, soil Zn concentrations, irrigation water pH and nitrogen fertilizer usage (Fig. 2). The results display a mean root-square error value of 0.17 and a variance (R^2) of 0.63, thus defining a good fit. Table 3 summarizes the PA analysis results of

Fig. 4. Measured and predicted cumulative probability distributions of Cd for: (a) total vegetable samples, (b) vegetables collected in the strong acidic group (soil pH ≤ 5.5) and, (c) vegetables from the natural group (soil pH > 5.5). The asterisks (☆) from the left to right refer to the minimum, mean, upper 95% confidence level and maximum PUF value, respectively. The cumulative probability distribution of PUF was matched by a Gaussian distribution function (Chen et al., 2009), so that $F(x) = 0.5 + 0.5 \times \text{erf}((x - x_0)/b\sqrt{2})$, where $F(x)$ is the cumulative probability that $\text{PUF} \leq x$, x_0 and b are the mean and standard deviations of the population and erf is the error function.

Table 2

Significant Spearman correlation coefficient between major environmental factors and PUFs.

PUF groups	Soil pH	SOM	Soil Zn	Irrigation water pH	Nitrogen fertilizer
Total vegetables	−0.667**	−0.428**	−0.412**	−0.099*	0.189*
<i>B. pekinensis</i>	−0.687**	−0.366**	−0.258*	−0.186*	0.153*
<i>B. rapa</i>	−0.710**	−0.381**	−0.358**	−0.106*	0.107*
<i>R. sativus</i>	−0.668**	−0.312*	−0.538**	/	0.080*
<i>L. sativa</i>	−0.746**	−0.421**	−0.559**	−0.160*	0.294**

*Correlation is significant at $p < 0.05$.**Correlation is significant at $p < 0.01$.**Table 3**

Direct effects (first column with bold letter) of environmental variables on PUFs and correlation coefficients (italic letter) among environmental variables.

Environmental factors	PUF	Soil pH	SOM	Soil Zn	Irrigation water pH
Soil pH	−0.239**				
SOM	−0.461**	<i>0.097*</i>			
Soil Zn	−0.419**	<i>0.244**</i>	<i>0.265**</i>		
Irrigation water pH	0.009	<i>0.116**</i>	<i>0.138**</i>	<i>0.054</i>	
Nitrogen fertilizer usage	0.017	<i>−0.144**</i>	<i>−0.269**</i>	<i>−0.068</i>	<i>−0.059</i>

*Correlation is significant at $p < 0.05$.**Correlation is significant at $p < 0.01$.

correlation coefficients and direct effects (D). The irrigation water pH are negatively correlated with the soil pH, but positively correlated with the amount of SOM. While soil pH ($D_{16} = -0.239^{**}$) and SOM ($D_{26} = -0.461^{**}$) had significant direct effects on PUF. Our results indicate the good correlation between irrigation water pH and PUF values are connected to the soil pH and quantity of SOM. The moist environment increases the biological activity, producing soil acidity (Reiser et al., 2014). The study area is characterized by a dense network of rivers (Fig. 1), providing enough water for irrigation. A higher field-moisture capacity generates a rapid acidification causing the migration of Cd to weaker bounding sites thus promoting the vegetables uptake. There is a poor correlation between the usage of nitrogen fertilizers ($D_{56} = 0.017$) and the PUF values, whereas a significant negative correlation between nitrogen fertilizer use and soil pH and SOM is shown in Table 3. The fixation of NH_4^+ and $\text{R}-\text{NH}_2$ present in nitrogen-based fertilizers cause the nitrification and a persistent soil acidification (Bolan et al., 1991). Extensive usage of nitrogen fertilizers promotes the microbial transformation of the soil organic carbon and leads to a decline in SOM (Khan et al., 2007). There was an intensification of agricultural practices in the study area since the 1980's inducing an excessive application of nitrogen fertilizers. This led to a substantial loss of SOM and a worsening of soil acidification ultimately causing increasing Cd accumulation in vegetables.

The soil Zn concentrations were also well correlated with the PUF values (e.g. $D_{36} = -0.419^{**}$), with no significant correlation with other environmental variables (Table 3). The evaluation of the Cd level in leafy vegetables, bio-solid modified soils, and in animal tissues, indicated any regulation on the Cd concentrations in crops should also consider the levels of Zn (Turner, 1973; Chaney et al., 1978). Cadmium and Zn are closely related from pollution sources and are accumulated in crops (McBride, 2002; Chaney, 2015). Our work reveals a significant correlation between Cd and Zn concentrations in soils ($r = 0.551^{**}$), but a poor correlation for vegetables ($r = 0.091$). Chaney (2015) demonstrated a soil Zn:Cd ratio $< 100:1$ leads to a greater Cd accumulation in crops. However, the average concentration of Zn in the sampled soils was 112.7 mg kg^{-1} (Table 1), slightly above the local background value of 94.4 mg kg^{-1} (SEPA, 1990). Only 2.3% of the analyzed samples possessed a Zn:Cd ratio $< 100:1$, suggesting that Zn had no control over the accumulation of Cd in vegetables. These results suggested the impacts of

soil Zn on the crop Cd uptake may be site-specific. The PA analysis was performed individually for the PUF values of the four major consumed vegetables. The soil pH and SOM contents showed a good correlation with the PUFs. These results mean a correlation analysis alone cannot characterize the PUF variations against environmental factors.

