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Original Research

Soil Metal Contamination and Fractionation of Tea Plantations: Case Studies in a Normal Tea Garden and in a Restored Mineland Tea Stand

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Abstract

Extensive soil samples were collected from two tea gardens (a noted tea garden in Guilin and a mineland tea stand in Bayi Mn mine) in Guangxi to assess the heavy metal contamination level. The concentrations of Cu, Zn, Mn, Pb, Cd, and Al in garden soil were determined by ICP-AES. The results showed that the total concentrations of 6 metals (Cu, Zn, Mn, Pb, Cd, Al) in mineland tea garden soils were all higher than their soil background concentrations in Guangxi. Except for Cu, the concentrations of 5 other heavy metals in mineland tea garden were all higher than those in Guilin tea garden. Although tea garden soils contained high concentrations of Cd, most of the Cd existed in the form of residual fractions that were unavailable to higher plants according to the results of the sequential extraction. Similarly, Mn was dominated by iron-manganese oxide and residual fractions in mineland tea garden soils, but dominated by residual fractions in Guilin tea garden soils. Pb was dominated by residual and iron-manganese oxide fractions in mineland tea garden soils but by residual, iron-manganese oxide and exchangeable fractions in Guilin tea garden soils. Cu, Zn, and Al were all dominated by residual fractions in soils. A simple pollution index and nemerow index assessment were used to assess heavy metal contamination in the two tea gardens. Results showed that Cd was the key pollutant in tea garden soils. According to simple pollution indexes, no Pb, Zn, or Cu pollution was found. However, because of the high contribution of Cd in composite pollution index, bulk soils (in-between and outside tea tree rows) in Guilin tea garden met the moderate pollution level with nemerow index assessment. All other tea garden soils reached the heavy pollution standard.

Keywords: metal contamination, tea garden, bulk soils, rhizosphere soils, metal fractionation

Introduction

Tea is a popular beverage in the world. It is reported that drinking tea has beneficial effects on immune function and inflammation, prevents many diseases, is responsible antioxidative detoxification, and removes cadmium [1-5]. However, heavy metal accumulation and contamination in tea has caused concerns for consumers and producers as discussed in many research publications [6-10].

Heavy metal pollution is a special concern as heavy metals are difficult to biodegrade and liable to accumulate. Particularly, heavy metals are available to crops in acidic soils [11-13]. Tea plant is an unusual crop because it usually grows in acidic soil and its planting makes soil more acidic

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[14-16], which may urge accumulation of heavy metals in tea. Therefore, heavy metal concentration of soils in tea gardens has remained a concern in recent years [13, 17-18].

Rhizosphere soils and bulk soils have different chemical and biological characteristics as root activity influences significantly the biological properties of soil, which can affect the bioavailability of heavy metals in soil [19-21]. In this paper, extensive soil samples, including rhizosphere and bulk soils (including bulk soil in-between tea tree rows and bulk soil outside tea tree rows) were collected from two tea gardens (a noted tea garden in Guilin and a mineland tea stand in Bayi Mn mine) in Guangxi to assess the metal (Cu, Zn, Mn, Pb, Cd, Al) contamination level. On the other hand, as the total amount of heavy metals in soil cannot be used to assess the biological availability of heavy metals effectively, metal species are separated into five operationally defined fractions, which are also discussed in this paper.

Materials and Methods

Study Sites

The two study sites are located at Guangxi Zhuang Autonomous Region in southern China. One is a noted tea garden in Lingui county of Guilin (short name: L tea garden), which belongs to a subtropical continental climate with an annual average temperature of 19.1ºC and an average rainfall of 1869 mm. The other, located in Xingbin district of Laibin city, is a mineland tea stand in Sirong mining area of Bayi Mn mine (short name: S tea garden) with annual temperature of 19.7ºC and annual precipitation of 1373.7 mm. L tea garden has been cultivated for 40-50 years, and S tea garden was constructed as a reclamation project of Mn mine wasteland in 1992.

