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ORIGINAL ARTICLE

Heavy metal distribution between parent soil and pepper in an unpolluted area, Hainan Island, China

Yu Guo · Yangrong Fu · Sen Yan · Yi Yang · Yuepin Guo · Zhengyu Bao

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Abstract Consuming edible plants contaminated by heavy metals transferred from soil is an important pathway for human exposure to environmental contaminants. In the past several decades, heavy metal accumulation in contaminated soil has been widely studied; however, few researches investigated the background levels of metals in plants and evaluated the difference in plants grown in soils produced from different parent rocks. In this study, a systemic survey of heavy metal distribution and accumulation in the soil–pepper system was investigated in an unpolluted area, Hainan Island, China. Levels of Cu, Pb, Zn and Cd were measured in soils and pepper fruits from five representative pepper-growing areas with different soil parent rocks (i.e. basalt, granite, sedimentary rock, metamorphic rock and alluvial deposits). Average concentrations of Cu, Pb, Zn and Cd in pepper fruits were 11.52, 0.84, 8.77 and 0.05 mg/kg, respectively. The concentrations of heavy metals in soils are controlled by the parent materials and varied greatly from in different areas. Heavy metal contents in all pepper samples were lower than the Chinese maximum contaminant levels. The relationship between heavy metals in soils and biological absorption coefficient (BAC) of pepper fruits suggests that the uptake ability of pepper for soil metals depends mainly on the physiological mechanism, while in some cases, the soil types and supergene environment are also important.

Keywords Heavy metals · Soil · Pepper fruits · Biological absorption coefficient (BAC)

Introduction

Heavy metal contamination in soil and plants is a worldwide issue of increasing concern due to the potential impact on human health (Calderon et al. 2001; Mantovi et al. 2003; Nan et al. 2002). Fruits and vegetables are important components of the human diet after cereals. Consuming the edible part of plants contaminated by the metals transferred from soil is an important pathway for human exposure to environmental contaminants. To date, most studies on metals accumulation in the soil–plant system were performed in the contaminated areas (Babula et al. 2008; Cheng et al. 2006; Fakayode and Onianwa 2002; Fernandez et al. 2000; Guan and Peart 2006; Krishna and Govil 2004, 2005; Li et al. 2006; We et al. 2009; Yoon et al. 2006). However, few researches were carried out in view of geological background, especially different soil parent rocks.

Agricultural production activities are conducted in certain geological backgrounds, and hence plants are inextricably linked to the parent material types. Hainan Island, which is surrounded by sea, is an unpolluted area, because there is no heavy industry and ecological tourism is the major human activity in the island. There are also some studies about heavy metals in plants and soil in Hainan Island (Bi et al. 2010; Guo et al. 2007; Wei et al. 2009). In this study, a systemic survey of

Y. Guo (✉) · S. Yan
Key Laboratory of Biogeology and Environmental Geology of Ministry of Educaion, China University of Geosciences, Wuhan 430074, People's Republic of China
e-mail: gy220090025@gmail.com

Y. Fu · Y. Yang · Y. Guo
Hainan Geological Survey, Haikou 570206, People's Republic of China

Z. Bao (✉)
State Key Laboratory of Geological Processes and Mineral Resources, China University of Geosciences, Wuhan 430074, People's Republic of China
e-mail: zybao@263.com

heavy metal distribution and accumulation in the soil–pepper system was conducted in Hainan; five representative pepper-growing areas with different soil parent rocks (i.e. basalt, granite, sedimentary rock, metamorphic rock and alluvial deposits) were selected. Biological absorption coefficient (Chojnacka et al. 2005) was used to examine the uptake ability of different metals in pepper plants.

The major objectives of this research were: (1) to determine the concentrations of Cu, Pb, Zn and Cd in soil and pepper fruits growing in Hainan; (2) to compare concentrations of heavy metals in pepper in different research areas in the world; and (3) to assess the accumulation capacity of heavy metals for pepper fruits.

