HEAVY METALS IN THE ENVIRONMENT

# Effects of Land Use and Parent Materials on Trace Elements Accumulation in Topsoil

Cheng-Long Tu, Teng-Bing He,\* Cong-Qiang Liu, and Xiao-Hui Lu

To determine the effects of parent material and land use on the concentration of trace elements in the agricultural topsoil of Guizhou Province, China, a total of 584 agricultural topsoil samples were collected in a typical region. The results indicate that the contents of trace elements (As, Cd, Cr, Hg, and Pb) in agricultural soils were greater than in the uncultivated soils, and the paddy fields exhibited higher contents of trace elements than dry lands. The enrichments of most trace elements in agricultural topsoil derived from carbonate rock were more serious. In paddy fields, Cd, Cr, and As showed positive relationships with soil organic matter (p < p0.01) but were not affected by pH, carbon-to-nitrogen (C/N) ratio, and clay (p > 0.05). Lead and Hg formed the second component in principal component analysis (PCA) and were closely related to pH and clay content. In dry lands, the trace elements were well correlated with pH, C/N, and clay (p < 0.05). Analysis of PCA and correlation showed that Cd, Cr, and Hg were mainly derived from inorganic fertilizers, whereas Pb and As were primarily from organic manures. These results suggest that the effect of anthropogenic activities on paddy fields is more serious than on dry lands. Parent materials not only serve as sources of soil trace elements but also control the loss and accumulation of trace elements by affecting soil physicochemical properties, especially in dry lands.

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TITH THE INTENSIFICATION of agricultural activities, increasing amounts of inorganic and organic manures, as well as pesticides, are added into agricultural soils. These fertilizers and pesticides contain hazardous trace elements such as arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb) (Mermut et al., 1996; Nicholson et al., 2003; Nziguheba and Smolders, 2008). Most trace elements are extremely persistent in the environment, cannot be degraded by microbes, and accumulate easily (Springael et al., 1993). These trace elements may be further enriched by the intensification of agricultural activities (Mantovi et al., 2003; Montagne et al., 2007; Smith et al., 1996). An excessive accumulation of trace elements in agricultural soils can restrict soil function, cause toxicity to plants, and contaminate the food chain (Jarup, 2003; Muchuweti et al., 2006). The accumulation of trace elements in agricultural soils has become an increasingly serious problem (Huamain et al., 1999; Wong et al., 2002).

The concentrations of heavy metals in agricultural soils are usually controlled both by the parent materials and landuse conditions (Wang and Chen, 1998; Wong et al., 2002). Generally, the trace elements in agricultural soils primarily depend on the composition of parent material during the initial stage of cultivation (De Temmerman et al., 2003). The loss and accumulation of trace elements are closely related to the physicochemical characteristics of soils (Dragovic et al., 2008; Kim et al., 2003; Vega et al., 2006; Yang et al., 2006). The concentrations of trace elements differ between parent materials, and the properties of soils derived from different parent materials have pronounced differences(Mao, 2007). Soil properties are the key factors in controlling the level and distribution of trace elements in soil (Kashem and Singh, 2001; Montagne et al., 2007). However, inconsistent results have been reported (Dragovic et al., 2008; Yang et al., 2006). Moreover, the accumulation processes of trace elements vary with landuse conditions (Wong et al., 2002). Land-use and related management practices can affect soil properties (Islam and Weil, 2000). Thus, the primary objectives of the current study are (i) to determine trace element content of soils from Wudang District,

C.-L. Tu and C.-Q. Liu, State Key Lab. of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550002, China; T.-B. He, Agricultural College, Guizhou Univ., Guiyang 550001, China; X.-H. Lu, School of Geographic and Environmental Sciences, Guizhou Normal Univ., Guiyang 550001, China. Assigned to Associate Editor Scott Young.

Abbreviations: CEC, cation exchange capacity; PCA, principal component analysis; SOM, soil organic matter.

southwest China, (ii) to assess the relative contribution of parent materials and anthropogenic inputs to the concentration of soil trace elements, and (iii) to analyze the factors that control the accumulation of trace elements in agricultural topsoil.