Soil Sampling

Three kinds of surface soil (0-20 cm depth) samples (rhizosphere soil, bulk soil in-between tea tree rows and bulk soil outside tea tree rows) were gathered once every two months respectively from the two tea gardens from August 2007 to June 2008. Section soil samples were gathered from the two gardens in October 2007. Soil profile depth is 100 cm, with 0~30cm being Layer A (LA, SA), 30~60cm Layer B (LB, SB), and 60~100cm Layer C (LC, SC), from top to bottom, respectively.

Chemical Analysis

All soil samples were air-dried, homogenized, and pulverized, and passed through a 100 mesh sieve. Precisely weighted soil samples were digested with concentrated HNO_3+HF+$HClO_4$ (5:2:1 v/v) using a microwave digesting system (Mars 5, CEM). Cu, Zn, Mn, Pb, Cd, and Al concentrations in the digest solution were determined by ICP-AES (PE Optima 2000 DV).

Quality assurance of metal determination was performed using certified Mn standard liquid added to the digested solution, and the average recovery rate was around 93%.

Fractionations of Metal Speciation

The speciation of selected metals, namely Cu, Zn, Mn, Pb, Cd, and Al of the section soil samples, was determined by the Tessier five-step extraction method [22]. The multiple-step sequential extraction scheme of the metals generated the following forms:
(1) exchangeable
(2) bound to carbonate
(3) bound to Iron and Manganese
(4) bound to organic matter
(5) residual

Results

Heavy Metal Concentrations of the Soil Samples of the Two Tea Gardens

Results of total heavy metal concentrations in the tea garden in Bayi Mn mine wasteland (Table 1) showed that Cu, Zn, Mn, Pb, Cd, and Al ranged 17.3~34.8 $mg{\cdot}kg^{-1}$, 15.4~102.9 $mg{\cdot}kg^{-1}$, 116.3~654.7 $mg{\cdot}kg^{-1}$, 20.6~45.1 $mg{\cdot}kg^{-1}$, 1.1~3.1 $mg{\cdot}kg^{-1}$, and 75,105.4~319,353.3 $mg{\cdot}kg^{-1}$. Annual average of Cu concentration of rhizosphere soil, bulk soil in-between tea tree rows, and bulk soil outside tea tree rows was respectively 24.3 $mg{\cdot}kg^{-1}$, 22.9 $mg{\cdot}kg^{-1}$, and 25.1 $mg{\cdot}kg^{-1}$, which were slightly higher than that of the soil background value of Guangxi (20.8 $mg{\cdot}kg^{-1}$). Annual average of Zn and Pb concentration was respectively 1.4~1.4 and 1.6~1.8 times higher than that of the soil background value of Guangxi (46.4 $mg{\cdot}kg^{-1}$, 18.8 $mg{\cdot}kg^{-1}$). Annual average of Mn concentration was 1.8~2.6 times higher than that of the soil background value of Guangxi (172.6 $mg{\cdot}kg^{-1}$), and it followed the pattern of bulk soil in-between tea tree rows > rhizosphere soil > bulk soil outside tea tree rows. Annual average of Cd and Al concentration far exceeded that of the soil background value of Guangxi with Al concentration of the three kinds of soils being up to 15,989.2~20,527.2 times and showing the pattern of rhizosphere soil > bulk soil outside tea tree rows > bulk soil in-between tea tree rows.

Results of total heavy metal concentrations in the tea garden in Guilin (Table 2) showed that Cu, Zn, Mn, Pb, Cd, and Al ranged in 22.4~39.2 $mg{\cdot}kg^{-1}$, 16.8~119.7 $mg{\cdot}kg^{-1}$, 20.7~84.3 $mg{\cdot}kg^{-1}$, 13.7~32.3 $mg{\cdot}kg^{-1}$, 0.3~2.3 $mg{\cdot}kg^{-1}$, and 33,498.6~105,537.3 $mg{\cdot}kg^{-1}$. In comparison with heavy metal concentrations in the tea garden in Bayi Mn mine wasteland, annual average of the first metal in this garden were higher, but those of the latter five metals were much lower. Annual average of Cu, Zn, and Mn concentrations of the three kinds of soils followed the same pattern of bulk soil in-between tea tree rows>bulk soil outside tea tree rows >rhizosphere soil.

