



Spatial Distribution and Bio-accumulation of Cadmium and Lead in Soil, Rice and Vegetables in Typical Pollution Areas, China

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Abstract

The adverse effects of Cd and Pb on human health have been widely reported. While, there were several typical pollution areas in China which has not been investigated comprehensively and deeply. In this study, 475 soil and 472 plant samples from these pollution areas were analyzed for Cd and Pb with ICP-MS. Here, the pollution levels of different areas were analyzed and compared; the spatial distributions in soil, rice and cabbage samples were illustrated in map; and the relationships of Cd and Pb in soil and rice, soil and cabbage were also discussed. The results showed that Cd pollution still existed in some south, central-south and south-east zones except other three zones located in suburb of Tianjin, north-east and north China which might be explained by the higher background values, soil acidity, Karst landscape and metallurgy industry. Comparatively, the Pb concentrations showed lower over-standard rate. Moreover, the spatial distribution of Cd and Pb in rice and cabbage showed similar trends with those of soil samples. Particularly, the average Cd and Pb concentrations in lettuces were much higher than those in other plants. Cd and Pb concentrations in rice and cabbage showed low linear dependence with the concentrations in soil. The results in this study added basic environmental data and would be helpful to future soil remediation of these areas.

Keywords Spatial distribution · Bio-accumulation · Lead · Cadmium · Soil · Rice

Many heavy metals and metallic compounds, cadmium (Cd) and lead (Pb) in particular, are harmful to human health. Cd and Pb metals in water and soil can be transferred to human body by the food chains, and subsequently distribute to many organs, e.g., liver, kidney and lung, by the circulation system, and finally hurt these organs. Consequently, the International Agency for Research on Cancer (IARC) classified cadmium and cadmium compounds into Group 1 (carcinogenic to humans) and inorganic lead compounds into Group 2A (probably carcinogenic to humans) respectively (IARC 2006). Besides, both Cd and Pb are specifically listed in the European Restriction of Hazardous Substances (The

Restriction of Hazardous Substances Directive 2002/95/EC, (RoHS 1), short for Directive on the restriction of the use of certain hazardous substances in electrical and electronic equipment). In addition, many countries also set maximum limit standards of Cd and Pb in soil, rice and vegetables, considering their unambiguous toxicities.

China has been facing a range of environmental challenges associated with soil pollution which were mainly induced by its huge population and fast economy growing. Waste water has been used for irrigation in many cities for decades that was beneficial for water conservation. However, there were large amount of toxic heavy metals in waste water which proved to be pollutants for soil and become a great risk to the health of local residents through food chain (Wang and Zhang 2005; Li et al. 2005; Wang et al. 2005a, b; Xu et al. 2016; Liu et al. 2017; Singh and Kumar 2017; Chen et al. 2018). Additionally, rapid development of local industry including coking, chemistry, metallurgy, manufacture and electronic industry added the emission of heavy metals and further aggravated the pollution. Previous studies have proved that pollution caused by industry did exist in soil and food in many areas (Wang et al. 2005a, b;

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Zhong et al. 2008, 2010; Li et al. 2009; Yuan et al. 2011; Ding et al. 2012).

However, mapping of the contaminated soil sites and evaluation of the pollution status are time consuming and expensive, requiring extensive amounts of geology, hydrology, chemistry and Geographic Information System in environmental contamination, as well as an appreciation of the history of industrial chemistry. Numerous previous studies only focused on the levels, profiles of heavy metals in specific area, for example, some waste water irrigated or metallurgy fields where were not systematic and detailed enough. On the other hand, heavy metals in foods were often neglected in many studies which generally emphasized on the soil. Finally, to our best of knowledge, the pollution status in many areas has not been monitored and evaluated for many years and no investigation has been conducted in this scale ever. So it is very necessary to survey the concentrations, contamination levels, spatial variation and bio-accumulation of heavy metals in these typical pollution zones.

