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Heavy metal concentrations of agricultural soils and vegetables from Dongguan, Guangdong

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Abstract: A total of 118 of agricultural soil and 43 of vegetable samples were collected from Dongguan City, Guangdong, China. The spatial distribution, sources, accumulation characteristics and potential risk of heavy metals in the agricultural soils and vegetables were depicted in details by three different approaches, including total contents of eight metal elements in soils and vegetables, GIS maps and multivariate analysis of heavy metals in soils in the study. The results show that there are higher accumulation of heavy metals such as Cu, Zn, Ni, Pb, Cd and Hg in agricultural soils, and the contents of Pb (65.38 mg kg⁻¹) and Hg $(0.24 \text{ mg kg}^{-1})$ are 1.82 and 2.82 times of the background contents of the corresponding heavy metals in soils of Guangdong Province, respectively. There are about 3.4% of Cu, 5.9% of Ni, 1.7% of Cd and 28% of Hg in all collected soil samples from all investigated sites which have overran the contents for heavy metals of the China Environmental Quality Standard for Soils (GB15618-1995, Grade II). The pollution characteristics of multi-metals in soils are mainly reflected by Hg. There are different sources to eight metal elements in soils, Cu, Zn, Ni, Cr and As are predominantly derived from parent materials, and Pb, Hg and Cd are affected by anthropogenic activities. The spatial distribution shows that the Cu, Zn, Ni, Cr, Pb, As and Hg contents of agricultural soils are high in the west and low in the east, and Cd contents are high in the northwest, southeast and low in the southwest in Dongguan. The ratios of vegetable samples which Ni, Pb and As concentrations higher than the Maximum Levels of Contaminants in Foods (GB2762-2005) are 4.7%, 16.3% and 48.8%, respectively. The order of bio-concentration factors (BCF) of heavy metals in vegetables is Cd > Zn > Cu > As > Ni > Hg > Cr > Pb. It is necessary to focus on potential risk of heavy metals for food safety and human's health from agricultural soils and vegetables in Dongguan City, Guangdong Province.

Keywords: agricultural soil; vegetable; heavy metal; pollution; spatial distribution; Dongguan

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1 Introduction

Heavy metal pollution in agricultural soils may lead to the functional disorder of soil, interference with crop growth, and even impair human health through a contaminated food chain (Lee *et al.*, 2006). With the quick progress of industrialization and urbanization, the dispersion of heavy metals into the environment has gained great concerns due to their potential impacts on the environment and on public health (Chen *et al.*, 2005; Zheng *et al.*, 2005a; Song *et al.*, 2006; Chen *et al.*, 2006; Zheng *et al.*, 2006).

The study area, Dongguan City, situated in the east central part of Pearl River Delta Economics Zone (PRDEZ), has been experiencing a rapid industrialization and urbanization process since China implemented the reform and opening up policy in the late 1970s. The gross domestic product (GDP) of Dongguan City in 2006 exceeded 210 billion RMB yuan, ranking the fourth in Guangdong Province, the most developed province in China. The amounts of industrial enterprises and motor vehicles in Dongguan City exceeded 21 thousand and 1.1 million in 2006, respectively (DCSB, 2007). With the quick progress of industrialization and urbanization, a large amount of pollutants including heavy metals were transported continuously into the agricultural soils directly and indirectly. In this study, agricultural soils and vegetables from Dongguan City were investigated. The characteristics of soil contamination and heavy metal accumulation in vegetables, the spatial variations and the sources of heavy metals in agricultural soils were studied to evaluate the soil environmental quality.

2 Materials and methods

2.1 Study area

Dongguan City can be divided into four totally different parts (the west plain area, the central area, southeast hilly area and the north water source protection area) based on geomorphologic types. The northwest, including 15 towns, is the delta plain of Dongjiang River and has abundant water resources. Parent materials of soil in this part are mainly sediments from Dongjiang River. There are four towns, including Shipai, Hengli, Qishi and Qiaotou, located in the eastern part of the upper Dongjiang River Delta. This part covers 213.0 km² and acts as the main water source for Dongjiang, Shenzhen and Dongguan canal. It is called the north water source protection area (I) due to the strict control against various polluting industries since 1991. The other 11 towns, with an area of 549.7 km², are called the west plain area (II) because of the abundant water and high density of town-owned enterprises. Parent materials of soil are dominated by the deposits from the Dongjiang River. Many resource-consuming enterprises developed at the initial stage of the reform due to the convenient traffic and abundant passing-by rivers on the west plain. The southeast, including nine towns, with an area of 1,018.6 km², is called the southeast hilly area (IV), where the water resource is short and the parent materials are mainly composed of granite, sandstone and shale. The center, including eight towns, such as Houjie, Humen etc., with an area of 693.5 km², is called the central area (III). Parent materials of soil are composed of the modern sediments from the Dongjiang River Delta, granite, red sandstone and shale (Lu, 1988).