3.3. Predicting the Cd accumulation in vegetables

The normal and log format Cd concentration in vegetables and soil showed poor correlations ($R^2 = 0.08$ and 0.11 , respectively). Adams et al. (2004) demonstrated the samples collected from highly contaminated areas to have a large control on the overall correlation. Even when 21 soil samples collected from high risk sites and containing Cd concentrations $> 1.25 \text{ mg kg}^{-1}$ were excluded, the log regression provided a low R^2 value (e.g. 0.10), suggesting the total soil Cd concentrations alone were a poor predictor of Cd vegetable accumulation. A stepwise multiple linear regression analysis was therefore used to define the relation between environmental factors and the Cd accumulation in vegetables. Table 4 summarizes the prediction models for the Cd transfer based on the extended Freundlich-type function (Ding et al., 2013). Compared to a simple regression function, the inclusion of the soil pH variable improved the variance from 0.11 to 0.49 (Eq. (2)). When the conditions become acidic, the binding sites of divalent metal ions are increasingly protonated, producing an enhanced competition for adsorption sites and effectively increasing the solubility of certain metals (Sauvé et al., 2000).

The PUFs were thus divided into two groups, following the national soil pH standard (GB15618-2008, SEPA, 2008), and were also described by a Gaussian distribution function (Fig. 4b–c). The probabilistic distributions of PUFs from strong-acidic ($\text{pH} \leq 5.5$) and natural ($\text{pH} > 5.5$) environments were similar at low levels of PUF values, but gradually differed for the elevated PUF values. Sampled vegetables that belong to the natural group tend to be more tolerant to Cd accumulation. Furthermore, the PUF values of the four major consumed vegetables extracted from strong-acidic ($\text{pH} \leq 5.5$) soils were significantly higher ($p < 0.001$) to that of samples belonging to the natural group. The results indicate the soil pH controls the Cd uptake by vegetables, in agreement with other published studies showing vegetables growing in acidic soils are

Table 4
Step-wise multiple linear regression models for Cd accumulation in vegetables.

No. of equations	Groups	Regression model $\log [\text{Cd}_{\text{vegetables}}] =$	n	Model validation		
				R ²	RMSE	p
2	Total vegetables	$0.06 + 0.54 \log [\text{Cd}_{\text{soil}}] - 0.22(\text{pH})$ (2)	791	0.49	0.20	<0.001
3	Total vegetables	$-0.69 + 0.45 \log [\text{Cd}_{\text{soil}}] - 0.36 \log (\text{SOM})$ (3)	791	0.18	0.25	<0.001
4	Total vegetables	$0.56 + 0.66 \log [\text{Cd}_{\text{soil}}] - 0.22(\text{pH}) - 0.36 \log (\text{SOM})$ (4)	791	0.56	0.26	<0.001
5	<i>B. pekinensis</i>	$0.64 + 0.58 \log [\text{Cd}_{\text{soil}}] - 0.23(\text{pH}) - 0.44 \log (\text{SOM})$ (5)	85	0.64	0.17	<0.001
6	<i>B. rapa</i>	$0.31 + 0.58 \log [\text{Cd}_{\text{soil}}] - 0.18(\text{pH}) - 0.35 \log (\text{SOM})$ (6)	113	0.65	0.13	<0.001
7	<i>R. sativus</i>	$0.68 + 0.67 \log [\text{Cd}_{\text{soil}}] - 0.27(\text{pH}) - 0.23 \log (\text{SOM})$ (7)	88	0.72	0.13	<0.001
8	<i>L. sativa</i>	$0.62 + 0.59 \log [\text{Cd}_{\text{soil}}] - 0.23(\text{pH}) - 0.36 \log (\text{SOM})$ (8)	56	0.59	0.16	<0.001

more prone to Cd accumulation (McBride, 2002; Hough et al., 2004; Nabulo et al., 2012). The plant available pool of trace elements is contained in solid phase in soil humus (Sauvé et al., 2000), and this effect is shown by an improved fit (from 0.11 to 0.18) where SOM is included in the model (Eq. (3), Table 4). The transfer function (Eq. (4)) reveals the total soil Cd content, soil pH, and SOM content exert a major influence on the Cd accumulation in vegetables, explaining 56% of the variance. The introduction of additional environmental variables, such as the soil Zn concentrations, CEC, temperature, solar radiation, and fertilizer use, only marginally improved the model fit, and therefore were not included in the model.