Materials and Methods

475 soil samples (0–10 cm soil layer, 1 kg each) were collected with stainless steel scoops and then stored in PE bags in August 2016 from sampling sites illustrated in Fig. 1. Simultaneously, rice and vegetable samples were collected at the same time. The samples sites from 6 different zones with different soil types, properties (Organic matter content (OM), pH, Cation Exchange Capacity (CEC)) and backgrounds were shown in Fig. 1 and Table 1. Soil samples

in area A ($n=78$) including Hebei province, Tianjin and Beijing, were composed of brown earths, cinnamon soils and castanozems. Most sampling sites were deployed in waste water irrigation fields. Soil samples in area B ($n=99$) of Liaoning province, were composed of brown earths, cinnamon soils, aeolian sandy soils and meadow soils. Most sampling sites were also deployed in waste water irrigation fields. Soil samples in area C ($n=27$) of Shandong province, were fluvo-aquic soils. Most sampling sites were chosen around industrial plants. Soil samples in area D ($n=85$) including Jiangsu, Zhejiang, province and Shanghai; E ($n=88$) from Hubei which were both paddy soils. Soil samples in area F ($n=99$) from Guangxi and Zhejiang province were red earths. Most sampling sites in D, E and F were deployed around industrial plants. And all of the sampling sites have been reported as serious pollution areas before.

Each sample was composed of 20 sub samples within $10 \times 10 \text{ m}^2$. Between sampling the scoop was rinsed by deionized water to preventing cross-contamination. And samples were placed in dark and transported to the laboratory as soon as possible. Soil and rice samples were air dried, ground and sieved through a 50 mesh sieve and stored in $-20 \text{ }^\circ\text{C}$ before analysis. Ultra pure water was used for dilution. All standard solution and internal standards were purchased from Agilent Technology Inc. Microwave-assisted digestion was carried out with a MarsXpress (maximum power: 1200 W) laboratory microwave digestion system (CEM, Matthews, NC, USA). Agilent 7500a ICP-MS (Agilent, USA) was employed for element analysis. Briefly, 0.1 g samples were added into the PTFE pressure bomb and then 1 mL HNO_3 , 1 mL HCl , 1 mL HF were added. The