Soil Heavy Metal Pollution Assessment

Single element pollution index and nemerow index were used to assess heavy metal contamination in the two tea gardens (Table 3). All of the indices were calculated

Table 1. Soil heavy metal concentrations (mg·kg^{-1}) in the tea garden in Bayi Mn wasteland (mean±SE, n=3).

Sampling time	Cu	Zn	Mn	Pb	Cd	Al
Rhizosphere soil						
2007.08.	18.5±0.4	68.6±4.9	554.1±26.6	32.5±3.2	2.6±0.2	76480.8±1061.7
2007.10	18.2±0.8	66.7±3.1	318.7±5.4	20.6±4.1	2.2±0.1	87158.3±3415.8
2007.12.	26.2±0.7	77.1±0.4	395.3±3.3	35.0±1.7	3.1±0.2	91455.9±5656.6
2008.02	28.1±0.5	102.9±4.5	489.5±38.9	43.3±4.6	2.8±0.3	117559.9±11893.0
2008.04	27.6±1.1	41.5±6.1	290.4±39.2	25.1±3.0	1.8±0.4	319353.3±2135.4
2008.06.	27.3±3.8	39.8±16.5	300.2±7.7	23.0±6.3	1.3±0.1	112615.7±2962.4
Average	24.3±1.9	66.1±9.6	391.4±44.7	29.9±3.5	2.3±0.3	134104.0±37592.5
Bulk soil in-between tea tree rows						
2007.08.	18.3±0.3	71.8±1.9	464.4±3.5	28.8±2.2	2.4±0.1	86450.7±3359.5
2007.10	18.6±0. 8	73.4±4.4	440.5±33.3	29.7±1.7	2.2±0.3	77696.8±12974.4
2007.12.	26.0±1.1	84.1±1.1	526.6±12.9	45.1±2.6	3.1±0.2	101406.9±3176.4
2008.02	26.7±0.4	88.0±5.5	654.7±49.9	42.8±1.7	2.8±0.3	119920.3±11362.0
2008.04	20.1±5.7	24.5±9.1	200.8±74.3	28.8±1.7	1.2±0.2	136966.9±11862.2
2008.06.	27.8±0.8	47.0±3.2	367.5±82.8	28.1±1.9	1.3±0.1	104301.7±3704.7
Average	22.9±1.8	64.8±10.0	442.4±62.4	33.9±3.2	2.2±0.3	104457.2±8842.5
Bulk soil outside tea tree rows						
2007.08.	17.3±0.4	88.9±9.6	538.9±16.0	28.3±1.0	2.6±0.1	75105.4±258.4
2007.10	19.3±0.9	74.0±1.6	270.7±16.2	24.9±2.7	2.4±0.3	96519.9±4050.2
2007.12.	27.6±0.3	81.8±2.0	332.1±5.2	31.7±3.1	2.8±0.1	118646.7±5367.3
2008.02	27.8±0.9	90.9±3.6	293.7±28.2	38.2±2.6	2.7±0.2	132882.3±13939.5
2008.04	30.0±4.8	15.4±2.8	116.3±60.3	29.0±2.2	1.7±0.1	243876.8±1182.0
2008.06.	28.7±0.5	47.6±11.1	256.7±114.0	27.4±3.9	1.1±0.3	102791.9±9098.5
Average	25.1±2.0	66.4±12.0	301.4±56.2	29.9±1.9	2.2±0.3	128303.8±24469.4

based on Chinese Environmental quality standard for soils (GB 15618-1995 II pH<6.5). As Mn and Al were not involved in the standard, only Cu, Zn, Pb, and Cd were evaluated. The results (Table 3) showed that the concentration level of Cu, Zn, Pb of soil in the two tea gardens were safe. However, the soils were seriously polluted by Cd. Comprehensive indices of soil pollution showed that rhizosphere soils of both tea gardens and the bulk soils of S tea garden were seriously polluted, and the bulk soils of L tea garden were moderately polluted. By comparing the pollution levels of the two tea gardens, we found that S tea garden was more seriously polluted by heavy metals.