2.2 Sample collection

An elaborate soil investigation was performed in May, 2002. A total of 118 surface soil (0-20 cm) and 43 vegetable samples were collected on the basis of the importance of crops, the sizes of agricultural area, industrial distribution, waste discharging, and irrigative water (Figure 1). At each soil sampling point, 10–15 sub-samples were randomly taken and then mixed to obtain a composite soil sample. Vegetable samples were mostly the edible parts of foliage.



Figure 1 Subdivision of Dongguan and location of the and samples sites

2.3 Sample analysis

All soil samples were air-dried at room temperature $(20-23^{\circ}C)$, removed stones or other debris, and then passed through 2 mm polyethylene sieve. Portions of soil samples (about 50 g) were ground in an agate grinder and sieved through 0.149 mm meth. The prepared soil samples were then stored in polyethylene bottles for analysis. Vegetable samples were washed carefully with tap-water and deionized-water. After washing, the samples were placed in an oven at 105°C for about 30 minutes and then dried at 60°C until a constant weight was achieved and ground to a fine powder. Soil samples were digested by with a solution of HNO₃-HCl-HF-HClO₄, and vegetable samples were digested by a mixture of HNO₃-HClO₄. Hg content was analyzed with cold atomic absorption spectrometer, As content was determined with DDCAg spectrophotometry, and Cu, Ni, Pb, Cr, Cd and Zn were determined with atomic absorption spectrometer (Liu, 2001). All samples were analyzed in testing center of Guangdong Institute of Eco-Environment and Soil Sciences. Blank and standard reference materials for soil (GSS-1) and plant (GSV-4) samples, obtained from the China National Center for Standard Reference Materials, were included for quality assurance. The concentrations of heavy metals in vegetable samples were based on the weight of fresh vegetables.

The soil pH was measured by pH meter with a soil/water ratio of 1:2.5. The soil organic matter was determined by wet oxidation with a mixture of potassium dichromate and sulfu-

ric acid. The testing results (Table 1) show that the agricultural soil in Dongguan City is acidic and the contents of soil OM is medium, even low.

	Range	Arithmetical mean (SD)	CV/%	Skewness	Kurtosis	Distribution
pH	3.69-7.73	5.89 (0.84)	14.26	0.22	-0.28	Normal
OM (%)	0.79-3.36	1.91 (0.54)	28.27	0.55	0.22	Normal

 Table 1
 Basic properties of agricultural soils in Dongguan

Note: SD is the standard deviation.

2.4 Risk evaluation

To quantify the pollution risk, the Nemero synthesis index evaluation method was performed. The synthesis index (Guo *et al.*, 2008) can be computed with the formula below:

$$P = \sqrt{\frac{\left(\frac{C_i}{S_i}\right)_{\max}^2 + \left(\frac{1}{n}\sum_{i=1}^n \frac{C_i}{S_i}\right)^2}{2}}$$

where *P* is the synthesis evaluation score, C_i is the measured content of a certain element at a sampling point, *i* denotes the element, and S_i is the evaluation criterion of the *i*-th element. In this study, the evaluation criterion is based on the China Environmental Quality Standard for soil metals (GB15618-1995, Grade II) (pH<6.5) (SEPAC, 1995). A result of $P \le 1$ means the soil is clean, $1 < P \le 2$ means the soil is slightly polluted, and $2 < P \le 3$ means the soil is heavily polluted with heavy metals.

2.5 Statistical analysis

Values for the contents of As, Cd, Hg, Cr, Cu, Ni, Pb and Zn were tested with normal distribution to avoid result distortions and low levels of significance (Shapiro–Wilk determination, p<0.05) by software of Origin7.5. The geostatistical analysis was carried out with Surfer 8.0. Maps were generated using ArcGIS 9.0. All statistical analyses were performed using Microsoft Excel 2003 and SPSS 13.0. Analysis of variance (ANOVA) was used to examine statistically significant differences in the mean concentrations of heavy metals among groups of soils and vegetables. A probability level of p<0.05 was considered significant.