Materials and methods

Description of the study area

The study area is in the northeast part of Hainan Island, southern China, an area of typical tropical monsoonal climate, with annual average temperature of 24.0°C and precipitation of 1,600 mm, which is very suitable for the growth of pepper.

Fourteen representative pepper plantations were selected in Wenchang and Qionghai city, the two largest pepper-growing areas in Hainan Island. The main soil parent rock types are basalt, granite, sedimentary rock, metamorphic rock and alluvial deposits, as described in Fig. 1.

Sampling and preparation

Seventy-four topsoils (0–20 cm) and thirty-two pepper fruits were collected in July 2005. The soil and corresponding plant samples were collected and all samples consisted of five subsamples. To compare with the topsoils, 34 deep soils (60–80 cm) were also sampled. After sampling, soil samples were air-dried at room temperature (25°C), rock, gravel, plant residues and other debris were picked out, and then ground to less than 100 μm for analysis. Pepper seeds were thoroughly cleaned by tapwater and then rinsed by Milli-Q deionized water to remove adhering particles. After that, the edible parts were separated and dried in an oven at 60°C. The dry samples were ground to fine powder for chemical analysis.

Analytical methods

Soil and plant samples were analyzed for heavy metals, including copper (Cu), lead (Pb), zinc (Zn) and cadmium (Cd). Soil pH was determined in a 5:1 (v/m) water/soil suspension using a pH meter. About 0.5 g of the pretreated soil samples was digested in Teflon beakers with a mixture of nitric acid (HNO_3) and perchloric acid ($HClO_4$). The concentrations of Cu, Pb, Zn and Cd in the digested solution were measured by inductively coupled plasma mass spectrometry (X7 ICP-MS, TMO, USA) at the Hubei Geological Analytical Center. For quality assurance and quality control (QA/QC), duplicates, method blanks and standard reference materials (GSS-1, GSS-2, GSS-3, GSS-8) were analyzed.

Data analysis

The SPSS (Statistic Program for Social Sciences) statistical program package (version 13.0) was used for statistical analyses of data. Pearson correlation coefficients (*r*) were used to examine the correlations between heavy metals in

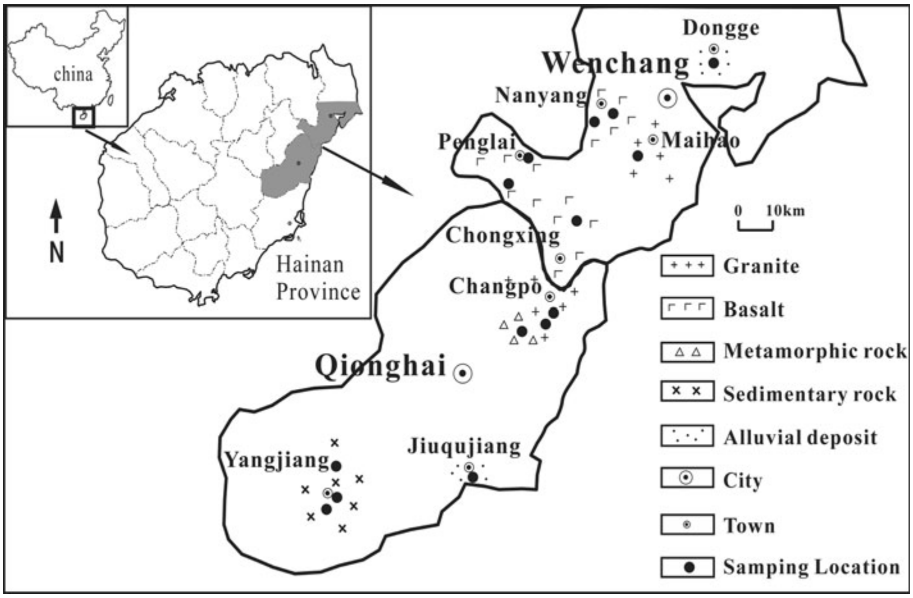

Fig. 1 Study area and sampling locations

topsoil and deep soil. Statistical significance of differences in both soil and plant concentrations of heavy metals was computed using one-way ANOVA.

