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Cadmium accumulation in edible flowering cabbages in the Pearl River Delta, China: Critical soil factors and enrichment models☆

Chengshuai Liu [a, b], Chunying Chang [c], Yingheng Fei [d], Fangbai Li [a, *], Qi Wang [a], Guangshu Zhai [a], Jing Lei [e]

[a] Guangdong Key Laboratory of Integrated Ago-Environmental Pollution Control and Management, Guangdong Institute of Eco-Environmental Science & Technology, Guangzhou 510650, China
[b] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, China
[c] Guangdong Key Laboratory of Contaminated Sited Environmental Management and Remediation, Guangdong Provincial Academy of Environmental Science, Guangzhou 510045, China
[d] School of Environmental Science and Engineering, Guangzhou University, Guangzhou 510006, China
[e] College of Agriculture, Guangxi University, Nanning 530005, China

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ABSTRACT

Although many previous studies have reported the soil pH and organic matter to be the most critical factors that affect the transfer of Cd in soil-crop systems in temperate zones, the behavior of Cd transfer is different in the Pearl River Delta (PRD), which is located in a subtropical zone with different climate and soil conditions. Therefore, we must determine the critical environmental factors that influence the transfer of Cd in the soil-vegetable system in the PRD region. Such knowledge can improve the safety of vegetables. In this study, the soil geochemical properties are investigated to explore the key soil factors that control the uptake of Cd by flowering cabbage, a popular leaf vegetable in China, from soils in the PRD region. The Cd contents in vegetables were most positively correlated to soil oxalate-Cd ($p < 0.01$), which indicates that amorphous Cd is the most available form for uptake into the cabbages. With the characteristics of rich in Fe oxide and Al oxide in the PRD soils, soil Fe and Al oxides were found to be the most relevant to the transfer factors of Cd from the soils to the cabbages. Soil secondary minerals are the key factor that affects the transfer of Cd, thereby influencing the migration and fate of Cd in soil-cabbage systems, with DCB-Fe significantly decreasing the Cd accumulation in cabbages. Additionally, models were developed to predict the enrichment of Cd in flowering cabbages, in which oxalate-Cd, DCB-Fe, and NaOAc-Al in soils were determined to be the most important factors that affect the Cd enrichment in flowering cabbages. In this study, we determine the important role of soil secondary minerals in affecting the transfer of Cd in soil-cabbage systems in the PRD. These observations are important to evaluate the accumulation of Cd in vegetables in subtropical zones.

1. Introduction

Cadmium (Cd) is well known as a nonessential heavy metal and extensively exists in soils from natural and anthropogenic sources (Mireles et al., 2004; Liang et al., 2013; Wang et al., 2013, 2015). The wide distribution of Cd in contaminated soils has caused profound environmental and health issues (Godt et al., 2006; Jihen et al., 2008; Brus et al., 2009; de Souza Predes et al., 2014). Furthermore, Cd can be slowly and consistently transferred from polluted soils into food crops to elevate Cd exposure to human beings in the long term through the food chain (Brus et al., 2009; Rizwan et al., 2016), leading to food safety incidents in many countries, such as Japan and China.

Humans can discontinue farming and cease the cultivation of fields with heavily Cd polluted soils to avoid the apparent harm from metals on human health. However, large areas of slightly and moderately Cd-polluted soils must be continually used to plant food crops, such as rice and vegetables, because of a lack of land resources in many developing countries (Hao et al., 2009; Okedeyi et al., 2014; Yu et al., 2015). Although people are increasingly

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* Corresponding author.
E-mail address: cefbli@soil.gd.cn (F. Li).

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concerned on food safety because of environmental pollution of Cd in China, the consumption amount and daily intake of these crops from slightly and moderately Cd-polluted areas are still large because of a shortage of available alternatives, especially rapidly growing leafy vegetables (Zhu et al., 2016).

The Pearl River Delta (PRD) is located in southern Guangdong Province, China (23°40'~21°30′N, 112°~115°30′E), which is suitable for agriculture with its typical subtropical monsoon climate. The PRD is one of the regions with the fastest economic growth and significant urban and industrial development in China, which inevitably resulted in the pollution of heavy metals, including Cd, in the soils (Wong et al., 2002; Cai et al., 2015; Hou et al., 2014). Furthermore, these slightly polluted soils are continuously used as agricultural land in the long term because of the extremely limited soil resources in China. The PRD region is one of the important national agricultural bases with 13,357 km^2 of farmland, of which 4829 km^2 is used for vegetable production (Chang et al., 2014a). Vegetable production can reach 1.13×10^7 tons annually (Zhang et al., 2014a). Inevitably, high concentrations of heavy metals, including Cd, have been detected in leafy vegetables in the PRD region (Chang et al., 2014a). However, these slightly polluted agricultural soils are markedly difficult to be remediated at a large scale with engineering methods because of both high costs and the potential ecological destruction of the soils. Therefore, we must elucidate the transfer mechanisms of Cd through soil and the soil properties alongside the climatic conditions and cultivars of vegetables to reduce the transfer of heavy metals from soils to vegetables.

A better understanding of the influences of Cd accumulation in vegetables is critical to produce vegetables with low amounts of Cd. Many studies proved that the heavy metal contents in vegetables are not simply correlated with the total concentrations of the soil, sometimes not even showing any direct correlation (Niesiobędzka, 2012; Raguž et al., 2013). Actually, despite the variations among different heavy metals and plant species, the soil properties, including the pH conditions, organic matter (OM) content, cation exchange capacity (CEC), and mineral and oxide contents, may play specific roles in influencing the enrichment of heavy metals in crops (Baltrenaite and Butkus, 2007; Niesiobędzka, 2012; Ding et al., 2013; Soriano-Disla et al., 2014; Liu et al., 2015a). Among these geochemical properties, the pH is considered the most critical factor for its significant role in affecting ion mobility and bioavailability, especially in soils in temperate zones, according to the viewpoints of European soil researchers (Baltrenaite and Butkus, 2007; Niesiobędzka, 2012). Additionally, OM and minerals are often applied when the transport and transfer of heavy metals in soils is modeled because of their relationship with the CEC (Baltrenaite and Butkus, 2007; Niesiobędzka, 2012; Zhang et al., 2014a).