3 Results and discussion

3.1 Descriptive statistics of heavy metals in agricultural soils

The contents of Cu, Zn, Ni, Cr, Pb, Cd, As and Hg in soils from the investigated sites vary greatly, ranging 5.08-105.60, 8.43-169.50, 2.29-57.46, 20.36-137.20, 0.02-0.67, 0.40-28.87 and 0.01-1.01 mg kg⁻¹, respectively (Table 2). The mean concentrations of Cu, Hg, Cd, Ni, Pb and Zn in the soils exceed background levels for soils in Guangdong Province (GPEMC, 1990). The contents of Pb (arithmetical mean) and Hg (geometric mean) in agricultural soils are 65.38 and 0.24 mg kg⁻¹, respectively, which are 1.82 and 2.82 times of the background levels. The background levels are exceeded by 66.1% of Cu, 65.3% of Zn, 61.0% of Ni, 30.5% of Cr, 92.4% of Pb, 69.5% of Cd, 46.6% of As and 89.8% of Hg in soil

Element	Range	Median	Arithmetical mean (SD)	Geometric mean (SD)	Backgr- ound value ^a	China Environmental Quality Standard for Soil Metals ^b	Ratio of overrunning (%) ^b
Cu	5.08-105.60	20.62	21.82(10.67)	19.38(1.67)	17.65	50	3.4
Zn	8.43-169.50	63.15	66.15(33.80)	53.97(1.87)	49.71	200	0
Ni	2.29-57.46	20.73	20.52(11.10)	16.91(1.98)	17.80	40	5.9
Cr	2.28-86.59	42.51	43.01(20.00)	35.07(2.00)	56.53	150	0
Pb	20.36-137.20	58.50	65.38(24.44)	62.36(1.46)	35.87	250	0
Cd	0.02-0.67	0.12	0.12(0.08)	0.10(1.77)	0.094	0.3	1.7
As	0.40-28.87	13.00	12.76(6.82)	10.13(2.14)	13.52	40	0
Hg	0.01-1.01	0.21	0.24(0.17)	0.24(2.16)	0.085	0.3	28

Table 2 Contents and pollution characteristics of heavy metals in agricultural soils of Dongguan (n = 118)

Note: SD is the standard deviation; ^a soil trace element background for Guangdong Province (GPEMC, 1990); ^b compared to the China Environmental Quality Standard for Soil Metals (GB15618-1995, Grade II).

samples. The results show the main contribution to heavy metal contamination of agricultural soils in Dongguan City is that of Pb and Hg, and the accumulations of Cu, Zn, Ni and Cd contents in soils are also found.

According to the Nemero synthesis index based on the mean concentrations of heavy metals in soils, P is 0.73. The China Environmental Quality Standard for Soil Metals (GB15618-1995, Grade II) (SEPAC, 1995) is exceeded by 3.4% of Cu, 5.9% of Ni, 1.7% of Cd and 28.0% of Hg in soil samples collected from the investigation sites. Table 2 shows there are multiple metal contaminations, and contamination by Hg is especially high.

3.2 Sources and spatial distributions of heavy metals in agricultural soils

3.2.1 Sources of heavy metals in agricultural soils

The results of Pearson partial correlation analysis (Table 3) show that the content of As has a significantly negative correlation with pH. Organic Matter (OM) exhibits a significantly positive linear relationship with Cu, Zn, Ni and Cr. The relationships between the contents of Cu and As and Zn, of Zn and Ni and Cr, of Ni and As and Cr, of Pb and Hg, of Cd and Hg in soils are significantly positive. These results suggest that the origin of As, Cu, Zn, Ni

	r r					· · · · ·				
	pН	OM	Cu	Zn	Ni	Cr	Pb	Cd	As	Hg
pН	1									
OM	-0.185	1								
Cu	0.140	0.281^{*}	1							
Zn	0.009	0.230^{*}	0.563**	1						
Ni	-0.048	0.495**	0.015	0.284^{*}	1					
Cr	-0.063	0.264*	-0.051	0.294^{*}	0.516**	1				
Pb	0.044	0.064	0.039	0.145	0.11	0.183	1			
Cd	0.194	-0.171	0.112	0.174	-0.127	0.128	-0.117	1		
As	-0.302**	0.078	0.37**	0.023	0.396**	0.116	-0.137	0.094	1	
Hg	0.010	0.194	0.015	0.113	-0.05	-0.105	0.317**	0.232^{*}	-0.059	1
*	**									

Table 3 Pearson partial correlations matrix for the heavy metals, pH and OM in agricultural soils

Note: *, p<0.05; **, p<0.01.

and Cr is possibly soil parent materials, and Pb, Cd and Hg have the same source.