Speciation Analysis of Metal Elements of Section Soil

The results of speciation analysis of metal elements of the section soil samples from the two tea gardens were shown in Fig. 1. Most of the Cd existed in the form of residual fraction. Mn was dominated by iron-manganese and residual fraction in S tea garden soils but dominated by residual fraction in L tea garden soils. Pb was dominated by residual and iron-manganese fraction in S tea garden soils, while it was dominated by residual, iron-manganese, and exchangeable fraction in L tea garden soils. Cu, Zn, and Al were all dominated by residual fraction in soils.

Correlation Analysis on the Concentrations of Heavy Metals in Soils

Correlation analysis was conducted among the total concentrations of heavy metals for the two kinds of tea garden soils. There was a highly significant positive correlation between Mn and Zn, between Pb and Zn, between Pb and Mn, between Cd and Zn, between Cd and Mn, and between Cd and Pb (Table 4). However, there existed a highly significant negative correlation between Mn and Cu. Al and other heavy metals showed no obvious correlation.

Table 2. Soil heavy metal concentrations in the tea garden in Guilin (mean±SE, n=3).

Sampling time	Cu	Zn	Mn	Pb	Cd	Al
Rhizosphere soil						
2007.08.	27.1±0.3	26.0±1.6	83.3±0.4	16.1±0.1	1.6±0.3	37456.9±956.4
2007.10	26.3±0.3	29.0±1.0	27.7±0.3	16.1±2.3	1.7±0.2	43723.3±1573.5
2007.12.	29.5±0.7	32.5±8.3	32.1±1.9	21.9±1.8	2.1±0.2	59537.0±414.4
2008.02	29.3±0.6	24.2±4.3	25.3±2.0	30.3±2.5	2.0±0.4	69610.0±1426.0
2008.04	27.8±0.4	16.8±0.9	34.0±5.7	20. 6±2.0	0.4±0.1	105537.3±2392.1
2008.06.	31.7±0.3	24.8±1.3	31.6±1.4	13.7±0.2	0.3±0.1	98966.0±4858.7
Average	28.6±0.8	25.5±2.2	39.0±9.0	19.8±2.5	1.3±0.3	69138.4±11484.2
Bulk soil in-between tea tree rows						
2007.08.	24.1±0.1	26.1±1.4	84.3±1.4	16.5±0.8	1.1±0.1	33761.5±238.5
2007.10	24.2±0.7	18.0±0.7	23.8±0.4	16.6±1.8	1.3±0.2	42201.8±3281.6
2007.12.	38.3±0.3	34.8±3.7	34.7±0.2	32.3±2.4	2.1±0.2	62226.1±2351.7
2008.02	37.2±3.6	119.7±86.6	59.1±11.6	30.8±5.0	1.7±0.5	55706.2±4941.6
2008.04	34.3±0.5	24.6±0.6	62.9±14.9	22.0±0.7	0.4±0.1	56681.5±3244.6
2008.06.	28.4±0.4	26.6±1.6	41.1±2.9	20.3±1.1	0.3±0.1	69721.9±1245.8
Average	31.1±2.6	41.6±15.8	51.0±9.0	23.1±2.8	1.1±0.3	53383.2±5389.7
Bulk soil outside tea tree rows						
2007.08.	22.4±0.1	23.3±2.1	73.6±2.2	17.5±0.2	1.5±0.2	33498.6±585.2
2007.10	23.8±0.2	19.3±0.2	20.7±0.5	21.0±1.9	1.1±0.1	41102.7±1243.9
2007.12.	39.2±0.2	30.8±2.1	38.5±0.1	25.8±1.1	2.3±0.3	72094.2±3018.2
2008.02	36.5±0.4	53.0±28.1	38.7±0.5	18.6±1.4	0.9±0.1	57769.1±8071.5
2008.04	27.3±1.0	37.5±11.5	51.5±2.1	19.6±5.7	0.7±0.3	76349.2±1495.6
2008.06.	25.2±0.7	20.9±7.2	30.4±6.6	16.1±2.1	0.3±0.1	61915.9±1799.5
Average	29.1±2.9	30.8±5.2	42.3±7.5	19.8±1.4	1.2±0.3	57121.6±6908.8