# **Materials and Methods**

# **Study Area**

Wudang District is part of Guiyang, the capital of Guizhou Province located in southwest China, and is home to 3 million people (Fig. 1). The district is situated between 26°32′ N and 26°55′ N and 106°43′ E and 107°3′ E, with a mean elevation of 1300 m above sea level. The area has a warm and humid subtropical climate, with an average annual temperature and rainfall of 15.3°C and 1197 mm, respectively. The agricultural soils are found mainly in mountainous fields and derived from carbonate rock, red residua, and sand-shale. The favorable climate and fertile soil make Wudang District an important base of agricultural product supply to Guiyang.

# **Sampling and Analysis**

A total of 584 surface soil samples (0-20 cm) were collected at random agricultural plots based on land-use types and parent materials, of which 89 uncultivated topsoil samples derived from different parent materials were taken as references (see Table 1, Fig. 1). A composite sample was taken from each plot, consisting of 4 to 8 soil cores excavated from the surface horizon (0-20 cm). The cores were mixed into one composite sample for each soil type and analyzed in duplicate or triplicate. All soil samples were transported to the laboratory where they were airdried for several days at ambient temperature. The samples were then finely ground, passed through a 100-mesh nylon sieve, and stored in closed bottles at room temperature.

The basic soil properties relevant to controlling the mobility and bioavailability of the trace elements were analyzed in accordance with the official laboratory methods of the Soil Science Society of China. Soil pH value was measured from a 1:2.5 (m/v) soil-to-water suspension using a pH electrode (Orion). The composition of soil particle size was calculated according to the sinking rates with hydrometer method. The content of soil organic matter (SOM) was determined by oxidation with dichromate based on the Walkley–Black method. Cation exchange capacity (CEC) was determined by saturation with sodium acetate solution, replacement of the adsorbed sodium with ammonium, and determination of the displaced sodium by flame atomic absorption spectrometry.

The soil samples were digested with nitric acid (HNO<sub>3</sub>) and hydrochloric acid (HCl) with a ratio of 3:1 (HNO<sub>3</sub>:HCl). Cadmium and Pb concentrations in the digested solution were measured using a graphite furnace atomic absorption spectrophotometer, As and Hg were analyzed using atom



Fig. 1. Location of the study area and distribution of the sampling villages.

fluorescence spectrometry, and chromium (Cr) was determined using a flame atomic absorption spectrophotometer.

## **Statistical Analysis**

Data analysis was performed using the SPSS 12.0 statistical package. The differences between trace element concentrations in the soils derived from different parent materials or under different land-use types were assessed by analysis of variance (ANOVA). A correlation matrix (Pearson correlation) was used to identify the relationships between the heavy metals concentrations and soil properties. Principal component analysis (PCA) was used to analyze the multivariate relationships among soil properties and trace element concentrations.

# Results

## **Basic Soil Physicochemical Properties**

The soil samples were divided into three groups based on the soil parent materials. In addition, each group was separated based on their land-use conditions. The descriptive statistics of the soil physicochemical properties for each group are presented in Table 1. In the uncultivated topsoil, the soil pH ranged from 4.1 to 8.2. Generally, the soil derived from carbonate rock showed alkaline properties, and the soil derived from red residua or sand-shale displayed acidic characteristics. The ranges of the basic physicochemical properties of the uncultivated topsoil were 12.7 to 98.6 g kg<sup>-1</sup> for SOM, 9 to 18 for C/N, 6.2 to 34.8 cmolc kg<sup>-1</sup> for CEC, and 28 to 71% for clay (<0.01 mm) content. The average values for the pH, SOM, C/N, CEC, and clay content in the uncultivated topsoil were 5.38, 42.1 g kg<sup>-1</sup>, 13, 15.9 cmolc kg<sup>-1</sup>, and 49%, respectively. Results of ANOVA show that the pH, C/N, CEC, and clay contents of soils derived from carbonate rock were not significantly different from those of dry lands, paddy fields, and uncultivated soils (p > 0.05). Dry land SOM was significantly lower than those in the other land-use types (p < 0.05). In the soils derived from red residua, the soil physicochemical properties changed greatly because of land-use conditions. The pH, SOM, and CEC in the paddy fields rose significantly (p < 0.05) and the C/N and clay content significantly decreased (p < 0.05) compared with those in the

uncultivated soils. The soil pH, C/N, and clay content in the dry lands were significantly different from those in the uncultivated soils (p < 0.05). In the soils derived from sand-shale, the influence of land use on the soil physicochemical properties was similar to that on the soils derived from residua. The pH, SOM, and clay content in the paddy fields were significantly higher than those in the other two land-use types (p < 0.05). The SOM and C/N ratio in the dry lands were lower than those in the uncultivated soils.