In environmental science, factor analysis (FA) is a useful statistics tool that can identify origins of heavy metals in soils (Facchinelli *et al.*, 2001; Loska *et al.*, 2003; Boruvka *et al.*, 2005; Martin *et al.*, 2006). The results of FA for heavy metal contents are presented in Table 4. According to the results, F1 is dominated by Cu, Zn, Ni, Cr and As. According to earlier discussions, the contents of these elements in soils are similar to background levels for soils in Guangdong Province (GPEMC, 1990), and have significant correlations with OM or pH (Table 3). It is generally considered that Ni and Cr in soils are controlled by parent rocks (Facchinelli *et al.*, 2001; Loska *et al.*, 2003), and Boruvka *et al.* (2006) also drew conclusions that Cu, Zn, Ni, Cr and As mostly come from soil parent materials.

Element -		Component matrix		R	otated component mat	rix
	F1	F2	F3	F1	F2	F3
Cu	0.660	-0.228	0.112	0.697	-0.033	0.115
Zn	0.809	-0.037	0.165	0.774	0.171	0.233
Ni	0.877	-0.054	-0.219	0.848	0.288	-0.131
Cr	0.885	-0.126	-0.059	0.878	0.180	0.003
Pb	0.485	0.613	-0.421	0.261	0.829	-0.186
Cd	0.230	0.271	0.880	0.118	0.062	0.940
As	0.812	-0.298	0.032	0.864	-0.026	0.036
Hg	0.265	0.834	0.050	-0.025	0.823	0.301

 Table 4
 Factors obtained by varimax orthogonal rotation and total varimax explained

Extraction method: Principal Component Analysis; Rotation method: varimax with Kaiser normalisation.

F2 includes mainly Pb and Hg. The higher contents of Pb and Hg contribute in the west plain area and the central area where township enterprises are prosperous (Figure 3). This implies the source of Pb and Hg in soils significantly correlates with the industrial activities. In addition, Pb is generally regarded as a tracer element for vehicle exhausts in urban soils (Facchinelli *et al.*, 2001; Hanesch *et al.*, 2003), and the source of Pb is significantly correlative with dense highway net and the rapidly increasing motor vehicles in Dongguan City. Therefore, F2 can be called as industrial activities and traffic sources.

F3 contains Cd only. Cd is usually considered as marking element of agricultural activities which include the use of pesticides and chemical fertilizers and so on (Nicholson *et al.*, 2003; Rodriguez *et al.*, 2006). According to statistics (DCSB, 2005), there have been 2 million tons of chemical fertilizers and 35 thousand tons of pesticides that were used in Dongguan since 1990. The amount is much higher than that of the average of China. Utilization efficiency of pesticides and chemical fertilizers is generally lower in China, where approximately 70% drained into soil, water and air (Chen *et al.*, 2002). Long-term agricultural production activities result in obvious accumulation of Cd in agricultural soils. In addition, industrial pollution is also a significant source of Cd in soils because the area is an important international manufacturing industrial base in China. Liu *et al.* (2007) also reported that industrial pollution was the main source of Cd in cropland soil in Cixi City of Zhejiang Province, China. So it seems reasonable to infer that F3 is an anthropogenic factor, related to industrial and agricultural activities. Cluster analysis (CA) is often coupled with FA to confirm results and provide grouping of variables (Facchinelli *et al.*, 2001). In order to discriminate distinct sources of heavy metals, CA was performed on eight elements. The eight contaminating metals in agricultural soils can be classified into three types based on the correlation coefficients using the furthest neighbor linkage method (Figure 2). Group I comprises As, Cr, Ni, Cu and Zn, Group II comprises Hg and Pb, and Group III only comprises Cd. The metals in each group have similar source or releasing principles in soils (Guo *et al.*, 2008). The result is in good agreement with FA results.

The contents of heavy metals in soils from different parent materials vary greatly (Table 5). The geometric mean contents of soil Ni, Cr and As from river and marine deposits are 2.87, 2.77 and 2.70 times of those from sandstone and shale, respectively. The contents of soil Cu, Zn, Hg, Ni, Cr and As from river and marine deposits are the highest in all parent materials. The contents of soil Pb from river and marine deposits and granite are higher than contents from the other three parent materials. Moreover, the mean content of Cd in soils from granite is the lowest.