The climate and soil conditions in the PRD region, which is located in a subtropical zone, are largely different from those of temperate zones. Additionally, the soil properties in subtropical zones are different from those in temperate zones. For example, ferralsols are the main soil type in subtropical zones, and the soil mineralogy is dominated by large amounts of Fe and Al minerals (Paduani et al., 2009; Bortoluzzi et al., 2015); therefore, the environmental behavior of soil metals is considered to be associated with these minerals (Yin et al., 2016; Chang et al., 2016). The models and key factors of Cd uptake from soil have been studied by using various plants in different types of soils (Tudoreanul and Phillips, 2004); however, the published results are hard to directly apply to regional soils in the PRD region, which is a different region in a typical subtropical monsoon climate whose soils are rich in Fe and Al oxides. The heavy metal contents in vegetables in the PRD region were found to be closely correlated with the concentrations in the soils (Chang et al., 2014a, 2014b; Zhang et al., 2014a), and understanding the crucial factors that limit the transfer of heavy metals from soils to crops is important to relieve the contamination of metals and enhance the safety of agricultural production. Previous reports found that cadmium (Cd) had higher potential to be transferred from soils into vegetables than other heavy metals, e.g., mercury (Hg), lead (Pb), chromium (Cr), arsenic (As), etc., and leafy vegetables such as flowering cabbages are among the most accumulative species (Zhang et al., 2014a; Chang et al., 2014b). However, Cd enrichment studies in soil-crop systems were mostly limited to pot experiments and few field experiments, while regional research in this field is rare, especially studies that focused on exploring the critical factors that affect the transformation of Cd in soil-crop systems in subtropical zones such as the PRD region.

Therefore, the transfer of Cd in soil-vegetable systems and the enrichment of Cd in flowering cabbages, a popular local leaf vegetable, are comprehensively studied in terms of the soil conditions and properties by examining 112 pairs of soils and flowering cabbages throughout the PRD region. Multivariate statistical analyses are used to explore the critical soil factors that affect the transfer and enrichment of Cd in cabbages. Additionally, suitable models that are based on the evaluated factors that affect the transfer of Cd are developed to potentially predict Cd enrichment in vegetables in the PRD region.

2. Materials and methods

2.1. Study area and sample collection

In total, 112 paired samples of soils and flowering cabbages (*Brassica campestris* ssp. *Parachinensis*) were collected from protected prime farmlands or vegetable production bases far from cities and industrial areas throughout the PRD region during the typical planting and collection season (September–November) for this vegetable in 2011 (Fig. 1) (Li and Wang, 2007). Each soil sample was a mixture of 4 sub-samples that were collected from the surface soil (0–20 cm) over an area of approximately 10 m^2, with each sub-sample collected in a 25 cm × 25 cm area. The corresponding cabbage sample was a mixture of shoots and leaves from 4 mature plants in each 10-m^2 soil sampling area. Both the soil and cabbage samples were sealed in individual polyethylene bags after collection and then transported to the laboratory within 6 h. After we removed the gravels, leaves and roots, the soils were air-dried at room temperature and then sieved through 80 meshes (~0.2 mm) before storage and analysis. Withered and decayed tissues were removed from the cabbage samples, and then the edible portions were washed twice in tap water before being dried in an oven at 60 °C. Afterwards, the samples were crushed with a wooden hammer in a carnelian mortar, passed through 80 meshes, and packed for storage and analysis.

2.2. Analyses of soil properties

The total Fe, Al, Ca, Mn, Mg, Na, and K contents of the soils were measured on an inductively coupled plasma-atomic emission spectrometer (ICP-AES, Optima 3300DV, Perkin Elmer, USA) after the soils were digested in HNO_3-$HClO_4$-HF (Pansu and Gautheyrou, 2006). The total Si content after digestion was measured by using the silicon-molybdenum blue colorimetric method. The total Si, Fe, Al, Ca, Mn, Mg Na, and K contents of the soils were recalculated and reported as the equivalent oxide contents of SiO_2, Fe_2O_3, Al_2O_3, CaO, MnO_2, MgO, N_2O, and K_2O, respectively. Amorphous Fe (oxalate-Fe), complexed Fe, and dithionite-citrate-bicarbonate (DCB)-Fe forms in the soils were extracted by using three different extractions, i.e., oxalic acid-ammonium oxalate buffer solution at a pH of

3.2, alkaline sodium pyrophosphate at a pH of 8.5, and dithionite-citrate-biocarbonate buffer solutions, respectively (Pansu and Gautheyrou, 2006).

2.3. Measuring the Cd concentrations

The sieved soil samples were mixed with concentrated HNO_3/$HClO_4$/HCl (87:13:10, v/v/v) for digestion and the cabbage samples were digested by a mixture of HNO_3/$HClO_4$/H_2O_2 (87:13:10, v/v/v) before measuring the total Cd concentrations. Different species of Cd in the soils, including amorphous-Cd, DCB-Cd, $CaCl_2$-Fe, NaOAc-Cd, and DTPA-Cd, were extracted by using oxalic acid-ammonium oxalate, DCB, $CaCl_2$ solution, acetic acid-sodium acetate, and diethylene triamine pentacetate acid, respectively (Gleyzes et al., 2002), following previously described methods (Li et al., 2000). The Cd concentrations were then determined by a graphite furnace atomic absorption spectrophotometer (AAS, SA-10, Titan, China).

2.4. Data analyses

A comprehensive statistical analysis was performed to determine how the soil geochemical properties managed the transfer of Cd from the soils into the cabbages. Pairwise correlations between the soil geochemical properties and multivariate statistical analyses of the geochemical properties and accumulated Cd in cabbages were performed with R, the Vegan package and FactoMineR (Oksanen, 2011). The regressed correlations were developed by using simple, straight-forward stepwise multiple regression analyses with SPSS 13.0.