Fig. 1 Sampling sites and soil types

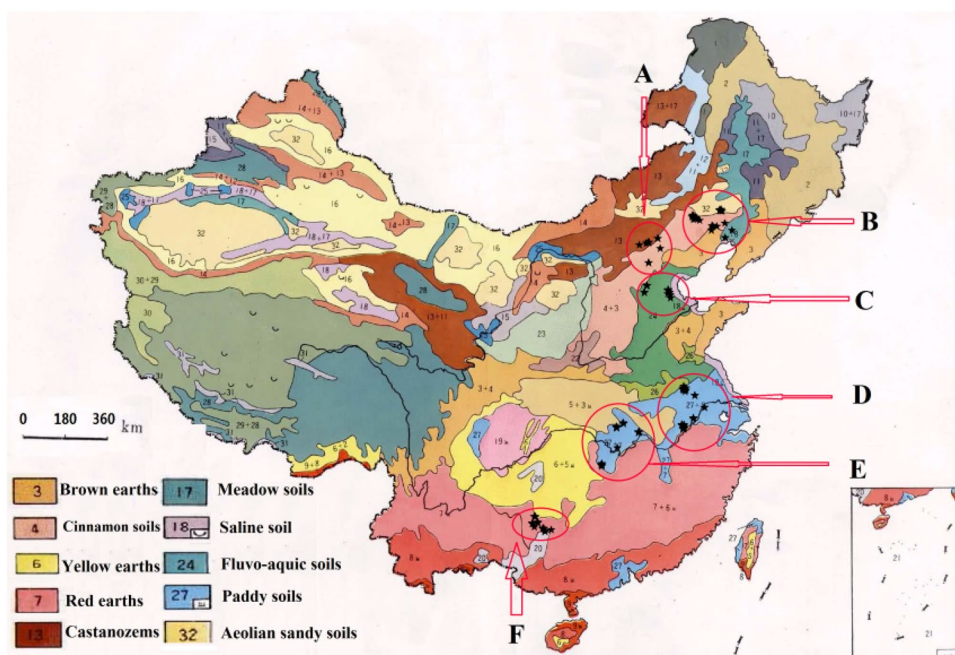


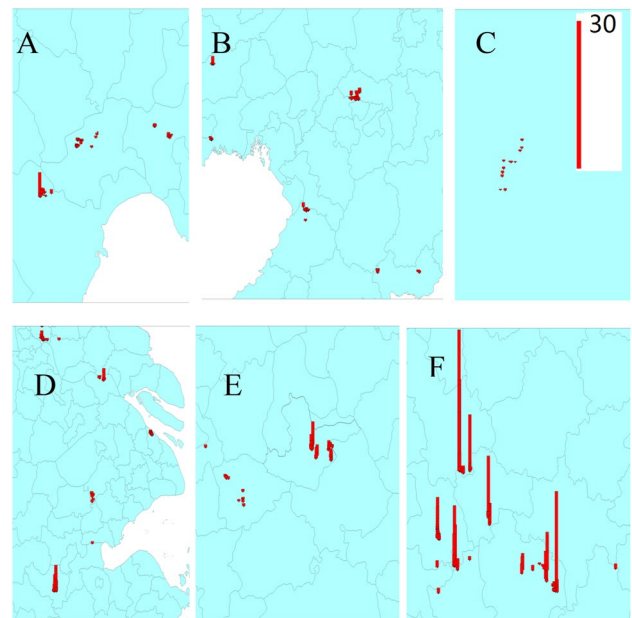
Table 1 Soil properties and Cd, Pb concentrations exceeding standard site numbers

	A (n=78)			Exceeding standard site numbers		B (n=99)			Exceeding standard site numbers	
	OM	pH	CEC	Cd	Pb	OM	pH	CEC	Cd	Pb
Min	0.58	4.95	7.52	2	1	0.59	3.65	5.4	2	0
Max	4.2	8.52	25.15			4.03	7.99	35.38		
Mean	1.92	7.62	19.42			2.1	6.19	17.88		
	C (n=27)			Exceeding standard site numbers		D (n=85)			Exceeding standard site numbers	
	OM	pH	CEC	Cd	Pb	OM	pH	CEC	Cd	Pb
Min	0.71	4.65	15.05	0	0	0.66	4.2	6.63	16	3
Max	3.53	8.78	22.63			6.04	8.98	25.44		
Mean	1.79	7.81	18.01			2.44	7.23	15.85		
	E (n=88)			Exceeding standard site numbers		F (n=98)			Exceeding standard site numbers	
	OM	pH	CEC	Cd	Pb	OM	pH	CEC	Cd	Pb
Min	0.62	4.19	4.57	24	4	0.58	4.77	6.88	61	18
Max	4.74	7.18	47.98			7.88	7.97	16.87		
Mean	2.8	6.12	16.87			3.55	5.68	11.87		

vessels were placed in microwave set at 150 °C for 1 h. After cooling down to the room temperature, shake the vessels for 1 min and digest with the same time procedure again. The supernatant were poured out, drawn through 0.45 µm filter and diluted to 50 mL with water for ICP-MS analysis. The concentrations of heavy metals were calculated by a five-point calibration curve (1 µg/L, 5 µg/L, 10 µg/L, 50 µg/L, 100 µg/L standard solution and 1 mg/L internal standard) with ICP-MS in which linearity of all target elements were higher than 0.99. Samples were measured for three times and the data were considered acceptable when RSD < 20%. Chinese Standard Reference Material-Coal Fly Ash (GBW08401) and GBW07405 (GSS-5) were employed to monitor the performance of the method. The average recovery of Cd was 92.1%, Pb 101.15% (GBW08401); Cd 102.5%, Pb 102.9% (GBW07405).