## **Trace Element Concentrations**

The average concentrations of the trace elements in the uncultivated topsoil were 0.1 mg kg<sup>-1</sup> for Cd, 21.7 mg kg<sup>-1</sup> for Pb, 53.6 mg kg<sup>-1</sup> for Cr, 0.06 mg kg<sup>-1</sup> for Hg, and 6.72 mg kg<sup>-1</sup> for As. The trace elements in the agricultural topsoil increased rapidly compared with those in the uncultivated topsoil, respectively. The contents of Cd, Pb, Cr, Hg, and As in agricultural topsoil were 2.5, 2.5, 3.2, 3, and 3 times greater than the values in the uncultivated topsoil. The contribution of anthropogenic trace elements was significant.

Table 2 lists the concentrations of trace elements and statistical characteristics of topsoil from the same parent materials but under different land-use conditions. Regardless of the parent material, the trace element concentrations in the agricultural soils were greater (p < 0.05) than those in the respective uncultivated soils. Most of the average trace element concentrations in the paddy fields were higher than those in dry lands. In the paddy soils, the content of Pb in soil derived from carbonate rock, the contents of Cd, Cr, and Hg in soil derived from red residua, and the contents of Cd, Pb, and Cr in soil derived from sand-shale were significantly higher than those in dry lands (p < 0.05).

Figure 2 shows the effect of the parent material on soil trace elements under the same land-use conditions. Based on the multicomparison results, most of the mean contents of the trace elements in the uncultivated topsoil derived from carbonate rock were slightly higher than those in soils derived from other parent materials. However, the parent materials did not significantly influence the concentrations of the trace elements, except for that of Hg (p < 0.05). The Hg content in the topsoil derived from carbonate

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Parent material	Land-use condition	No. of soil samples	рН	Soil organic matter	C/N†	CEC‡	Clay content (<0.01 mm)
		n		g kg <sup>-1</sup>		cmolc kg <sup>-1</sup>	%
Carbonate rock	Dry land	60	7.7 ± 0.3 a§¶	34.9 ± 14.5a	$11.4 \pm 1.4$ a	17.2 ± 5.0 a	48.1 ± 11.9 a
	Paddy field	75	7.7 ± 0.4 a	45 ± 14 b	11.3 ± 1.3 a	18.2 ± 4.3 a	44.4 ± 15.5 a
	Uncultivated soil	19	7.7 ± 0.3 a	48.7 ± 21.7 b	11.9 ± 2.1 a	19.4 ± 5.4 a	44.2 ± 12.6 a
Red residua	Dry land	56	6 ± 0.8 a	32.9 ± 10.1 a	11.4 ± 1.5 a	14.1 ± 3.5 a	58.5 ± 7.8 a
	Paddy field	76	6.3 ± 0.7 b	44.1 ± 10.8 b	11.4 ± 1.3 a	16.1 ± 3.5 b	60.4 ± 7.8 b
	Uncultivated soil	13	$4.6 \pm 0.7 \ c$	36.2 ± 11.4 a	14 ± 1.9 b	13.5 ± 3.6 a	62.7 ± 6.2 c
Sand-shale	Dry land	149	5.9 ± 0.8 a	33.5 ± 10.3 a	11.7 ± 1.6 a	14.8 ± 4.4 a	50.9 ± 9.4 a
	Paddy field	168	6.2 ± 0.8 b	43.3 ± 11.7 b	10.9 ± 1.3 b	16 ± 3.9 b	51.7 ± 10.1 b
	Uncultivated soil	57	$4.9\pm0.8$ c	40.6 ± 18.1 b	13 ± 2.2 c	15.5 ± 5.3 ab	50.5 ± 5.3 c

† Mass ratio.

‡ CEC, cation exchange capacity.

§ Mean ± SD.

¶ In soils derived from the same parent material, means with different letters indicate significantly different ( $p \le 0.05$ ) probability levels (LSD) within each column.