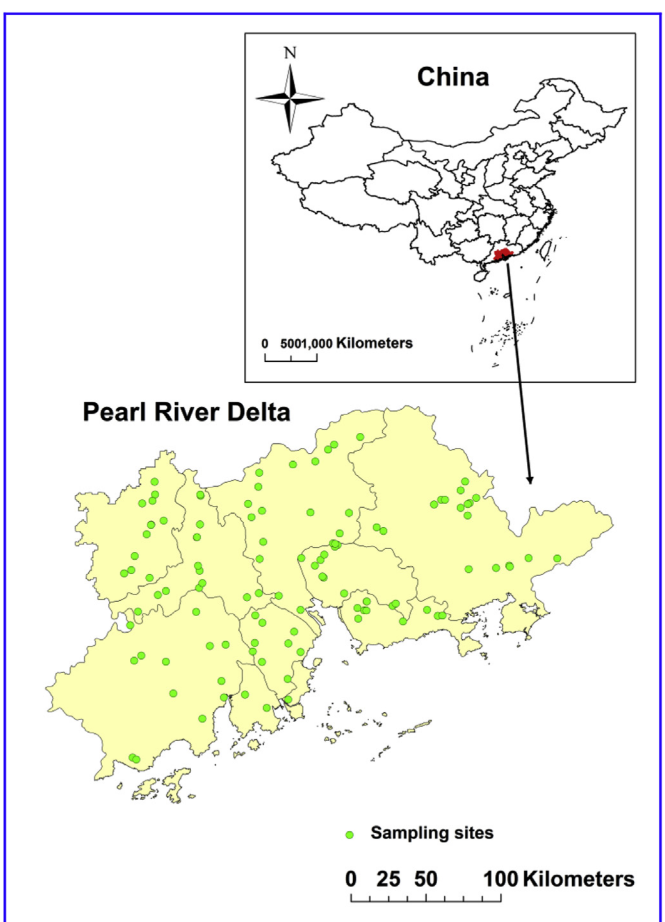

Fig. 1. Map of the study area and sampling sites.

3. Results and discussion

3.1. Geochemical properties of the soils in the PRD region

The detailed values of the physicochemical properties of the soil samples from the PRD are listed in Table S1. The average values of the physicochemical properties were calculated, and the contents of the major soil elements are provided in Table 1. Briefly, the average pH (H_2O) and pH (KCl) were 6.12 ± 0.96 and 5.56 ± 1.04, with ranges of 3.86–8.07 and 3.26–7.40, respectively. Most of the soil samples were acidic, with only 26.8% of the soils having a pH (H_2O) higher than 7 and 1 sample higher than 8.0. These results may suggest that the amount and mobility of ionic Cd would probably have been increased because lower pH values often enhance the bioavailability of soil metals, including Cd (Niesiobędzka, 2012; Liu et al., 2015a).

The total soil organic carbon was 0.59–2.76%, with an average value of 1.26 ± 0.48%, and the OM content was 0.97–5.51%, with an average value of 2.56 ± 0.930%. These results were consistent with others' reports of the PRD area that the soils in this region presented relatively low OM levels (Zhang et al., 2014a, 2014b; Cai et al., 2015). Correspondingly, the CEC of the soil was also low (4.09–21.1 cmol/kg), with an average value of 12.67 ± 4.59 cmol/kg. As previous studies demonstrated, the OM and CEC were always negatively proportional with the bioavailable Cd and other heavy metals (Spence et al., 2014; Liu et al., 2015a), which indicates the high transfer potential of Cd in the studied soils.

Soil weathering and leaching were relatively high in the PRD because of this area's warm, moist climate. Easily mobile elements, such as K^+, Na_+, Ca^{2+}, and Mg^{2+}, were consistently leached, while hardly mobile oxides, including Fe_2O_3, Al_2O_3 and MnO, accumulated during the weathering process. These hardly mobile oxides, e.g., Fe_2O_3, Al_2O_3 and MnO, had high contents of 8.37–121.6 g/kg, 46.0–247.8 g/kg, and 0.00–1.48 g/kg (average values of 38.55 ± 21.70 g/kg, 136.8 ± 46.91 g/kg, and 0.405 ± 0.251 g/kg), respectively, which would affect the main characteristics of the PRD's soils. As previously reported, soil desilication and enrichment in Fe-oxides and Al-oxides were universal in southern China (Cai et al., 2015), which affect the uptake of heavy metals by growing plants (Kabata-Pendias, 2004; Chang et al., 2014a).

3.2. Cd contents and species in the soils

The concentration ranges of Cd in 112 soil samples from the PRD were 0.05–1.25 mg/kg, with an average value of 0.36 ± 0.25 mg/kg (Fig. 2a). The Cd concentrations in all the soils were higher than the background concentrations of soil Cd in Guangdong Province (0.04 mg/kg) (Zhang et al., 2011). Compared to the maximum permissible concentrations of Cd for agricultural soils, which is 0.30 mg/kg (SEPAC, 1995), 52 of the 112 soil samples exceeded the safety standard. The high variations among the different soil samples suggest great heterogeneity throughout the PRD region.

The mobility and bioavailability of Cd were more related to the Cd species than the total content (Niesiobędzka, 2012; Raguž et al., 2013). Generally, the physically adsorbed, ion exchange species (e.g., $CaCl_2$ extract) and carbonate species (NaOAc extract) of heavy metals were the most available species for the plants, while DCB-Cd and DTPA-Cd, which are the sum of amorphous, oxidation and crystalline species and the chelation species, respectively, were relatively more stable. The residual species, other than those that were extracted in this study, were the most stable species and were almost never utilized by the plants. According to the results, the Cd in most of the soil samples was relatively stable and not easily

Table 1
Average values of the physicochemical properties and the major soil element contents of the 112 soil samples (calculated from the data in Table S1).

pH(H_2O)	pH(KCl)	TOC (%)	OM (%)	CEC (cmol/kg)	Sand (%)	Silt (%)	Clay (%)
6.12 ± 0.96	5.56 ± 1.04	1.26 ± 0.44	2.56 ± 0.93	12.7 ± 4.59	37.4 ± 22.6	36.3 ± 13.3	26.3 ± 11.0
Fe_2O_3	Al_2O_3	SiO_2	MnO	CaO	MgO	Na_2O	K_2O
38.6 ± 21.7	136.8 ± 46.9	679 ± 98.5	0.405 ± 0.351	7.36 ± 6.02	5.96 ± 4.99	2.09 ± 1.03	19.9 ± 7.48

available to the plants. The concentrations in individual extracts followed the order of DTPA-Cd > DCB-Cd > Oxalate-Cd > NaOAc-Cd > $CaCl_2$-Cd. Thus, DTPA-Cd and DCB-Cd comprised 41.2% and 40.9% of the total soil Cd, respectively, while the other three species comprised less than 20% of the total. These results are different from others' reports that exchangeable Cd ($CaCl_2$-Cd) and carbonate-bound Cd (NaOAc-Cd) are dominant in soils (Wang et al., 2012a; Zhang et al., 2014a), which could be attributed to the naturally low OM content and CEC but abundant Fe or Mn oxides in the studied soils.