3.4. Soil Cd threshold for vegetables cultivation

Transfer functions were used to predict the likelihood of certain Cd concentration in four major consumed vegetables under different soil conditions. Fig. 5 presents the predicted Cd concentration in vegetables using Eqs. (5)–(8), combining the SOM content (20 g kg^{-1} , similar to the average SOM content of the investigated soil) and the Cd concentrations of four soil samples including the local background value (e.g. 0.13 mg kg^{-1} , SEPAC, 1990), the Chinese agricultural soil limit (0.25 mg kg^{-1}), average Cd soil concentrations (0.50 mg kg^{-1}), and severely contaminated soil samples (e.g. 1.25 mg kg^{-1} , five-fold greater than the prescribed soil Cd limit), respectively. As expected, the predicted values increased with the soil Cd concentrations and decreased with the soil pH. *L. sativa* can be safely cultivated in the study area, since the predicted Cd concentrations were always lower to that of the standard limit. While *B. pekinensis* and *B. rapa* were likely to contain Cd concentrations exceeding the Cd limit when planted in soils with local Cd levels and a pH value < 5.4, the cultivation of *R. sativus* was potentially hazardous when growing at sites having Cd concentrations within the prescribed limits but a pH value < 4.8. Results also suggested Cd-rich soils should not be harvested in the production of *B. pekinensis*, *B. rapa*, *R. sativus*, unless the soil pH value is maintained near 6.5. When the soils are acidic, the Cd accumulation in vegetables surpasses that of the food guideline, even if the soil Cd concentrations meet the quality standard.

A soil Cd threshold was computed from Eq. (4), under different combinations of soil pH values and SOM contents based on the Chinese food regulations (SHAC, 1994) to compare with the present

Chinese soil quality standard (GB 15618-2008, SEPAC, 2008). Fig. 6 demonstrates the derived Cd threshold increased with the augmentation of the soil pH and SOM content. Strongly acidic ($\text{pH} \leq 5.5$) and low SOM (10 g kg^{-1}) soils present a Cd threshold ranging from 0.17 to 0.36 mg kg^{-1} . The derived threshold rose from 0.66 to 4.50 mg kg^{-1} , where the natural $\text{pH} > 5.5$ and the SOM content was high (30 g kg^{-1}).

The national soil quality Cd standard is unrelated to the SOM content and is set at 0.25 mg kg^{-1} for soils having $\text{pH} \leq 5.5$ (Fig. 6). However, in southern China, there is a continuous decrease in the soil pH and SOM content (Guo et al., 2010; Zhao et al., 2015). Fig. 6 suggests a derived Cd threshold varying from 0.17 to 0.66 mg kg^{-1} corresponding to pH values between 4.5 and 5.5. The current soil quality standard for vegetables crops under a soil pH value < 5.0 and moderate SOM contents may be over-estimated, and be conservative for vegetables crops under soils pH values > 5.2.

At present, the national soil quality standard is only valid for a limited combination of total soil Cd concentrations and soil pH values, and fails to provide an accurate and rigorous soil Cd regulation for the cultivation of vegetables. The soil quality standard should be reevaluated in view of the control of environmental variables on the Cd accumulation in vegetables. To reduce the potential health risk, China needs a policy requiring higher soil pH in contaminated areas, and the application of liming materials and biochar is reported to improve the acidity of farmland and increase the vegetables yield (Hossain et al., 2010; Zhao et al., 2015).

4. Conclusion

High Cd concentrations were observed in paired soil and vegetable samples collected in the Youxian prefecture, southern China. Evaluation of the Cd accumulation potential in four major consumed vegetables, indicates the *R. sativa* potential was larger to that of the other three vegetable species. The PUF values exhibited comparable results and appeared to define a reasonable and consistent Cd risk assessment. Many environmental variables exhibited significant correlations with the concentrations of Cd in the soil-vegetable system, with the soil pH values, soil Zn concentrations, and SOM contents being the dominant factors. A transfer model combining the soil Cd concentrations, soil pH values, and SOM content variables better predicted the Cd concentration in

Fig. 5. Predicted Cd concentrations in (a) *B. pekinensis*, (b) *B. rapa*, (c) *R. sativus*, and (d) *L. sativa* under different soil conditions.

Fig. 6. Derived soil Cd thresholds for cultivated vegetables. The 25th, 50th, and 75th percentiles of the SOM contents represent concentrations of 10, 20, and 30 g kg⁻¹, respectively, corresponding to low, moderate, and high SOM contents.

vegetables. Under the current Chinese soil quality standard, we should pay more attention to the cultivation of *B. pekinensis*, *B. rapa*, and *R. sativus* in the study area. The current national soil quality standards may not fully acknowledge the control of environmental variables on the Cd uptake of vegetables. An improved management of soils pH values and SOM content is needed for a better and safer vegetable production.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.envpol.2016.09.003>.

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