Results and discussion

Soil properties and heavy metal concentrations

The average concentrations of heavy metals in topsoils and deep soils are shown in Table 1. The environmental disturbance by anthropogenic activities in deep soil is much less than that in topsoil; therefore, the deep soil can represent the soil parent material and background. Relationships of heavy metal concentrations between topsoils and deep soils can reveal the heritage from parent materials to soils and element mobility.

As shown in Table 1, the Cu and Zn contents in the basalt area are the highest among the five different sampling areas, and Cu and Zn contents in the granite area are higher than those in metamorphic, sedimentary and alluvial areas; whereas the highest Pb and Cd contents are observed in the metamorphic rock area, and the lowest Pb and Cd contents are found in alluvial and basalt areas, respectively. The ranges of average pH values in topsoils and deep soils are 4.79–5.73 and 4.73–5.50, respectively, which indicate that both topsoils and deep soils are acidic, and deep soils are generally more acidic than topsoils. For instance, the average pH of topsoils in the basalt area is 4.79, while the average pH of deep soils in the basalt area is 4.73.

Table 1 The average pH and heavy metal concentrations in soils growing pepper (mg/kg)

Parent material types (*N*)	Cu	Pb	Zn	Cd	pH
BA					
Topsoil (31)	97.15	28.47	144.94	0.05	4.79
Deep soil (14)	99.70	26.25	141.14	0.03	4.73
GA					
Topsoil (16)	50.38	45.18	71.93	0.07	5.36
Deep soil (8)	71.80	77.06	90.38	0.07	5.14
MA					
Topsoil (6)	10.09	108.4	54.6	0.26	5.73
Deep soil (3)	14.59	89.97	74.27	0.15	5.50
SA					
Topsoil (13)	13.92	24.08	47.26	0.05	4.90
Deep soil (5)	16.40	23.64	53.28	0.09	4.92
AA					
Topsoil (8)	13.32	21.35	39.5	0.07	5.63
Deep soil (4)	14.58	22.90	51.45	0.12	5.09

BA basalt area, *GA* granite area, *SA* sedimentary rock area, *MA* metamorphic rock area, *AA* alluvial deposits area

As shown in Fig. 2 and Table 2, there are significant correlations between heavy metals in topsoils and deep soils, which indicate that heavy metals in the topsoils are mainly controlled by the parent materials, with minor influence from anthropogenic activities. The correlation coefficients of Cu, Pb, Zn are larger than that of Cd, which could be attributed to the different geochemical behavior for metals in the supergene environment. In general, the difference in heavy metal concentrations in topsoils and in deep soils can be attributed to various anthropogenic and natural activities, such as the usage of chemical fertilizer and pesticides, sewage irrigation and runoff leaching. However, the study area is a world-famous tourist destination in China, and the environment is less affected by human activities. Thus, the difference can be mainly caused by natural factors. As shown in Fig. 2, Cu and Zn concentrations in topsoils are comparable with those in deep soils, while Pb and Cd contents are depleted in topsoils. This is probably due to the different mobility of elements in the soil. Pb and Cd are more mobile than Cu and Zn, thus, are likely to migrate into the deep soil via runoff leaching. In addition, soil pH values between topsoils and deep soils have no significant correlation, with the correlation coefficient of 0.338 (Table 2), probably because pH showed different behaviors in the supergene environment and in the deep. Compared to the deep, the surface environment appears more complex, the pH value related to organic matter, rainfall, climate, etc. For example, the value will decrease when the soil organic matter content increases.