3.3. Cd transfer from soils to flowering cabbages

The Cd concentrations in the cabbage samples varied from 0.128 mg/kg dw to 2.80 mg/kg dw, with an average value of 0.627 ± 0.475 mg/kg dw. Compared to the maximum permitted level of Cd in leafy vegetables (0.20 mg/kg dw) in China (National Food Hygiene Standard of China, GB2762-2012), the Cd concentrations in 65.2% of the studied flowering cabbages were above the national maximum level. The transfer factors (TF) of Cd from the soils to the flowering cabbages were obtained by dividing the vegetable Cd content with the soil's total Cd concentration (Zhang et al., 2014a). The TF values were 0.39–11.8, with an average of 2.32 ± 1.75, in which 85 of the 112 samples had a TF value higher than 1 (Fig. 2b). This result indicates the significant accumulation of and enrichment of Cd in vegetables in the PRD region, which were attributed to the high availability of Cd in these acidic soils. The spatial distribution of the TF did not show great heterogeneity, except for some extremely high TF values in the samples from the middle and northeastern areas, which did not match the spatial distribution of the Cd concentration in the corresponding soils. Others also reported similar spatial distributions of Cd contents in vegetables from the PRD region (Zhang et al., 2014a). These results were also consistent with our previous finding that Guangzhou and Dongguan, which are located in the middle of the PRD region, had the most serious heavy metal contamination in leafy vegetables (Chang et al., 2014b). Despite the possibly high degree of atmospheric pollution in these areas from high industrialization (Luo et al., 2011; Zhang et al., 2014a), heterogeneous soil properties might be the main cause of the different enrichment extents of Cd in vegetables, which are thoroughly discussed later.

3.4. Correlation between vegetable and soil Cd fractions and soil geochemical properties

A soil's properties are the most important factors that influence the availability and transfer of metals in soil-plant systems and have received much attention in studies of different regional soils. Pearson correlation analysis was conducted to illustrate the correlations among the soil's Cd concentration, the vegetables' Cd accumulation and the properties of the PRD's soils. The results are summarized in Table 2. The vegetables' Cd content was significantly positively correlated with either the soil's total Cd concentration or individual species ($p < 0.01$). The coefficients varied from 0.29 to 0.67 for vegetable-Cd with different soil Cd species, which had the highest correlation coefficient ($r = 0.673$) (Fig. 3A), might have been the most direct factor to the accumulation of Cd in the flowering cabbages, followed by NaOAc-Cd ($r = 0.536$), DTPA-Cd ($r = 534$), $CaCl_2$-Cd ($r = 0.358$) and DCB-Cd ($r = 0.290$). Oxalate-Cd was expected to be the most available for transfer and accumulation by the vegetables.

Generally, basic soil properties such as the pH, OM and CEC are considered to be important factors that influence the transport and transfer of heavy metals in soils, which has been reported in many previous reports (Ding et al., 2013; Soriano-Disla et al., 2014; Chang et al., 2014a). The pH has been declared the most predominant soil factor that controls the transfer of heavy metals from soils to plants, especially in soils in temperate zones such as Europe (Baltrenaite and Butkus, 2007; Niesiobędzka, 2012). However, no significant correlation was observed between the soil pH and vegetable Cd

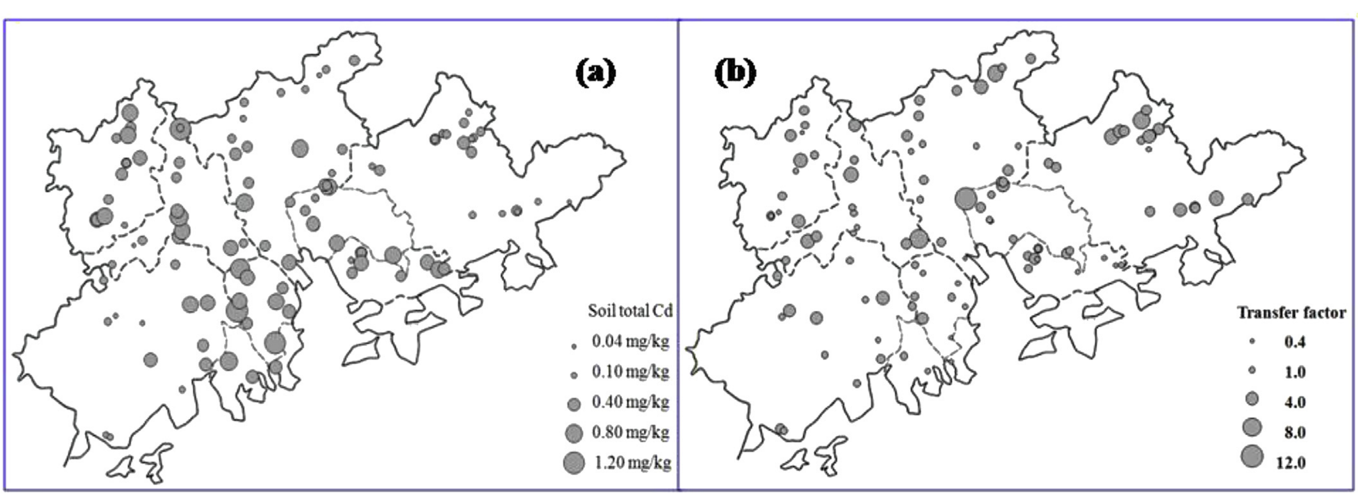

Fig. 2. Spatial distribution of the sampling points, soil total Cd concentration (upper) and vegetable-Cd transfer factor (lower).

content in this study (Table 2), which contrasts the previous statement of the pH's role in soils in temperate zones. As introduced above, Al minerals are the main minerals in subtropical soils, and Al (hydr)oxides were indeed the most abundant components in the studied soils from the PRD region (Table 1). Aluminum ions on the edges of faces (100 and 110 planes) of Al (hydr)oxides usually have unsatisfied positive charges, and these surface Al sites can act as Lewis acid sites (Nassar et al., 2011; Liu et al., 2012). Furthermore, exchangeable aluminum is considered to be predicative of a soil's pH conditions (Hochman et al., 1992) and is more reliable for indicating pH conditions, especially in Al-rich soils. The effect of pH (H_2O) on the transfer of Cd was highly disturbed by these solid acid sites and exchangeable aluminum in Al (hydr)oxides because of the high amounts of Al minerals in the studied soils. Therefore, the soil's pH did not significantly affect the transfer of Cd in the soil-vegetable system.