(p < 0.05). In the paddy fields, Hg and Cr contents were influenced by the different parent materials. The soils derived from carbonate rock had higher Hg and Cr concentrations than the other soil types. In dry lands, the concentrations of Hg, Cd, and Cr in the soils derived from carbonate rock were higher than those of the other soil types (p < 0.05). The content of Pb in the soils derived from red residua was higher than those of the other soil types (p < 0.05).

# Discussion

# **Effects of Different Land-Use Types**

Several reports have suggested that agricultural soils receive a comparatively high input of anthropogenic trace elements, possibly related to the use of agrochemicals, atmospheric deposition, and other soil amendments (Alloway, 1995; Kachenko and Singh, 2006; Montagne et al., 2007; Smith et al., 1996). According to the present investigation, fertilizers contain certain amounts of trace elements, and the contents of trace elements in organic manure are higher than those in inorganic fertilizers, except for Cr (Chen et al., 2006). In fact, fertilizer applications were not consistent under different landuse conditions. Generally, the amount of organic manure applied is 15 to 45 t  $ha^{-1}yr^{-1}$  and that of inorganic fertilizers is 0.45 0.6 t ha<sup>-1</sup> yr<sup>-1</sup> in the paddy fields. In dry lands, the amount of organic manure applied is 1.5 5 t ha<sup>-1</sup> and that of inorganic fertilizers is 0.75 to 1.2 t ha<sup>-1</sup>. Thus, the amount of organic manure input in the paddy fields is clearly higher than in dry lands, but the amount of inorganic fertilizer input in the paddy fields is lower. In addition, Table 2 shows that most of the contents of trace elements in the paddy soils were higher than those in dry lands. Therefore, organic manures are key anthropogenic sources of trace elements and greater contributors in the paddy fields than in dry lands. Moreover, agricultural soils are widely distributed on the sloped land in this region. For the paddy fields, the hillside is terraced, which is an effective method of reducing soil erosion and facilitates the accumulation of trace elements. In dry lands, local farmers always cultivate the soil along the slope, which usually results in more serious soil loss and transports trace elements during soil erosion. Thus, paddy field cultivation has caused greater trace elements accumulation in soil.

## **Effect of the Parent Material and Soil Properties**

The initial source of trace elements in soils is typically the parent material, and the input of anthropogenic trace elements from fertilizers, manures, and other sources may be lower than their initial contents in soils (Facchinelli et al., 2001). Thus, the content of trace elements in agricultural soils is strongly associated with the parent material even after long-term cultivation (Mico et al., 2006; Montagne et al., 2007; Wang and Wei, 1995). Furthermore, trace element concentrations in soil may differ with different parent materials (Station, 1990). The uncultivated soils derived from carbonate rock showed higher average trace element contents (except for that of Pb) than the other soil types. However, no significant differences among the three types of topsoil were observed, except for that of Hg concentration (p > 0.05) (Fig. 2). Similarly, with the exception of Cr and Hg, the contents of the other trace elements were not significantly different among the paddy topsoil samples. The effects of the parent materials on the contents of trace elements in the uncultivated or paddy soils were clearly not consistent. However, the soil parent material greatly influenced the contents of trace element in dry lands. The parent materials clearly not only serve as sources of trace elements in soil but may also control the loss and accumulation of trace elements by affecting soil physicochemical properties (Facchinelli et al., 2001).

Soil pH influences the mobilization of cations and the solubility of heavy metals (Basta et al., 1993; Kashem and Singh, 2001; Yang et al., 2006). Normally, under acidic conditions, most of the trace metal elements are solubilized and made available for plant use. In contrast, alkaline conditions promote the accumulation of trace metals in soils (Nan et al., 2002; Remon et al., 2005). In the current study, the pH range in dry lands was from 4.2 to 8.3, and the topsoil derived from carbonate rock showed a significantly higher value than the other soils. Moreover, the correlations between the trace elements and soil pH were significant in dry lands (p < 0.01) (Table 3), which indicates that soil pH is a key factor in controlling the accumulation of trace elements in dry lands. In the paddy fields, the pH of the soils derived from carbonate rock was basic, whereas the other soils were acidic (below 7.0). However, waterlogging may cause the pH of acidic soils to increase while drainage may cause a decrease in pH. Ultimately, the soil pH in the paddy fields should be close to neutral. Thus, the correlations among the trace elements and soil

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Table 2. Descri	prive statistics of trac	Le metal contents m ac	gricultural topsoli samples.