Heavy metal concentrations in pepper

The average concentrations of heavy metals in pepper reported in this study and the literature are presented in Table 3. Compared to the pepper cultivated in Serbia, the concentrations of Cu and Pb in this study are higher than those grown in open fields and plastic sheds in Belgrade, Serbia (Markovic et al. 2010), while the Zn and Cd concentrations are approximately two and five times lower than those found in Serbia, respectively. Compared with the pepper reported in Korea, only Cu content in pepper from Hainan is slightly higher than that in controlled areas from Korea; the other metals are all lower in Hainan than that both in the controlled area and non-controlled areas from Korea (Jung and Thornton 1996). Compared to the pepper grown in Egypt, the contents of Cu and Pb in Hainan are higher and Zn lower; Cd appears equal (Radwan and Salama 2006). Moreover, the Cu, Pb and Cd contents in pepper are much lower than the Chinese maximum contaminant levels. Through the comparisons of heavy metals in pepper produced from Hainan to other countries and Chinese maximum contaminant levels, the levels of

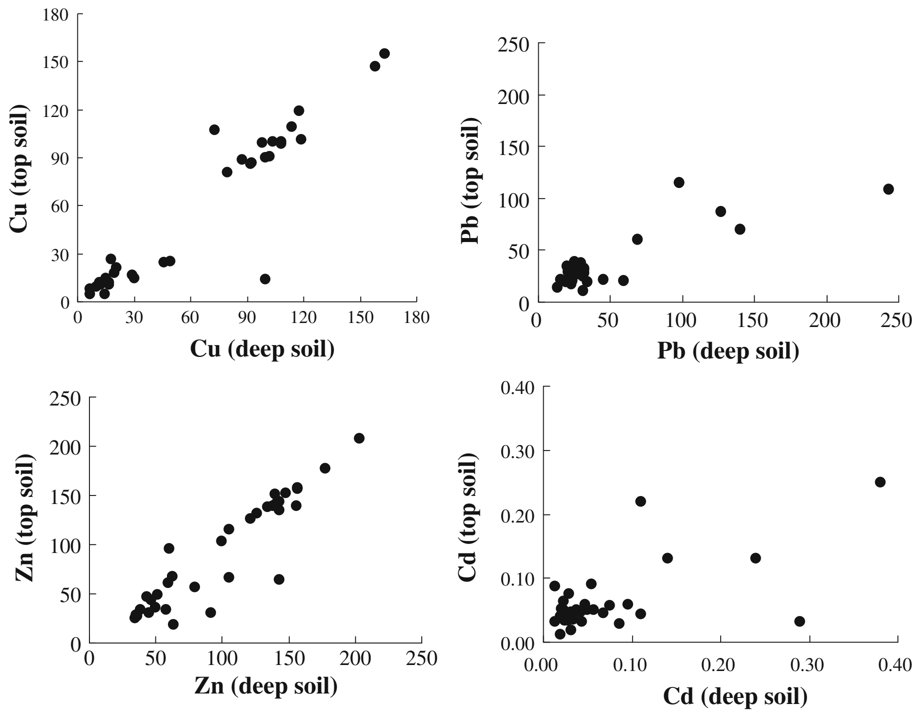

Fig. 2 Scatter plots of heavy metal concentrations between topsoils and deep soils (mg/kg)

Table 2 Pearson correlation of heavy metals and pH between topsoil and deep soil

Correlation coefficients	Cu	Pb	Zn	Cd	pH
r	0.937	0.845	0.922	0.638	0.338

Correlation is significant at the 0.01 level (two-tailed)

Table 3 Comparison of heavy metals in pepper and other plants (mg/kg)

Locations	Plant types (DW)	Cu	Pb	Zn	Cd	Reference
Hainan, China	Pepper	11.52	0.84	8.77	0.05	Present study
Belgrade, Serbia	Pepper (open field)	8.21	0.25	15.1	0.24	Markovic et al. (2010)
	Pepper (plastic shed)	6.55	0.41	17.0	0.24	
Korea	Red peppers[a]	11.6	1.34	51.0	0.27	Jung and Thornton (1996)
	Red peppers[b]	9.1	1.20	42.0	0.20	
Egypt	Green pepper	4.53	0.47	12.5	0.05	Radwan and Salama (2006)
Chinese maximum contaminant levels		20.0	5.0	–	0.3	(WM/T2-2004)

[a] In and around a lead–zinc mine

[b] Control area

copper and lead content were medium; and zinc and cadmium concentrations were the lowest, which indicated good quality of Hainan pepper.