The most significant and highest correlation for the pH and soil Cd fractions was found with $CaCl_2$-Cd ($r = -0.689$ and $r = -0.746$ for pH(H_2O) and pH(KCl), respectively, $p < 0.01$). This result is reasonable because acidic condition often favored the solubilization of heavy metal ions (Spence et al., 2014). Another significant positive correlation was also observed between the pH and NaOAc-Cd ($r = 0.350$ and $r = 0.380$ for pH(H_2O) and pH(KCl), respectively, $p < 0.01$). Soils with higher pH had higher amounts of carbonate-bound Cd because carbonates would be more easily dissolved in soils with lower pH, and the prolonged dissolution of carbonates in soils with higher pH would lead to more carbonate-bound Cd in the soils (Spence et al., 2014). These two Cd species, i.e., $CaCl_2$-Cd and NaOAc-Cd, were determined to have contributed less Cd to the vegetables than oxalate-Cd (Table 2).

No significant correlation could be identified between the OM and TOC of the soils and the vegetable Cd content, while the most significant and highest correlation coefficients were found with NaOAc-Cd and DTPA-Cd ($r = 0.336$–0.478, $p < 0.01$) (Table 2). Additionally, the OM was significantly correlated with the soil's DCB-Cd ($r = 0.336$, $p < 0.01$). We can easily understand why the carbonate-bound Cd (NaOAc-Cd) was correlated with the TOC content. Soil OM is important for the adsorption and organo-metal complexation of heavy metals (Kabata-Pendias, 2004; Spence et al., 2014); thus, higher OM contents would surely increase the oxidized-Cd (OM-bound Cd, which was included in DCB-Cd) and chelation fractions (DTPA-Cd).

Another important variable, the CEC, was significantly positively correlated with all the vegetable and soil Cd contents and fractions, except for $CaCl_2$-Cd (Table 3). In addition to OM, the CEC is an important factor in terms of the soil adsorption of heavy metals (Guo et al., 2013). Lower CECs are expected to translocate Cd in the soil, while higher CECs would increase the amount of bound Cd in

Table 2
Pearson correlation between the Cd contents in soils and vegetables and the soil properties.

	Vegetable-Cd	Soil Total-Cd	CaCl2-Cd	NaOAc-Cd	Oxalate-Cd	DCB-Cd	DTPA-Cd
Vegetable-Cd	1	0.485(**)	0.358(**)	0.536(**)	0.673(**)	0.290(**)	0.534(**)
pH(H_2O)	−0.052	0.372(**)	−0.689(**)	0.350(**)	−0.08	0.038	0.189(*)
pH (KCl)	−0.144	0.384(**)	−0.746(**)	0.380(**)	−0.122	0.091	0.231(*)
TOC	0.01	0.239(*)	0.087	0.336(**)	0.123	0.237(*)	0.391(**)
OM	0.037	0.354(**)	0.144	0.413(**)	0.232(*)	0.336(**)	0.478(**)
CEC	0.320(**)	0.451(**)	0.175	0.501(**)	0.463(**)	0.506(**)	0.554(**)
sand	−0.417(**)	−0.329(**)	−0.261(**)	−0.357(**)	−0.435(**)	−0.383(**)	−0.350(**)
silt	0.312(**)	0.168	0.174	0.231(*)	0.296(**)	0.212(*)	0.191(*)
clay	0.405(**)	0.423(**)	0.280(**)	0.395(**)	0.465(**)	0.468(**)	0.433(**)
Fe_2O_3	0.493(**)	0.457(**)	0.185	0.476(**)	0.470(**)	0.421(**)	0.477(**)
Al_2O_3	0.182	0.207(*)	0.081	0.252(**)	0.224(*)	0.181	0.249(**)
SiO_2	−0.218(**)	−0.416(**)	−0.064	−0.432(**)	−0.296(**)	−0.338(**)	−0.409(**)
MnO	0.350(**)	0.449(**)	0.095	0.471(**)	0.338(**)	0.371(**)	0.519(**)
CaO	0.135	0.554(**)	−0.390(**)	0.524(**)	0.093	0.229(*)	0.393(**)
MgO	0.554(**)	0.546(**)	0.146	0.583(**)	0.552(**)	0.534(**)	0.563(**)
Na_2O	0.264(**)	0.132	0.161	0.201(*)	0.276(**)	0.209(*)	0.295(**)
K_2O	0.136	0.048	0.180	0.103	0.126	0.122	0.111
$CaCl_2$-Fe	−0.016	−0.026	0.237(*)	0.020	0.208(*)	0.235(*)	0.067
NaOAc-Fe	0.060	−0.200(*)	0.289(**)	−0.082	−0.013	−0.153	−0.089
Oxalate-Fe	0.353(**)	0.439(**)	0.185	0.521(**)	0.488(**)	0.425(**)	0.543(**)
DCB Fe	−0.291(**)	0.368(**)	0.082	0.376(**)	0.366(**)	0.385(**)	0.374(**)
DTPA-Fe	0.088	−0.130	0.604(**)	−0.102	0.228(*)	0.132	0.014
$CaCl_2$-Al	0.061	−0.110	0.315(**)	−0.132	0.129	0.174	−0.031
NaOAc-Al	0.284(**)	−0.092	0.563(**)	−0.038	0.221(*)	0.043	0.028
Oxalate-Al	0.264(**)	0.409(**)	0.091	0.345(**)	0.332(**)	0.361(**)	0.362(**)
DCB-Al	0.159	0.158	0.083	0.059	0.188(*)	0.212(*)	0.114
DTPA-Al	−0.043	−0.375(**)	0.574(**)	−0.370(**)	0.009	−0.088	−0.253(**)
$CaCl_2$-Si	−0.117	0.344(**)	−0.313(**)	0.345(**)	−0.084	0.052	0.333(**)
NaOAc-Si	0.024	0.482(**)	−0.464(**)	0.489(**)	0.014	0.065	0.307(**)
Oxalate-Si	0.280(**)	0.644(**)	−0.216(*)	0.669(**)	0.296(**)	0.305(**)	0.576(**)
DCB-Si	0.033	0.328(**)	−0.147	0.366(**)	0.037	0.053	0.362(**)
DTPA-Si	−0.264(**)	−0.020	−0.102	0.009	−0.212(*)	−0.191(*)	−0.013
$CaCl_2$-Mn	0.215(**)	0.056	0.795(**)	0.057	0.242(*)	0.199(*)	0.190(*)
NaOAc-Mn	0.291(**)	0.547(**)	0.141	0.575(**)	0.323(**)	0.459(**)	0.651(**)
Oxalate-Mn	0.276(**)	0.522(**)	−0.033	0.525(**)	0.296(**)	0.353(**)	0.526(**)
DCB-Mn	0.328(**)	0.592(**)	0.014	0.557(**)	0.415(**)	0.442(**)	0.603(**)
DTPA-Mn	0.282(**)	0.451(**)	0.368(**)	0.424(**)	0.400(**)	0.508(**)	0.535(**)

*Correlation is significant at the 0.05 level (2-tailed).
**Correlation is significant at the 0.01 level (2-tailed).