Parent material		Cd	Pb	Cr	Hg	As
				mg kg <sup>-1</sup>		
Carbonate rock	Dry land	0.28 ± 0.15 a†‡	50.9 ± 27.1 a	179 ± 125 a	0.24 ± 0.15 a	18.8 ± 16.9 a
	Paddy field	$0.29 \pm 0.13$ a	62.4 ± 40.2 b	236 ± 159 a	$0.24 \pm 0.16$ a	20.2 ± 16.7 a
	Uncultivated soil	$0.12 \pm 0.03$ b	25 ± 10.2 c	63 ± 47 b	0.09 ± 0.06 b	8.1 ± 4.6 b
Red residua	Dry land	$0.20 \pm 0.14$ a	66.1 ± 46.4 a	130 ± 79 a	$0.12 \pm 0.07$ a	21.6 ± 13.1 a
	Paddy field	0.29 ± 0.17 b	63.2 ± 37.6 a	177 ± 87 b	0.18 ± 0.11 b	19.5 ± 11.2 a
	Uncultivated soil	$0.12 \pm 0.09$ a	26.5 ± 9.5 b	58 ± 32 c	$0.08 \pm 0.16$ a	$7.4 \pm 9.7 \text{ b}$
Sand-shale	Dry land	0.23 ± 0.11 a	48 ± 29.1 a	146 ± 116 a	$0.18 \pm 0.19$ a	21.1 ± 17 a
	Paddy field	0.26 ± 0.13 b	54.7 ± 31.7 b	184 ± 114 b	$0.21 \pm 0.18$ a	20.9 ± 16.7 a
	Uncultivated soil	0.09 ± 0.06 c	20.8 ± 10.2 c	51 ± 30 c	$0.04\pm0.05~b$	$6.8\pm8.1$ b

†Mean ± SD.

 $\pm$  In soils derived from the same parent material, means with different letters indicate significantly different ( $p \le 0.05$ ) probability levels (LSD) within each column.

pH are generally not significant, except for Cr and Hg content (p > 0.05). However, the paddy soils derived from carbonate rock contained a certain amount of carbonate, which rendered the soil pH neutral to slightly alkaline. Therefore, the increase concentration of Cr and Hg were partly due to soil alkalinity.

Soil organic matter facilitates the formation of trace elements to chelate complexes, which are difficult to biodegrade and result in trace element enrichment (Chen et al., 1997; Huang and Jin, 2008; Wu et al., 2003). In addition, the adsorptions of some trace elements were found to increase with higher SOM content (Alloway, 1995; Bradl, 2004; Weng et al., 2001). A number of studies showed that organic matter has a positive linear relationship with trace elements (Dragovic et al., 2008; Li et al., 2009; Liu et al., 2006; Mico et al., 2006; Rodríguez Martín et al., 2006). In the current study, because of the large amount of organic manures added into the paddy fields, the trace elements (except for Pb and Hg) exhibited significant relationships with SOM (p < 0.01) (Table 3), which means that SOM is a key factor affecting the accumulation of trace elements. By contrast, the relationships among the trace elements and SOM were very weak in dry lands (Table 3), in which the loss in SOM was high and the effect of SOM on the accumulation of trace elements was weakened.

Normally, the C/N ratio reflects the humification level of SOM and soil microbial activity. Farmers adjust the C/N ratio and satisfy the crop N nutrition by using inorganic chemical fertilizers to improve the release of available nutrition from SOM. A high C/N ratio suggests poor SOM humification. Moreover, some studies have reported on whether the C/N ratio indirectly affects the forms and contents of trace elements by changing the humification level of SOM and soil microbial activity. In dry lands, the C/N ratio showed a significant positive correlation with the content of trace elements, with the exception of As, which suggests that inorganic chemical fertilizers can solubilize trace elements, resulting in the transference of these trace elements. In the paddy fields, no significant relationships among the C/N ratio and trace elements were observed. The amount of inorganic fertilizer input in paddy fields was minimal compared with that of organic manures, which contain more trace elements. Thus, the input of inorganic fertilizers into the paddy field did not significantly affect the contents of trace elements (p > 0.05).