Results and Discussion

Figure 2 illustrated the spatial distribution of Cd contents in soil samples. Maximum value was illustrated with the height of the scale-bar in the upper-left corner. Concentrations could be observed and compared to each other with the heights. The results showed that Cd concentrations of 2/78, 2/99, 0/27, 16/85, 24/88 and 61/98 sites exceeded the China Environmental quality standard for soils (GB 15618–1995)

**Fig. 2** Spatial distributions of Cd in soil (mg/kg)

Grade III, 1.0 mg/kg in the six areas respectively. Soils in area F were polluted most seriously. There were several reasons which should account for the abnormally serious pollution. Firstly, most sites of F were located in Hechi, Guangxi,

where the background values of Cd in soil were significantly high. It is reported that the background of Cd in soil of Xijiang watershed where Hechi located in were 0.15–0.51 mg/kg, much higher than the average value in Guangxi province (0.0617 mg/kg) and the whole country (0.097 mg/kg) (Wei and Zhang 2016). Many areas without mining plants and metallurgy industry in Hechi, where people were found with longer lives, were still detected higher Cd concentrations than the national standard. Secondly, mining plants and metallurgy industry deteriorated the pollution status. Thirdly, Karst landscape was the main landscape type which account for 66% of the whole landscape in area F. Specifically, there were 162 subterranean streams, 1326 underground water reservoirs in Hechi which would aggravate the metals transfer in soil from the polluted area to the non-polluted area with the existence of water. Lastly, the average pH value of area F was 5.68, which was the lowest among all investigated areas. Acidity of soil enhanced the dissolving of Cd and the dissociation of Cd complex state and subsequently improved the mobility in soils. Moreover, serious Cd pollution also was found in low CEC values in area F, 11.87 mol/kg, which was much lower than the other five areas. Similarly, soils in area E were also polluted seriously, which mainly because most sites were chosen in Daye, Hubei province, a Cu and Fe metallurgy base of China. Relatively lower soil pH (6.12) was also an important factor for the abnormal Cd concentrations. Soils in area D were found to be polluted to some extent. In comparison, slight pollution was detected in area A and B and no pollution was detected in area C.

Pb concentrations showed similar spatial distribution but lower over-standard rate compared with Cd (Fig. 3, Table 1). 1/78, 0/99, 0/27, 3/85, 4/88, 18/98 sites exceeded the China Environmental quality standard for soils (GB 15618–1995) Grade II (pH < 6.5, 250 mg/kg; 6.5 < pH < 7.5, 300 mg/kg; pH > 7.5, 350 mg/kg) in the six areas respectively. The over-standard rates of Pb in soil were lower than Cd suggesting that Pb pollution were not as serious as Cd. The over-standard rates of Pb in area D, E, and F (south of China) were higher than those in area A, B and C (north of China). Particularly, the rate in area F was significantly higher than those in other areas. Xiang (2011) has investigated the migration and transformation of heavy metals such as Sb and Pb in lead and antimony mine smelting area of F, in which study the Pb concentrations ranged from 128 to 39,374 mg/kg with average values 57.7 times of the background (24 mg/kg) proved that lead and antimony mine smelting imposed serious influence on soil. However, the Pb concentrations in this study varied from 19 to 1948 mg/kg with average values of 201 mg/kg, which was much lower than the report before. This divergence may be explained by the differences between sampling sites of the two studies. In detail, Xiang (2011) focused on the smelting area directly, while



Fig. 3 Spatial distributions of Pb in soil (mg/kg)

we focused on the arable soil around the smelting area. Besides, readjustment steps from the government to reduce and shut down the smelting factories might also played an important role.

Figure 4 showed the spatial distributions of Cd (A) and Pb (B) contents in rice samples. Total 247 soils sites were planted with rice, mainly located in area B, north-east of China and D, E, F, south of China. Cd in rice ranged in 0.0111–3.5631 mg/kg, with a mean value 0.2285 mg/kg. Pb ranged in 0.0048–22.980 mg/kg, with a mean value 0.2864 mg/kg (Table 2). According to the National Standard for Food Safety: Maximum Permission Limits of Pollutants (GB 2762–2012), 53 samples exceeded Cd standard (0.2 mg/kg), 54 samples exceeded Pb standard (0.2 mg/kg) respectively. All samples in this study were collected from pollution areas, so the over-standard rate of Cd (21.4%) were much higher than the whole country average (10.3%) surveyed by Supervision and Testing Center for Rice and Products Quality, Ministry of Agriculture in 2002. Cd pollution of rice in area E and F were much higher than those in other sites which appeared the similar trends with the spatial distribution of Cd in soil. We may adduce two possible reasons for this phenomenon. Firstly, the Cd molecule size was small, which make it easier to transfer from soil to rice. Secondly, rice cultivation needs lots of water, which facilitated the dissolving of cadmium, the dissociation of Cd complex state, and the absorption and assimilation of Cd by the rice, especially in acid soil environment. In comparison, Pb varied significantly among same area. Pb pollution in