Relationships between heavy metal concentrations in soils and in peppers

To understand the uptake of metals from soil to pepper, correlation analysis was performed between metal concentrations in topsoils and peppers. Through correlation analysis, complex linear correlations between concentrations of heavy metals in soil and pepper were calculated, which enabled the interpretation of heavy metals transfer from soil to pepper.

Scatter plot of Cu shows an approximately linear relationship between the element in soil and in pepper. As shown in Fig. 3, there are two groups: one group with higher Cu contents in soil and pepper and the other group

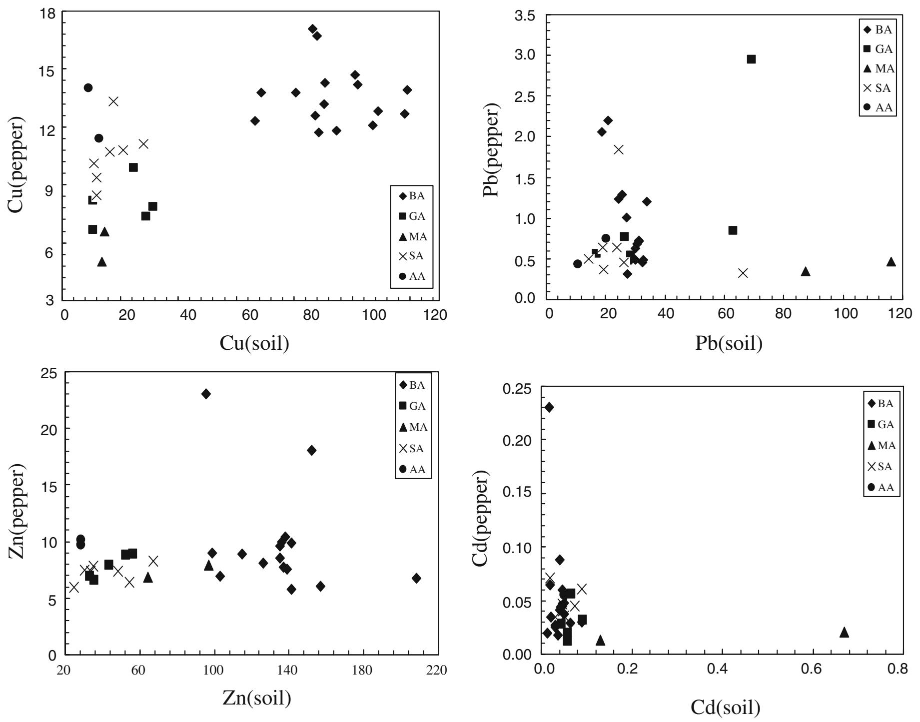

Fig. 3 Scatter plots of heavy metal concentrations between top soils and peppers (mg/kg). Refer Table 1 for explanations for BA, GA, MA, SA and AA

with lower Cu contents in soil but variable Cu contents in pepper. This phenomenon could be attributed to two main factors: Cu concentration in soil and its bioavailability. Cu concentrations in pepper grown in granite and basalt areas increase with increasing Cu contents in soil, which reveals that an increase of Cu content in soil can directly lead to an increase of Cu content in pepper. In addition, Cu contents in peppers grown in the soils with low Cu contents fluctuate from 5.0 to 14.0 mg/kg. For example, the average Cu content in pepper from the sedimentary rock area is twice that in the metamorphic rock area, while the Cu concentrations in soils are almost the same. This is probably caused by the different bioavailability of Cu in different types of soil. The bioavailability of metal in soil is a function of soil pH, organic matter content, metal speciation, oxidation–reduction potential, etc. As shown in Table 1, the average pH value (4.9) of soils in the sedimentary rock area is lower than that (5.7) for the metamorphic rock area, indicating the increasing bioavailability of Cu in more acidic soil.