Figure 2 Clustering tree of heavy metals in agricultural soils

Table 5 Geometric mean for contents of heavy metals in the soils from different parent materials

	Content of heavy metals (mg kg ⁻¹)							
Element	River and marine deposit $(n = 30)$	Granite $(n = 23)$	River alluvial deposit $(n = 26)$	Metamorphic rock $(n = 14)$	Sandstone and shale $(n = 25)$	of F		
Cu	27.53(1.34)a	20.04(1.72)ab	19.69(1.34)b	19.23(2.24)b	15.93(2.0)c	2.46^{*}		
Zn	89.66(1.78)a	49.47(2.19)b	53.8(1.78)b	49.4(1.87)b	46.63(1.91)b	5.61**		
Ni	29.67(1.71)a	15.63(2.23)b	19.06(1.71)b	16.36(1.97)b	10.29(1.89)c	11.65**		
Cr	59.28(1.4)a	37.52(1.79)b	40.38(1.41)b	35.84(2.19)b	21.43(2.63)c	9.93**		
Pb	70.79(1.51)a	72.28(1.5)a	58.49(1.51)b	63.58(1.3)ab	52.63(1.44)b	2.97^{*}		
Cd	0.12(1.38)a	0.08(1.77)b	0.12(1.38)a	0.10(1.77)ab	0.12(1.75)a	1.62		
As	18.82(1.42)a	7.63(2.3)c	12.00(1.42)b	9.02(2.41)bc	6.97(2.43)c	14.22**		
Hg	0.25(2.25)a	0.19(2.09)ab	0.22(2.25)a	0.17(2.78)ab	0.16(2.23)b	1.29		

Note: *, p < 0.05, **, p < 0.01; the different letters in the same line show differences among the contents of heavy metals in soils from different parent materials; the data in brackets are the geometric standard deviations.

3.2.2 Spatial distributions of heavy metals in agricultural soils

In order to know the distribution patterns of heavy metals, Kriging interpolation was used to obtain the filled contours maps. The spatial structure analyses to soil Zn, Ni, Cr, Pb and As can directly be performed without proportional effect because the contents of these elements follow normal distributions (Zhang *et al.*, 1998). The experiment semivariograms of these

elements contents were calculated and fitted right with theoretic models according to their attributes, respectively. It was found that the experiment semivariograms of soil Zn, Ni, Cr, Pb and As can be well fitted with the exponential, spherical, spherical, spherical and exponential model, respectively. Then spatial variation maps of these elements were obtained with ordinary Kriging estimation according to the above semivariograms parameters (Figure 3). Logarithmic transformations were applied to soil Cu, Cd and Hg contents because of following lognormal distributions, then the spatial structure analyses were performed with these logarithm transformed data. The experiment semivariograms were calculated, and were fitted with suitable theoretic models according to their attributes, respectively. It was found that the experiment semivariograms of these data can be well fitted with the spherical, spherical and exponential model, respectively. Then spatial variation maps of these data were obtained with ordinary Kriging estimation according to the above semivariograms parameters, finally spatial variation maps of soil Cu, Cd and Hg contents were gained by taking inverse logarithm transformation (Figure 3).

Figure 3 shows that the contents of soil Cu, Zn, Ni, Cr, Pb, As and Hg are all relatively high in the west, and low in the east of Dongguan City, but the content of soil Cd is relatively high in the northwest and southeast, and low in the southwest. The geometric mean contents of heavy metals for each district were calculated and listed in Table 6. It shows the highest geometric mean contents of all eight heavy metals are in the west plain area, especially soil Zn, Pb and Hg have significantly accumulated there and the geometric mean contents of these elements are 1.67, 1.98 and 3.17 times of the background levels for soils in Guangdong Province (GPEMC, 1990), respectively. In the central area the accumulation of soil Hg is high, and the geometric mean content is 2.47 times of the background level, but the accumulations of soil Pb and Zn are only faint, and the mean contents of soil Cu, Ni and Cd are approximate to the background levels. In the southeast hilly area Pb and Hg have accumulated to a certain extent, the mean contents of soil Cu and Cd are close to the background levels, but Zn, Ni, Cr and As show evident depletions. In the north water source protection area Pb and Hg have rather accumulated, too, and the mean contents of soil Cu, Ni, As and Cd are approximate to the background levels, but Zn and Cr show significant depletions.