Table 3. The pollution indices and their pollution grade in the two tea gardens.

Soil sample	Cu		Zn		Pb		Cd		Nemerow index	
	Pi	grade	Pi	grade	Pi	grade	Pi	grade	P_N	grade
S tea garden										
Rhizosphere soil	0.16	safe	0.33	safe	0.12	safe	7.61	serious	5.57	serious
Bulk soil in-between tea tree rows	0.15	safe	0.32	safe	0.14	safe	7.18	serious	5.26	serious
Bulk soil outside tea tree rows	0.17	safe	0.33	safe	0.12	safe	7.31	serious	5.36	serious
L tea garden										
Rhizosphere soil	0.19	safe	0.13	safe	0.08	safe	4.41	serious	3.23	serious
Bulk soil in-between tea tree rows	0.21	safe	0.21	safe	0.09	safe	3.79	serious	2.79	medium
Bulk soil outside tea tree rows	0.19	safe	0.15	safe	0.08	safe	3.83	serious	2.81	medium

Pi – pollution index
P_N – nemerow index

Table 4. Correlation coefficient between different heavy metals' concentrations in soils (n=36).

	Zn	Mn	Pb	Cd	Al
Cu	-0.147	-0.504*	-0.002	-0.231	0.000
Zn		0.721*	0.708*	0.707*	0.087
Mn			0.721*	0.698*	0.300
Pb				0.754*	0.325
Cd					0.159

*highly significant correlation (P<0.01)

Discussion

The total concentrations of 6 heavy metals (Cu, Zn, Mn, Pb, Cd, Al) in mineland tea garden soils were all higher than their soil background concentrations in Guangxi. Except for Cu, the concentrations of 5 other heavy metals in the mineland tea garden were all higher than those in Guilin tea garden. In particular, the concentrations of Mn and Cd in the mineland tea garden were 7.1~10.0 and about 2 times higher than that in Guilin tea garden, implying that mineland had been polluted by Mn and Cd due to exploitation of manganese deposits.

The total concentration of heavy metals can reflect the pollution level of soil environment, but cannot predict its