The CEC is normally composed of alkaline and alkalineearth cations and does not include a substantial proportion of the soil trace element content. The results show that CEC did not significantly relate to any trace elements in dry land, but the relationships reached a significant level in paddy field, except for that with Pb (p < 0.05) (Table 3). This result suggests that the exchangeable trace elements in the paddy fields were influenced by the total trace element content; however, this was not the case in dry lands.

Chen et al. (1999) reported that the concentrations of the five trace elements are strongly correlated with clay content in Florida surface soils. Mermut et al. (1996) also found strong correlations between concentrations of As, Cd, Cr, and Pb and clay content in Canada soils. The authors suggested that trace elements can be adsorbed on the surface of clay and remain in the soil. Similarly, the correlations among the trace elements and clay content (<0.01 mm) were significant (p < 0.05) in dry



Dry land Paddy field Uncultivated

Fig. 2. Effects of different parent material on soil trace elements in same land-use type.

lands (Table 3). In contrast, some researchers presented inverse or weak correlations among trace elements and clay content (Li et al., 2009; Lucho-Constantino et al., 2005; Navas and Mach, 2002). They suggested that clay content was not important in determining the distribution and content of trace elements in soils. In the paddy fields of the current study, the concentrations of trace elements (except that of Pb) were not significantly correlated with the clay content (p > 0.05) (Table 3).

Table 3. Pearson correlation coefficient matrix between the concentrations of trace elements and basic p	hys	sicochemical	prop	pertie	s
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	pH (H <sub>2</sub> O)	SOM	C/N	CEC	<0.01 mm clay content
		Uncu	ltivated soil		
Cd	0.10	0.08	-0.04	-0.14	0.04
Pb	0.08	0.01	-0.05	0.03	0.21
Cr	0.06	-0.01	-0.12	-0.18	0.08
Hg	0.15	-0.07	-0.13	0.01	-0.11
As	0.01	-0.06	0.01	0.02	0.12
		D	ry land		
Cd	0.31**	0.04	0.18**	0.05	0.28**
Pb	0.20**	0.10*	0.17**	0.01	0.12*
Cr	0.17**	-0.03	0.11*	-0.01	0.11*
Hg	0.16**	0.05	0.22**	0.06	0.22**
As	0.28**	0.11*	-0.02	0.05	0.15**
		Pa	ddy field		
Cd	0.09	0.14**	0.14*	0.13*	-0.05
Pb	0.07	0.02	0.03	0.05	0.18**
Cr	0.11*	0.29**	0.08	0.28**	-0.10
Hg	0.16**	0.01	0.04	0.15**	-0.05
As	0.00	0.16**	0.06	0.17**	-0.01

\* Correlation is significant at the 0.05 level.

\*\* Correlation is significant at the 0.01 level.

### **Correlation Matrix and Principal Component Analysis**

Several studies reported on the strong relationships among soil trace elements, indicating that the trace elements are from the same sources (Li et al., 2009; Rodríguez Martín et al., 2006). Thus, PCA was used to identify the sources of the trace elements (Mico et al., 2006). In the present study, with the exception of Pb/Cd, Pb/Hg, and Hg/Cr, the relationships among the trace elements were significant in the uncultivated soils (p < 0.01) (Table 4). These results suggest that the contents of Pb, Cd, Cr, and Hg were significantly different in the soils derived from different parent materials or were closely related to the parent

Table 4. Pearson correlation coefficient matrix among the concentrations of trace elements.

	Cd	Pb	Cr	Hg
	L	Incultivated so	il	
Cd				
Pb	0.08			
Cr	0.38**	0.33**		
Hg	0.29**	0.03	0.13	
As	0.54**	0.39**	0.31**	0.296**
		Paddy field		
Cd				
Pb	-0.04			
Cr	0.38**	0.04		
Hg	0.23**	-0.06	0.12*	
As	0.28**	0.06	0.25**	-0.07
		Dry land		
Cd				
Pb	0.14*			
Cr	0.45**	0.03		
Hg	0.32**	0.09	0.29**	
As	0.35**	0.15**	0.10*	-0.05

\* Correlation is significant at the 0.05 level.