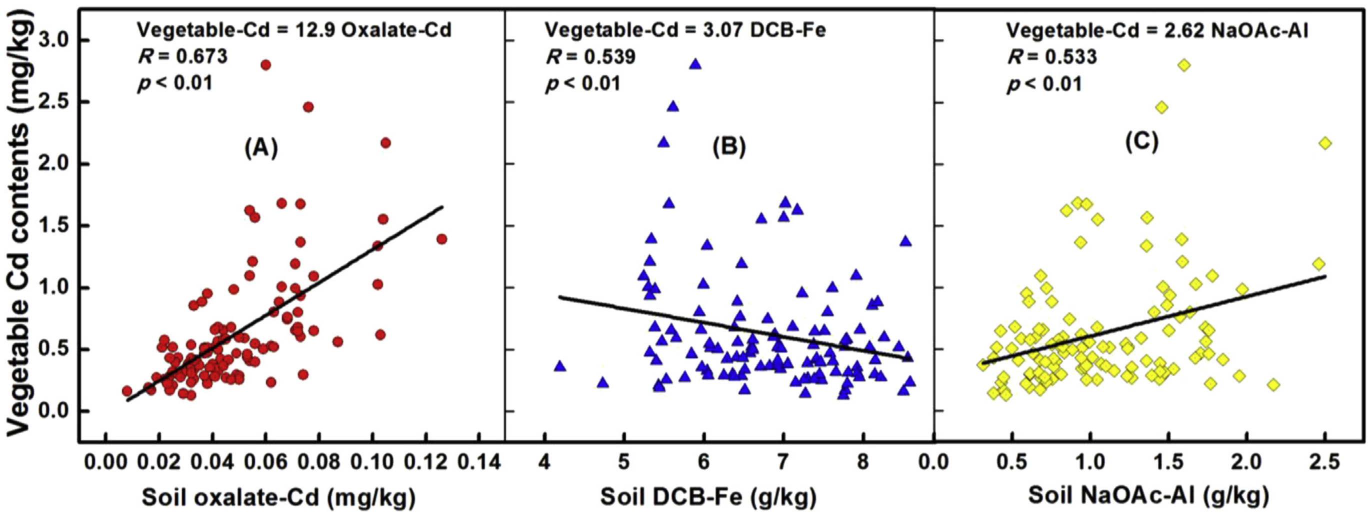

Fig. 3. Correlation between the accumulated Cd in the cabbages and the (A) soil oxalate-Cd, (B) Soil DCB-Fe, and (C) NaOAc-Al contents.

the soil and thus decrease the amount of Cd that is available to vegetables (Spence et al., 2014). However, this scenario obviously did not occur in this study. The soil-plant transfer of Cd is a complex process that is affected by multiple factors, and active factors such as the pH, OM and CEC also interact with each other. The effects of the low CEC values in the studied soil samples may have been covered by other actions, while the observed positive correlation might be a spurious correlation. Meanwhile, the soil samples with higher OM contents were mostly from contaminated areas in the PRD region (i.e., central and northwestern areas), so the higher plant Cd contents could probably be attributed to the original pollution rather than the higher OM content, which may require further investigation to obtain an assured explanation.

The most interesting finding is the critical role of mineral constitutes towards Cd accumulation in the cabbages. Soil minerals are important soil constitutes that affect the motilities of soil elements. For example, Fe and Mn (hydr)oxides were reported to be important media that retain high amounts of Cd in soil through sorption (Wang et al., 2012a). Our study also indicated the high correlations between vegetable Cd accumulation and the main soil minerals. Among the main oxides, including Fe, Al, Si, Mn, Ca, Mg, Na and K oxides, MgO and Fe oxides may have been the most important minerals in terms of vegetable Cd accumulation, as suggested by their high correlation coefficients ($r = 0.554$ and $r = 0.493$, respectively, $p < 0.01$) (Table 3). A similar correlation was found for oxides with soil Cd and different species ($p < 0.01$). Specifically, DCB-Fe and NaOAc-Al were the two species that had significant correlations with the vegetable-Cd content, as suggested by their correlation coefficients ($p < 0.01$) (Fig. 3B and C).

3.5. Multiple factor models of Cd transfer from soils to flowering cabbages in the PRD

The transfer factor (TF) of metals is expressed as the ratio between the metal concentration in the vegetables and the metal concentration in the soils, which is a very important parameter to study the mobility and accumulation of heavy metals in soils and their bioavailability in soil-vegetable systems (Rattan et al., 2005; Nabulo et al., 2011; Wang et al., 2012b; Raguž et al., 2013). Therefore, TFs were used to evaluate the soil Cd mobility and vegetable Cd accumulation. The vegetables might have been optimized via their reasonable layout and the selection of plant species or cultivars with relatively small concentrations in the edible portions of the vegetables. Therefore, exploring the critical factors that determine the transfer availability of Cd from soils to vegetables according to the above factors is very important.

Transfer models of soil-vegetable systems for heavy metals have been investigated by many researchers (Dudka et al., 1996; Jung and Thornton, 1997; Zhao et al., 2010), who focused on soil heavy metal contents and their basic physicochemical properties (e.g., OM, pH) in terms of the heavy metal contents in crops. Actually, the species of heavy metals in the soils were the key factor for heavy metal transfer models in soil-vegetable systems. In particular, most of the existing models mainly involved lab or field experiments that controlled the metal concentrations and species. Multiple factors may drive the transfer of Cd from soils according to their contributions in the models, such as the soil's basic physicochemical properties, nutrient elements, and secondary minerals. Therefore, Cd transfer models of soil-vegetable systems were developed based on the soil Cd species and Fe, Al and Mn oxides to analyze the correlation analysis results in the above section. Generally, three types of models, including linear models, exponential models and logarithmic models, can describe the transfer of heavy metals from soils to plants. Moreover, linear models, logarithmic models or a combination of the two are preferentially utilized for relatively low amounts of heavy metals in the soil (Dudka et al., 1996). In this study, the concentrations of Cd in the soils were relatively low, and the vegetable Cd and soil Cd demonstrated a good correlation. Therefore, the multiple step regression models were developed according to the relationship between the soil Cd data (or log-transformed data) and vegetable Cd data.