Cu, Zn, Ni, Cr and As mostly originate from soil parent materials, and their spatial distributions in soils are controlled by parent rocks and landform in Dongguan City. The higher contents of Cu, Zn, Ni, Cr and As are distributed in the west of the study area (Figure 3). Compared to the soil parent materials, the distribution of higher contents of Cu, Zn, Ni, Cr and As show good similar distribution trends with the distribution of river and marine deposits. Moreover, the landform in Dongguan City is higher in the east than in the west (Lu, 1988), so it is propitious for Cu, Zn, Ni, Cr and As to accumulate in the west of the study area. However, the spatial distributions of Pb, Cd and Hg in soils are mainly affected by anthropogenic activity. The higher contents of Pb and Hg are distributed in the west plain and central areas, especially in Humen and Chashan towns (Figure 3). It is evident that the accumulations of Pb and Hg are induced by traffic and industrial pollution. Because of the prominent regional advantages, convenient traffic and abundant water resources, some resources-consuming enterprises had been developed rapidly in the west plain area in the initial stage of the reform. With the increasing development of heavy pollution industries, a



Figure 3 Spatial variation maps of heavy metals distribution in the soils of Dongguan City

	Content of heavy metals (mg kg ⁻¹)							
Element	West plain area $(n = 46)$	Central area $(n = 27)$	Southeast hilly area $(n = 31)$	North water source protection area $(n = 14)$	of F			
Cu	25.2(1.45)a	18.82(1.62)b	16.38(1.96)b	18.81(1.35)b	2.65			
Zn	82.97(1.33)a	62.34(1.78)b	38.51(2.1)c	37.46(1.65)c	12.84**			
Ni	26.88(1.44)a	18.11(1.89)b	9.38(1.97)c	16.00(1.75)bc	19.7**			
Cr	53.36(1.36)a	42.59(1.65)ab	20.78(2.61)c	36.86(1.36)b	13.24**			
Pb	71.18(1.34)a	68.63(1.55)a	53.02(1.38)b	46.65(1.48)b	7.13**			
Cd	0.13(1.67)a	0.09(1.61)b	0.09(2.04)b	0.10(1.6)ab	1.47			
As	16.46(1.38)a	8.47(2.11)bc	6.39(2.62)c	11.00(1.55)b	13.79**			
Hg	0.27(1.71)a	0.21(2.0)a	0.13(2.36)b	0.13(2.21)b	6.35**			

Table 6 Geometric mean for contents of heavy metals in the soils from different areas

Note: *, p < 0.05, **, p < 0.01; the different letters in the same line show differences among the contents of heavy metals in soils from different area; the data in brackets are the geometric standard deviations.

great deal of heavy metal pollutants were discharged into the environment, and the area has become the most severely polluted one in Dongguan City. The number of all the enterprises exceeded 140 in Humen town in 2006, including 50 electroplating factories and three power plants. There were 25 major pollution enterprises in Chashan town, too, which were mostly paper mills and electroplating factories (DCSB, 2007). Moreover, dense highway net and rapidly increasing motor vehicles in the west plain and central areas are also important factors to accumulations of soil Pb and Hg. Spatial distribution characteristic of soil Cd in Dongguan City is completely different from those of other elements, the higher contents of Cd are distributed in the southeast hilly area, and this probably correlates with the especial agricultural activities such as use of pesticides, phosphorus fertilizer and plastic film containing Cd and so on (Zheng *et al.*, 2005d).

3.2.3 Contents of heavy metals in agricultural soils for different land uses

Mean contents of Cu, Zn, Ni, Cr and Pb in agricultural soils for different land uses all followed the order of paddy soil > orchard soil > vegetable soil (Table 7), the order of Cd and Hg is paddy soil > vegetable soil > orchard soil, and the order of As is orchard soil > paddy soil > vegetable soil. The results are not all the same with those obtained in an investigation of Beijing City (Chen *et al.*, 2005; Zheng *et al.*, 2005a, 2005b, 2005c, 2005d; Zheng *et al.*, 2006), and this probably correlates with different soil parent materials and agricultural customs between Beijing and Dongguan cities. The geometric mean contents of Cu, Zn, Ni, Cr, As and Hg in paddy soils are 1.60, 2.26, 1.66, 2.15, 1.68 and 2.17 times of those in vegetable soils, respectively, and the geometric mean content of Hg in paddy soils is 2.29 times that in orchard soils (Table 7). These results indicate that the accumulation of heavy metals in soils is significantly affected by land use.