\*\* Correlation is significant at the 0.01 level.

materials. The As content in the uncultivated soils showed significant positive relationships with the other trace elements, which means that the effect of the parent materials on As may have been weak. In fact, our data did not show significant differences in the trace element contents, with the exception of Hg, among the soils derived from different parent materials (p > 0.05). However, the uncultivated soils derived from carbonate rock showed higher average trace element contents, except for that of Pb. In the agricultural soils (either the in the paddy fields or in dry lands), significant relationships among Cd, Cr, and As were observed. The contents of the trace elements were far higher than those of the uncultivated soils because of the use of fertilizers. Moreover, the association of Pb with the other elements was weak, meaning that the loss and accumulation of Pb differed from those of the other trace elements. Some reports showed that the soil parent material, Pb-containing agrochemicals and Pb emitted from gasoline combustion are the main sources of Pb in agricultural soils (Markus and McBratney, 2001). As indicated by our previous research, Pb produced by gasoline combustion is far less than that from parent materials and agrochemicals (Wu et al., 2008). In addition, Pb showed positive linear relationships with soil pH, SOM, C/N, and clay content in dry lands, as well as with the clay content in the paddy fields. These results indicate that Pb content is controlled not only by sources but also by soil properties.

Results of PCA show that Cd, Cr, and As comprised the first component in the paddy field (Fig. 3), which can be considered as an anthropogenic component. The concentrations of these trace elements were far beyond those of the uncultivated soil background, and significant relationships among these trace elements were observed (p < 0.01). In addition, these trace elements were closely related to SOM, which is always controlled by organic manures in paddy soils. In dry lands, the first component included Cd, Cr, and Hg (Fig. 3), which can be derived from inorganic fertilizers. The concentrations of these trace elements were far beyond those of the uncultivated soil



Fig. 3. Principal component analysis loading plots for the two rotated components.

background, and these trace elements had no significant relationships with SOM. In addition, Pb and As, which can be derived from organic manures, formed the second component. These two trace elements were closely related to SOM. Furthermore, the government of China has forbidden the use of inorganic forms of As in agriculture as insecticides, herbicides, fungicides, desiccants, defoliants, and additives to animal feeds since the 1970s (Chen et al., 1997).

#### Conclusions

The effect of land-use conditions on soil trace elements was evaluated by discussing the difference in the cultivation methods between paddy fields and dry lands based on statistical analysis. The soil properties controlling the loss and accumulation of trace elements were also analyzed. A number of principal conclusions are presented as follows:

- Our data reveal that the agricultural topsoil, either in paddy fields or in dry lands, had significantly higher trace element contents compared with the uncultivated soil background, which indicates that the contribution of anthropogenic trace elements is significant in agricultural soils. The accumulation of trace elements in the paddy fields is more significant than in dry lands because of the difference in fertilizer application and in cultivation methods.
- 2. In the paddy fields, the topsoil derived from carbonate rock had higher Cr and Hg contents. The concentrations of Cd, Pb, and As in the soils from different parent materials were not significant (p > 0.05). Cadmium, Cr, and As had significant relationships with SOM (p < 0.01) and were not closely related to other soil properties (p > 0.05). These results indicate that organic manures were the key sources of these trace elements, and Cd, Cr, and As were strongly controlled by anthropogenic activities. Lead and Hg formed the second component in PCA analysis. Lead and Hg did not correlate with SOM but were closely related to soil pH or physical clay content. These results suggest that soil properties can control the loss and accumulation of Pb and Hg to a certain extent.
- 3. In dry lands, the topsoil derived from carbonate rock had higher Cr, Cd, and Hg contents, and the topsoil derived from red residua had a higher Pb content. The contents of trace elements were well correlated with soil pH, C/N, and clay content (<0.01mm) (p < 0.05). In the soil

derived from carbonate rock, the high background values of the trace elements, as well as the alkaline conditions, contributed to the significant enrichment of Cd, Cr, and Hg. In the soils derived from red residua, the Pb and As contents were the highest among the soil categories, partly because these soils have higher clay contents. (<0.01 mm). Principal component analysis and correlation analysis showed that Cd, Cr, and Hg were mainly derived from inorganic fertilizers, whereas Pb and As were mainly from organic manures. These results show that parent materials not only serve as sources of trace elements but also control the loss and accumulation of trace elements by affecting the physicochemical properties of soils.

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