The basic parameters in the Cd regression model were the soil Cd concentration (Cd_{soil}) and plant Cd concentration (Cd_{plant}). Other parameters, such as the soil Cd species, soil basic physicochemical properties, and Fe/Mn oxide contents, were stepwise added into the equations to study their contributions and correlations. Therefore, the following equations for Cd in plants were gradually added: (1) the total soil Cd; (2) species of soil Cd; (3) species of soil Cd plus soil physicochemical properties; (4) species of soil Cd, soil physicochemical properties and total Fe, Al, and Si oxides; and (5) species of soil Cd, soil physicochemical properties and species of Fe, Al, and Si oxides. Only the factors that were correlated with the vegetable-Cd at the 0.01 and 0.05 confidence levels (Table 2) were included in the regression to avoid spurious correlations.

The obtained regression models of Cd in plants are listed in Table 3. Different performances were recorded for models with

Table 3
Regression model of Cd in flowering cabbages and the soil system (only the factors that were correlated with vegetable-Cd at the 0.01 and 0.05 confidence levels were included in the regression).

No.	Main factor	Fitting equation	R^2	S.E.	*p*
1	Total soil Cd	$\text{Log Cd}_{plant} = -0.063 + 0.425 \log \text{Cd}_{total\text{-}soil}$	0.251	0.232	<0.01
2	Cd species	$\text{Log Cd}_{plant} = 0.937 + 0.910 \log \text{Cd}_{oxalate\text{-}soil}$	0.456	0.194	<0.01
3	Cd species, soil properties and total Fe	$\text{Log Cd}_{plant} = 1.697 + 0.682 \log \text{Cd}_{oxalate\text{-}soil} - 0.295\ \text{Sand}_{soil} + 0.107\ \text{Fe}_{total\text{-}soil}$	0.484	0.195	<0.01
4	Cd species, soil properties and Fe species	$\text{Log Cd}_{plant} = 1.341 + 0.864 \log \text{Cd}_{oxalate\text{-}soil} - 0.139\ \text{Fe}_{DCB\text{-}soil} + 3.17\ \text{Fe}_{oxalate\text{-}soil}$	0.541	0.182	<0.01
5	Cd species, soil properties and Al species	$\text{Log Cd}_{plant} = 0.857 + 0.924 \log \text{Cd}_{oxalate\text{-}soil} - 0.011\text{Sand} + 2.67\text{Al}_{NaOAc\text{-}soil} - 2.76\ \text{Al}_{DTPA\text{-}soil}$	0.535	0.187	<0.01
6	Cd, Fe and Al species	$\text{LogCd}_{plant} = 0.948 + 0.911 \log \text{Cd}_{oxalate\text{-}soil} - 0.336\ \text{Fe}_{DCB\text{-}soil} + 0.272\text{Al}_{NaOAc\text{-}soil}$	0.555	0.158	<0.01

different factors. Model 1 exhibited the worst performance and the lowest regression coefficient value ($R^2 = 0.251$), which suggests that the total soil Cd had the lowest correlation with the accumulated Cd in the plants. The regression coefficient greatly increased to $R^2 = 0.456$ and the S.E. reduced to 0.194 when the total Cd content in the soil ($Cd_{total\text{-}soil}$) was replaced by the soil oxalate-Cd contents ($Cd_{oxalate\text{-}soil}$), which had the highest regression coefficient among the various species of soil Cd. The oxalated-Cd was mainly the Cd content combined with amorphous Fe in the soil via physical adsorption and simple chemical bonding, which is also known as “active” Cd (Guo et al., 1997). Model 2 suggested that the oxalate-Cd, not the total soil Cd or other Cd species, was the most available species that collected by the flowering cabbage in the soils. Furthermore, the soil oxalate-Cd had the best correlation with the Cd content in the plants (Table 3). When the soil physicochemical properties with their previously determined correlations (Table 2) were included in the equation, the soil sand contents and total Fe were found to improve the performance, with R^2 increasing from 0.456 to 0.4841 (model 3), which suggests that the soil Fe had an apparent relationship with the plants' uptake of Cd from the soils.

Soil Fe and Al oxides are important minerals to stabilize and adsorb heavy metals, especially in mineral-rich soils (Manceau et al., 1992; Liu et al., 2015b, 2016), such as the studied soils in the PRD region. The selective adsorption of heavy metals on the surface of oxides influences the geochemical cycle of heavy metals in soil-water-plant systems. Our results also indicate that the Cd accumulation in flowering cabbages showed a positive correlation with Fe species (DCB-Fe and oxalate Fe) and Al species (NaAc-Al and oxalate-Al), which suggests that these Fe species and Al species played important roles in influencing the Cd accumulation in the plants in the PRD. The soil Fe species were gradually included in model 4, and the inclusion of DCB-Fe and oxalate-Fe increased the regression coefficient ($R^2 = 0.541$) when only considering species of Fe, Al, Si, and Mn. In terms of the Al species, soil properties and Cd species, the inclusion of NaOAc-Al and the soil sand content increased the regression coefficient ($R^2 = 0.535$) (model 5).

When the correlated Cd, Fe, and Al species were adopted, the regression coefficient achieved the largest value ($R^2 = 0.552$) with the lowest standard error (0.158) (model 6). Table 3 shows that all 6 regression models reached high significance levels ($p < 0.001$), and the order of the performances of the equations from best to worst was model 6, model 4, model 5, model 3, model 2, and model 1. Furthermore, the regression coefficient of model 6 was higher than those of the other models, which suggests that model 6, including the oxalate-Cd, DCB-Fe, and NaOAc-Al factors, was the best regression model for flowering cabbages to accumulate Cd from the soils in the PRD region.