3.3 Concentrations of heavy metals in vegetables

The testing result of As content in vegetables is total As, but only inorganic As was mentioned in the Maximum Levels of Contaminants in Foods (GB2762-2005) (Ministry of Health, 2005), so they can not be compared directly. Some literatures (Ociel *et al.*, 2002; Oscar *et al.*, 2004) have reported about 87% of As content in vegetables is inorganic [As(III) and As(V)]. The content of total As in vegetables was converted into that of inorganic As

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Element	Vegetable so	(n = 61)	Paddy soil	(<i>n</i> = 26)	Orchard so	Orchard soil $(n = 31)$		
	Arithmetical mean (SD)	Geometric mean (SD)	Arithmetical mean (SD)	Geometric mean (SD)	Arithmetical mean (SD)	Geometric mean (SD)	of F	
Cu	20.11(12.46)	17.25(1.73)	33.65(29.53)	27.57(1.81)	25.61(9.09)	23.73(1.55)	4.06*	
Zn	48.97(25.59)	42.02(1.8)	97.15(21.75)	94.81(1.28)	78.27(31.65)	68.49(1.87)	18.02**	
Ni	18.29(10.48)	14.81(2.05)	26.62(8.27)	24.53(1.67)	27.99(14.11)	23.12(2.1)	7.15**	
Cr	34.43(18.76)	27.67(2.18)	63.52(22.38)	59.42(1.51)	54.2(19.01)	49.35(1.67)	14.73**	
Pb	63.75(22.87)	60.1(1.41)	86.27(25.7)	82.04(1.44)	78.05(25.47)	74.08(1.4)	5.31**	
Cd	0.12(0.06)	0.11(1.64)	0.13(0.03)	0.13(1.29)	0.11(0.06)	0.09(1.95)	0.5	
As	10.82(6.2)	8.36(2.37)	14.75(5.12)	14.04(1.39)	17.24(7.62)	15(1.83)	8.81**	
Hg	0.24(0.17)	0.18(2.24)	0.47(0.27)	0.39(2.02)	0.22(0.13)	0.17(2.45)	7.17**	

Table 7 The contents of heavy metals in the soils under different land use types

Note: *, p<0.05; **, p<0.01; SD is the standard deviation.

according to this ratio in the paper. The concentrations of Ni, Pb and As in fresh vegetables ranged from 0.04 to 0.67, 0.02 to 0.45 and 0 to 0.54 mg kg⁻¹, respectively (Table 8). The Ni, Pb and As concentrations in vegetable samples exceeded the Maximum Levels of Contaminants in Foods (GB2762-2005) (Ministry of Health, 1991, 1994, 2003, 2005) for 4.7%, 16.3% and 48.8% of the samples, respectively, implying that contamination by heavy metals in vegetables is comparatively serious. Furthermore, the arithmetic mean contents of Zn, Cr, Cd and Hg in the west plain area are higher than those in the other areas, and the arithmetic mean contents of Ni and As in the north water source protection area is higher than those in other areas (Table 9).

The ratio of element concentration in vegetables to that in the corresponding soil was calculated to appraise the bio-accumulation effects of vegetables uptaking toxic elements from the soils (Guo *et al.*, 2008). The results show that bio-concentration factors (BFs) of vegetables for these considered elements are in the order of Cd > Zn > Cu > As > Ni > Hg > Cr > Pb (Table 8), on the whole, which agrees with the literature data (Cd > Zn \approx Ni \approx Cu \approx Pb > As \approx Cr) for midstream and downstream of the Xiangjiang River in Hunan Province given by Guo *et al.* (2008). The other previous studies have shown that BFs of vegetables

_		Conter	nt of heavy meta	Ratio of				
Element	Range	Median	Arithmetical mean (SD)	Geometric mean (SD)	China Food Quality Standard	overrunning (%)	BF (SD)	
Cu	0.17-0.74	0.38	0.39(0.12)	0.37(1.35)	10	0	0.028(1.75)	
Zn	1.76-12.21	3.32	3.5(1.75)	3.22(1.47)	20	0	0.077(1.16)	
Ni	0.04-0.67	0.1	0.13(0.12)	0.11(1.88)	0.3	4.7	0.014(1.49)	
Cr	0.08-0.37	0.19	0.2(0.08)	0.18(1.54)	0.5	0	0.004(1.35)	
Pb	0.02-0.45	0.06	0.13(0.14)	0.09(2.43)	03	16.3	0.002(2.47)	
Cd	0.01 - 0.07	0.03	0.03(0.15)	0.027(1.65)	0.2	0	0.137(1.41)	
As	0-0.54	0.04	0.079(0.1)	0.04(2.52)	0.05	48.8	0.021(5.09)	
Hg	0-0.01	0.002	0.002(0.001)	0.002(1.76)	0.01	0	0.011(2.09)	