area E and F were also much higher than those in other sites as the pollution of Cd.

Figure 5 showed the spatial distributions of Cd (A) and Pb (B) contents in cabbage samples. 188 soils sites in total were planted with cabbage. Cd in cabbage ranged in 0.0018–3.6580 mg/kg, with a mean value 0.1466 mg/kg. Pb ranged in 0.0007–0.4362 mg/kg, with a mean value

0.0464 mg/kg. According to National Standard for Food Safety: Maximum Permission Limits of Pollutants (GB 2762–2012), 25(13.3%) samples exceeded the Cd standard for leaf vegetables and 4 (2.1%) samples exceeded the Pb standard for leaf vegetables respectively. Cd concentrations in cabbage in area E and F were higher than those in other four areas. The spatial distribution of Cd in cabbage was

Fig. 4 Spatial distributions of Cd (A) and Pb (B) contents in rice samples (mg/kg)

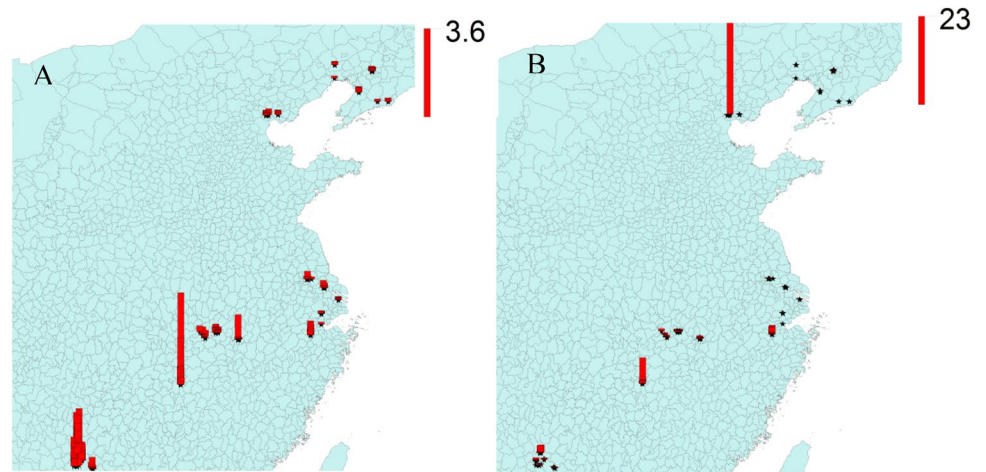
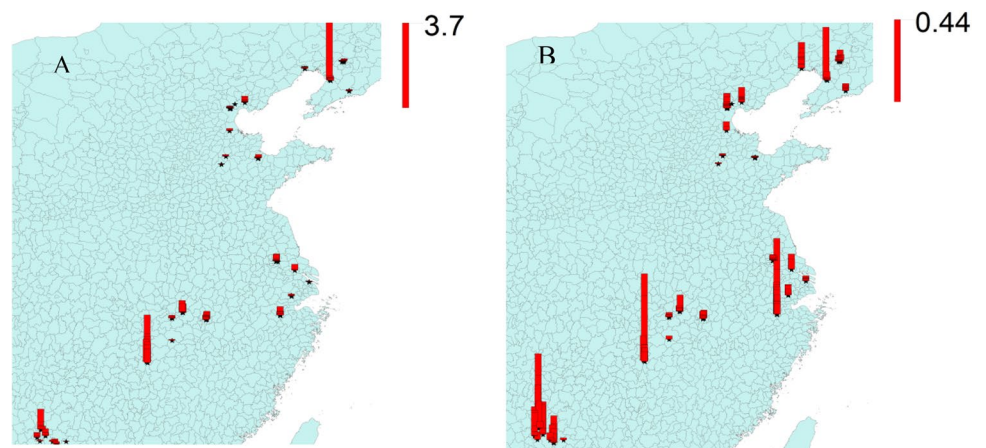