3.6. *Soil critical factors that influence the transfer of Cd in soil-vegetable systems in the PRD*

The soil factors that affect the transfer of Cd in soil-plant systems are critical to evaluate Cd accumulation in crops to ensure food safety, especially in areas with high Cd concentration levels in farmland soils (Dudka et al., 1996; Brus et al., 2009; Ding et al., 2013; Chang et al., 2014b; Rizwan et al., 2016). Therefore, the soil factors that affect the transfer of Cd by crops have been extensively investigated in agricultural regions around the word, e.g., Europe and China. In temperate zones, the pH, OM content, and cation exchange capacity (CEC) are the most important and sensitive soil factors that influenced the transfer and availability of heavy metals, including Cd, in soil-crop systems (Baltrenaite and Butkus, 2007; Niesiobędzka, 2012; Liu et al., 2015a). Indeed, the bioavailability of Cd is increased by soil acidification, which increases the concentration of available Cd. Organic matter can reduce the availability of Cd by adsorption or the formation of stable complexes, e.g., with active functional groups of −OH (Emmanuel and Erel, 2002). The CEC is considered a measure of the amount of available adsorption sites for metals, and higher soil CEC values increase the sorption amount and decrease the bioavailability of Cd (Sterckeman et al., 2004; Pauget et al., 2012).

Although many individual studies on specific regions showed significant effects from the soil pH, OM content, and CEC on the transfer of metals in soil-crop systems, predicting metal availability for one area by using relationships that obtained from another area is difficult (Niesiobędzka, 2012). For example, the air and temperatures of different regions were reported to play important roles in Cd accumulation in vegetables (Li et al., 2013), and the critical factors that affect the transfer of Cd would be different in different climate zones. The PRD region is located in South China, which is a subtropical zone with high temperatures and a rainy climate, and this area's soils are usually acidified because of seasonal acid rain and abundant vegetation (Tao et al., 2014) and have high OM contents (He et al., 2008) and low CECs (Cai et al., 2015). As our results showed, the studied soils had an average pH (H_2O) of 6.12, OM content of 2.56% and CEC of 12.67 cmol/kg, with the values ranging from 3.86 to 8.07, from 0.97% to 5.51%, and from 4.09 to 21.1 cmol/kg, respectively (Table S1). The Cd transfer factors did not exhibit correlations with these three soil properties because of the relatively uniformly low pH and CEC of the soils in this study. In other words, the transfer of Cd exhibited similar behavior for all the studied soils in the PRD region, usually occurring under low pH, high OM content and low CEC. Regular differences among different soils could not be observed because of the uniformity of the pH, OM content, and CEC in different soils in the PRD region.

In addition to the soil pH, OM content, and CEC, clay and oxide minerals in soils affect the transfer of metals in soil-plant systems because of their high affinity to metals, although these materials are considered less critical in most regions, as suggested in previous reports (Pauget et al., 2011). Actually, minerals are excellent binding agents for pollutants in soils, especially Fe and Al oxides, which have been confirmed to influence the geochemical behavior of metals in Earth's surface (Pauget et al., 2012; Wang et al., 2013). The soils in the PRD usually have high amounts of Fe and Al minerals because of active soil formation. As our results showed, the average Fe_2O_3 and Al_2O_3 contents in the 112 studied soils were 47.8 ± 21.7 and 152 ± 46.9 g/kg, respectively (Table 1). Therefore, these active soil minerals strongly affected the transfer of Cd in the soil-cabbage systems. According to the regression results and the above discussion, the accumulation of Cd in the cabbages depended on complex Fe and NaOAc-Al, and oxalate-Cd was the most readily accumulated soil Cd species by the cabbage. Indeed, the important roles of Fe and Al minerals in the transfer of metals from soils to plants have been suggested in previous studies. For example, Wang et al. (2013) studied the influence of soil properties on the transfer of Cu and Zn into wheat in China's Yangtze River delta region. Although the soil pH was disclosed to be the most important factor in the accumulation of Cu and Zn in wheat grains, the available Fe in the soil provided conjunct ions and co-transports for transferring Cu and Zn in the soil-wheat system, thus promoting the uptake of Cu and Zn by the wheat. In this study, the available Fe in the soil, together with the soil's pH and S content, exhibited a more important role than other soil parameters in accumulating Cu and Zn in wheat grains. Pauget et al. (2012) studied soil parameters to predict the bioavailability of metals to snails in northern France and observed that the addition of the pH and the iron and aluminum oxide contents enabled variations in Zn assimilation fluxes to be predicted, which indicated that the affinity of Fe and Al oxides in soils was the most important factor that affected the availability of Zn to snails.

Although the soil pH was observed to be the most critical factor that affects the transfer of metals in soil-plant systems, Fe and Al oxides were also important parameters, even for soils in temperate zones such as eastern China and northern France. Fe and Al oxides are expected to play important roles in the transfer of Cd in soil-cabbage systems with mineral-rich soils, such as those in the subtropical PRD region. Oxalate-Cd, DCB-Fe, and NaOAc-Al produced a significant ($p < 0.001$) regression model for the transfer of Cd in soil-cabbage systems with an R^2 value of 0.552 (model 6). While the soil pH, OM content, and CEC are the most important factors that affect the transfer of metals in soil-plant systems in temperate zones, Fe and Al oxides more prominently affect the transfer of metals in soils in subtropical zones, such as the PRD region. According to our results, the roles of Fe and Al oxides in the transfer of metals in soil-plant systems have been underestimated, and the corresponding mechanisms of Fe and Al oxides that affect the transfer of metals should be further investigated in the near future to more accurately predict the accumulation of metals in crops in the PRD region.

4. Conclusions

The concentrations of Cd in 112 pairs of soils and flowering cabbages from the PRD in China and the critical soil geochemical properties that affect the transfer of Cd in soil-cabbage systems were comprehensively studied. The results showed that all the soil samples had higher concentrations of Cd than the background soil Cd concentration in the study area (0.04 mg/kg), with 46.4% of the soil samples having higher concentrations than the maximum concentration limit of Cd for agricultural soils (0.30 mg/kg) in China (SEPAC, 1995). The transfer factors of Cd in these soil-cabbage systems were high because of the acidic soil conditions in the PRD region, with 75.9% of the sample pairs (the soil and cabbage) having transfer factors value higher than 1. Soil minerals greatly affected the bioavailability of Cd in the PRD region, in which Fe and Al oxides were found to be the most relevant to the transfer factor of Cd from soils to cabbages, and the transfer of Cd in soils mostly depended on secondary mineral-associated Cd forms, which influenced the migration and fate of Cd in the soil-vegetable systems. A model that included the oxalate-Cd, DCB-Fe, and NaOAc-Al factors was regressed to predict the accumulation of Cd in cabbages in the PRD region according to the obtained Cd concentrations in the pairs of soils and cabbages and the critical soil factors that affect the transfer of Cd in soil-cabbage systems. Fe and Al oxides in the soil were also important factors alongside the pH and CEC that affected the transfer of Cd in the soil-cabbage system in the subtropical PRD region.

Notes

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2017.08.092.

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