The range of Zn contents in most pepper samples is 5–10 mg/kg, and Zn contents in pepper are relatively stable with different soil types and Zn contents in soil. This suggests Zn uptake by pepper is mainly controlled by physiological mechanism and genetic characteristics of pepper.

Pb and Cd show a certain degree of heavy metal antagonist, i.e., the lower concentration of metals in soil, the easier absorption of plants; when the metal concentration increased in the environment, the plants showed resistance to absorb, i.e. plants appear to have low levels of heavy metal concentration in high content soils. As shown in Fig. 3, there is obvious negative correlation between Cd concentrations in soil and pepper.

In short, pepper has a different uptake mechanism to different elements, which is dependent on many factors including soil types, parent materials, soil pH, moisture, aeration, temperature, organic matter, biomass, rooting depth, age and other environmental factors (Waldrop et al. 2000).

Biological absorption coefficient (BAC) of heavy metals from soil to pepper

Plants can absorb heavy metals from soil and have different absorption and accumulation ability to different metals. To illustrate the migration ability of metals in the soil–plant system, BAC can be used to examine the absorption and accumulation of different metals in plants. Absorption coefficient is the ratio of a certain element concentrations in plants (dry weight) and the corresponding soil, which can represent migration of elements in the soil–plant system, and reflect the accumulation capacity of elements in plants.

BAC for element i is calculated as follows:

$$\mathrm{BAC}_i = C_i / S_i$$

where C_i is the concentration of i in plant (mg/kg) and S_i is the concentration of i in corresponding top soil (mg/kg).

The BACs for Cu, Pb, Zn and Cd are plotted in Fig. 4. Among these four metals, Cu and Cd were easily absorbed

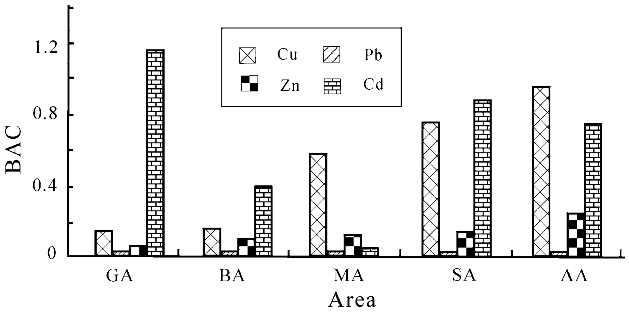

Fig. 4 Biological absorption coefficient (BAC) of heavy metals in pepper fruits. Refer Table 1 for explanations for BA, GA, MA, SA and AA

and accumulated by pepper; Zn was less absorbed, while Pb was hard to accumulate in pepper. Moreover, the BACs for Cu and Cd varied significantly in different sampling areas, while the variations of BACs for Pb and Zn were less pronounced. For instance, the largest BAC of Cd in the basalt area was over 10 times larger than the lowest one in the metamorphic rock area. Previous studies indicated that Cd is more labile and bioavailable under more acidic conditions (Bi et al. 2010; Guo et al. 2007; Wei et al. 2009). In this study, the average soil pH value is highest in metamorphic area (Table 1); thus the Cd mobility and bioavailability are suppressed in MA soil, which resulted in the decrease of Cd uptake by pepper. Furthermore, the average Cd content is the highest in MA soil (Table 1). Therefore, the BAC value of Cd in MA is the lowest among all the sampling areas.

Conclusions

This study investigated the contents of heavy metals in soils and peppers. The results indicated that heavy metal contents changed significantly in soils collected from different parent material areas, while heavy metal contents in plants varied slightly. Compared to the data reported in previous studies, Cd levels in this study were considerably low, while Cu levels were relatively high, and Pb and Zn levels were at a medium level. Furthermore, heavy metal concentrations in all pepper samples did not exceed the Chinese maximum contaminant levels. In addition, the relationship between heavy metals in soil and pepper, and BAC suggest that metals uptake by pepper from soil mainly depend on the physiological mechanism, while in some cases, the soil types and supergene environment are also important.

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