Table 2 Cd and Pb concentrations in plants (mg/kg)

	Cauliflower (n = 11)		Shepherd’s purse (n = 19)		Lettuce (n = 7)	
	Cd	Pb	Cd	Pb	Cd	Pb
Min	0.007	0.007	0.002	0.006	0.294	0.016
Max	0.158	0.291	0.156	0.17	0.928	35.69
Mean	0.057	0.054	0.072	0.037	0.475	5.134
	Rice (n = 247)		Cabbage (n = 188)			
	Cd	Pb	Cd	Pb		
Max	0.0111	0.0048	0.0018	0.0007		
Min	3.5631	22.980	3.6580	0.4362		
Mean	0.2285	0.2864	0.1466	0.0464		

Fig. 5 Spatial distributions of Cd (A) and Pb (B) contents in cabbage samples (mg/kg)



similar to that in soil. Arthur et al. (2000) reported that cabbage could easily accumulate Cd from soil which was coincided well with this study, so it must be considerate to plant cabbage in those Cd polluted areas.

Table 2 also showed the Cd and Pb concentrations in other plants. 11 cauliflower, 19 shepherd's purse and 7 lettuce samples were analyzed. Cd and Pb concentrations in lettuce were much higher than others. The results revealed that lettuce could highly accumulate Cd and Pb from soil which consistent with the previous studies (Xu et al. 2008).

Linear regression was conducted to reveal the relationship between the Cd and Pb concentrations in soil and rice, soil and cabbage as shown in the Fig. 6. Low correlation was observed in these two plants although samples were all collected from polluted soil with high Cd and Pb concentrations, which indicate that the plants only could absorb or assimilated certain amount of Cd and Pb. Both in rice and cabbage, Cd concentrations showed stronger linear correlation than Pb suggesting that Cd transferred more easily from soil to rice and vegetables mainly because of its lower molecule size and higher solubility in acid soil.

Table 3 showed the bio-concentration factors (BCF) of different sampling areas and plants. As for different areas, the average BCFs of Cd were 0.4151, 0.3550, 0.1259, 0.3159, 0.5292, and 0.2261 from area A to F. Interesting, area C, with the lowest organic matter contents (1.79%) and highest pH (7.81) was found with the lowest Cd bio-accumulation ability (BCF 0.1259). In contrast, area E, with low pH value (6.19) was found with the highest Cd bio-accumulation ability (BCF 0.5292). Cd bio-accumulation

ability (BCF 0.2261) in area F was not as high as that in E, although pH was the lowest in area F (5.68). This would be explained by the limits of plant assimilation and the abnormally high soil concentrations (the highest soil over-standard rate, 61/98). The pH values and organic matter contents are both important factors affecting the transference of Cd. The average BCFs of Pb were 0.0008, 0.0012, 0.0003, 0.0011, 0.0538 and 0.0020 from area A to F. A same trend of Pb was identified with Cd. No correlation of the average BCFs and average CECs was found in this study. Table 3 also showed that the average BCFs of Cd and Pb in 247 rice samples (0.4247, 0.0124) which were both higher than those in 188 cabbage samples (0.2520, 0.0064) manifest that rice could accumulate more Cd and Pb than cabbage.