Table 8 Content and accumulation of heavy metals in vegetables of Dongguan

Note: SD is the standard deviation; BF is the bio-concentration factor, which is the ratio of the element concentration in vegetables to that in soil.

	Concentrations of heavy metals (mg kg ⁻¹)							
Element	West plain area $(n = 13)$	Central area $(n = 12)$	Southeast hilly area $(n = 10)$	North water source protection area $(n = 8)$				
Cu	0.35(0.11)a	0.43(0.13)a	0.34(0.08)a	0.43(0.09)a				
Zn	3.98(2.98)a	3.19(0.87)b	3.51(1.54)ab	3.34(0.80)b				
Ni	0.11(0.02)b	0.11(0.08)b	0.12(0.07)b	0.24(0.22)a				
Cr	0.25(0.77)a	0.19(0.08)ab	0.17(0.08)b	0.22(0.09)a				
Pb	0.14(0.15)ab	0.14(0.14)ab	0.16(0.17)a	0.06(0.05)b				
Cd	0.033(0.01)a	0.026(0.01)a	0.031(0.02)a	0.029(0.18)a				
As	0.11(0.08)a	0.08(0.08)ab	0.06(0.08)b	0.14(0.22)a				
Hg	0.0025(0.001)a	0.0018(0.001)b	0.0021(0.002)ab	0.0016(0.001)b				

Table 9 Arithmetic mean for contents of heavy metals in vegetables from different areas

Note: The different letters in the same line show the significant differences among the contents of heavy metals in soils from different areas (p<0.05); the data in brackets is the standard deviation (affiliated to the normal distribution).

from Beijing are in the order of $Cd > Cu > As > Pb \approx Ni > Cr > Zn^{1}$ and those from Chenzhou City are in the order of Cd > Zn > Cu > Pb > As (Liu *et al.*, 2005). These results suggest that Cd is easily transferred from soil to plants.

4 Conclusions

Some conclusions are drawn from this study as follows:

(1) Cu, Zn, Ni, Pb, Cd and Hg have accumulated to a certain extent in agricultural soils from Dongguan City. The main contributors to metal contamination are Pb and Hg. There is co-contamination of heavy metals, especially for Hg in agricultural soils.

(2) There are different sources to eight metal elements in soils, Cu, Zn, Ni, Cr and As are predominantly derived from parent materials, but Pb, Hg and Cd are affected by anthropogenic activities. The contents of heavy metals in soils from different parent materials vary widely.

(3) The Cu, Zn, Ni, Cr, Pb, As and Hg contents of agricultural soils are high in west, and low in the east, and Cd contents are high in the northwest, southeast and low in the southwest in Dongguan City according to GIS-based spatial distribution maps. The contents of heavy metals in soils from different areas vary greatly, and the highest mean contents of all eight heavy metals are in the west plain area, especially soil Zn, Pb and Hg have significantly accumulated and the mean contents are 1.67, 1.98 and 3.17 times of the background levels for soils in Guangdong Province, respectively.

(4) The accumulation of heavy metals in soils is significantly affected by land use. The mean contents of Cu, Zn, Ni, Cr and Pb in agricultural soils for different land uses all followed the order of paddy soil > orchard soil > vegetable soil, the order of Cd and Hg is paddy soil > vegetable soil > orchard soil, and the order of As is orchard soil > paddy soil > vegetable soil.

(5) The concentrations of Ni, Pb and As in vegetables exceed the pollutant standards of edible food quality in China by 4.7%, 16.3% and 48.8% of the testing samples, respectively.

¹⁾ Song B, 2007. GIS-based risk assessment of heavy metals in regional soils and planting regionalization of harmless vegetables: A case study in Beijing [D]. Institute of Geographic Sciences and Natural Resource Research, Chinese Academy of Sciences, Beijing, 40-83. (in Chinese)

The bio-concentration factors of vegetables in Dongguan City are in the order of Cd > Zn > Cu > As > Ni > Hg > Cr > Pb.

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