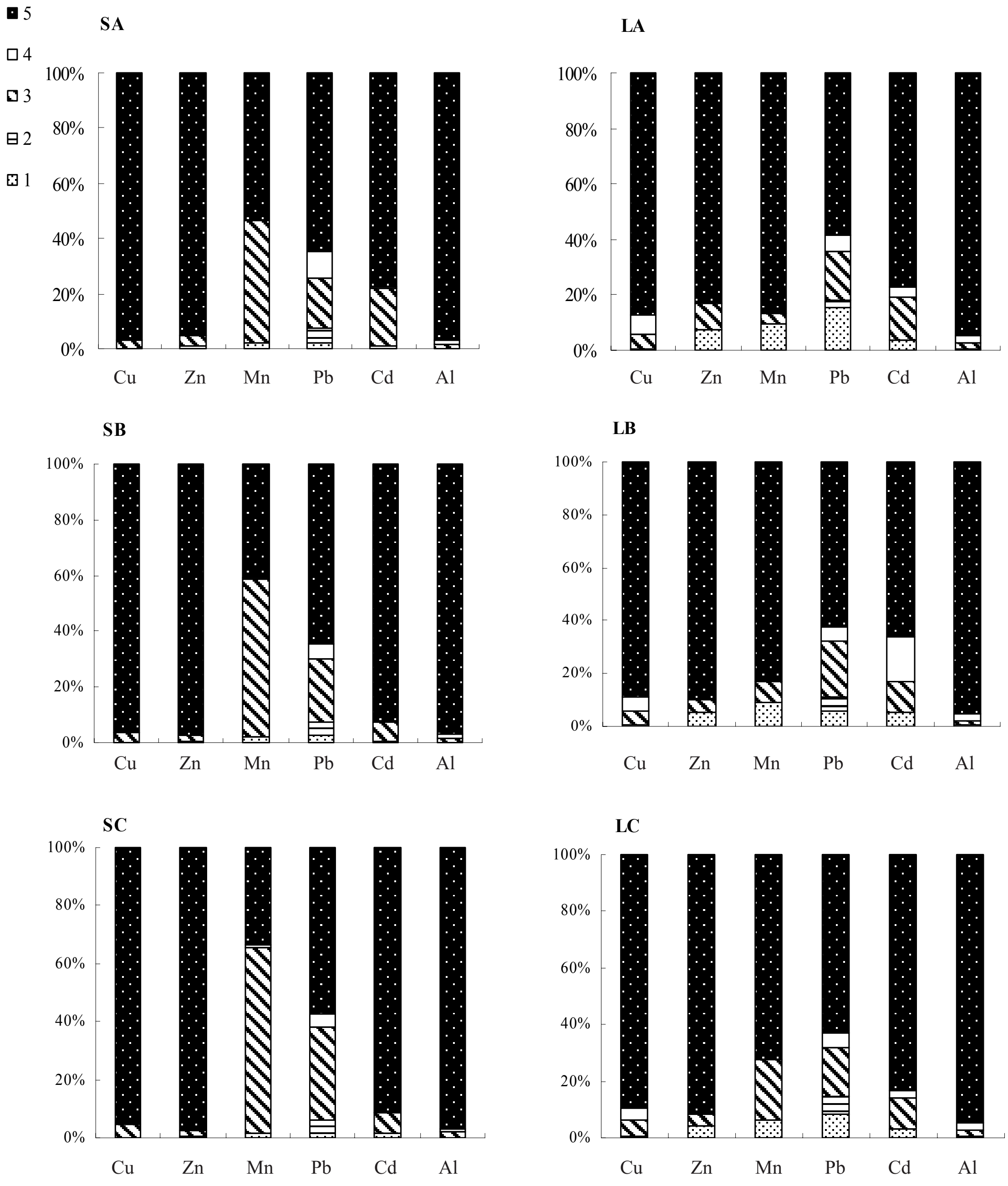

Fig. 1. Speciation analysis of heavy metals in soil.
1 – exchangeable, 2 – bound to carbonate, 3 – bound to Iron and Manganese, 4 – bound to organic matter, 5 – residual.

bioavailability [23]. Hence, the speciation of heavy metals in soils is necessary to be investigated. Section soil samples collected from the two tea gardens contained high concentrations of Cd, but most of the Cd was bound in residual fraction that was unavailable to higher plants according to the results of Tessier's sequential extraction. Similarly, Mn was dominated by iron-manganese and residual fraction in mineland tea garden soils but dominated by residual fraction in Guilin tea garden soils.

Tea plant is a typical kind of Al-rich plant. A number of papers have been published regarding the relationship between growth of tea plants and Al concentration of tea garden soils, and aluminum activation and enrichment processes were reported to exist in tea garden soils [15, 24, 25]. Ding et al. found that Al concentration in soil increased significantly after planting tea [15]. Ruan et al. found that tea planting intended to increase water solubility and exchangeable Al fractions [24]. In this study, Al concentration of the soils in the two tea gardens was respectively up to 15,989.2~20,527.2 times higher than the soil background concentration and showed the pattern of rhizosphere soil>bulk soil outside tea tree rows>bulk soil in-between tea tree rows, which may be due to tea planting. From sequential multiple-step extractions, the amounts of Al fractions were dominated by residue state mixed with small part of Fe-Mn oxide-bond and organic matter-bond.

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