Levels, spatial distribution and bio-accumulation of Cd and Pb in soil, rice and cabbage from 6 polluted arable areas were illustrated and investigated in this study. Specifically, Cd pollution in soil samples of two areas (area E and F) in south of China was more serious than those in the other four areas. Higher background values, soil acidity, Karst landscape and metallurgy industry might result in the pollution. Pb concentrations showed similar spatial distribution with Cd but lower over-standard rate in soil. As for foods, the concentrations of Cd and Pb in both rice and cabbage exceeded standards to a certain extent. Moreover, Cd showed stronger linear correlation than Pb both in rice and cabbage. Cd was readily bio-accumulated by cabbage while rice could accumulate more Cd and Pb than cabbage. Lettuce could highly accumulate Cd and Pb from soil. Consequently, it must be carefully considered to plant cabbage in Cd polluted

Fig. 6 Relationships of Cd and Pb concentrations in soil and rice, soil and cabbage (mg/kg)

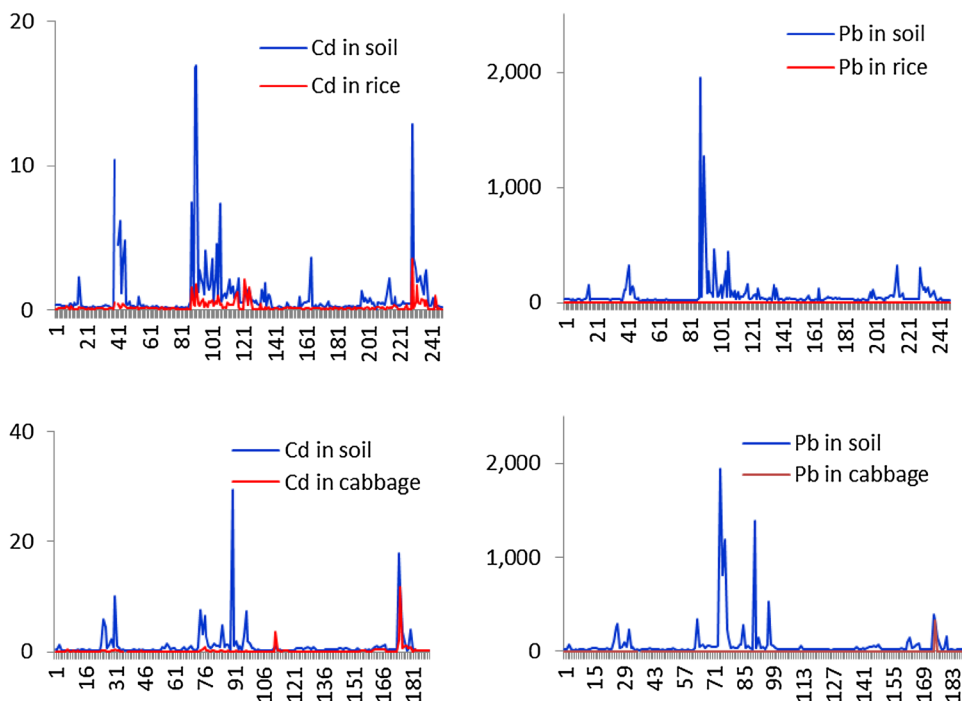


Table 3 Bio-concentration factors of different sampling areas and produces

Area	A		B		C	
	Cd	Pb	Cd	Pb	Cd	Pb
Min	0.0093	0.0000	0.0241	0.0001	0.0000	0.0000
Max	1.9739	0.0069	3.4880	0.0068	0.5011	0.0009
Mean	0.4151	0.0008	0.3550	0.0012	0.1259	0.0003
Area	D		E		F	
	Cd	Pb	Cd	Pb	Cd	Pb
Min	0.0059	0.0000	0.0000	0.0000	0.0012	0.0000
Max	1.6906	0.0130	5.6531	2.3420	2.0649	0.0278
Mean	0.3159	0.0011	0.5292	0.0538	0.2261	0.0020
Produce	Rice		Cabbage			
	Cd	Pb	Cd	Pb	Cd	Pb
Min	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
Max	4.0829	2.3420	5.6531	1.0000	5.6531	1.0000
Mean	0.4247	0.0124	0.2520	0.0064	0.2520	0.0064

areas and lettuce in Cd and Pb polluted areas. In summary, various remediation practices to decrease source pollution were still urgently needed in some areas in this study.

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