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Environmental Impact Assessment of Tea Garden Soils by the Heavy Metal Concentration in Shandong Province, China

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Scientific basis was presented to fertility management of tea garden soils, and concentrations of selected heavy metals in them were measured, followed by the environmental impact assessment, for keeping quality and sustainable development in the tea production. For this purpose, twelve surface soil samples were collected at different tea gardens in Qingdao and Jimo Cities, Shandong Province, China.

Total concentrations of Cu, Zn, Cr and Pb in the tea garden soils were below the soil environmental standards. The total Cd concentration ranged from 0.32 to 0.81 mg kg^{-1} and exceeded the soil environmental standard. The simple pollution index of 1.90 for Cd confirmed pollution of the tea garden soils by Cd. The overall pollution index was 1.38, which indicated the pollution status of "slightly polluted" in the tea garden soils. Based on the results, soil quality management practices in the tea garden soil were proposed.

INTRODUCTION

Tea cultivation has expanded directly or indirectly from China to about 50 countries in the world (Hu and Li, 2003). At the end of 20th century, tea cultivation area was 260×10^4 ha, and annual tea production and amount of export exceeded 300×10^7 kg and 120×10^7 kg, respectively, in the whole world. Tea cultivation area is 110×10^4 ha in China and equivalent to 42% of the tea cultivation area of the world. Annual tea production in China is 68×10^7 kg, occupying 23% of the world tea production, and annual amount of export in China is 20×10^7 kg, occupying about 17% of the world amount of export (Fuchinokami and Fuchinokami, 1999). In China, people of near one hundred million are working in tea production and associated industries.

Tea production in Shandong Province is limited by the low temperature in the climatic condition, which is a northern limit in the tea production of China. Shandong Province set out a policy for "security of the economic effect due to maintenance of the present tea cultivation area and increase in the yield and production of tea" in 1980s in the 20th century. Tea garden area was 11,400 ha and the gross production reached 4×10^6 kg in 2002, and now tea production is a main industry of agricultural production in Shandong Province. Present major targets in the tea production are reduction in the production cost, increase in the total production, improvement in the tea quality, emphasis on the ecological environmental conservation and improvement of its environment, and promotion of sustainable development.

Types and amounts of chemical substances added to agricultural land are rapidly increasing with development of industrial technology. As a result, soil pollution by heavy metals becomes conspicuous. Therefore, control of tea quality and soil management for keeping it becomes very important.

In the present study, scientific data were presented for fertility management of tea garden soils in Shandong Province, and heavy metal concentrations of the tea garden soils were measured to assess environmental impact for the purposes of maintenance of tea quality and sustainable development of tea production.

MATERIALS AND METHODS

Soil samples

Twelve surface soil (0–20 cm) samples were collected at different tea gardens in Laoshan region of Qingdao City and in Dahainanchun region of Jimo City, Shandong Province, China, on the early October of 2005. Collected soil samples were air–dried in the natural condition and were subjected to measurement of fundamental chemical properties and analysis of heavy metal concentrations.

Chemical analyses

Chemical properties of soils were mainly analyzed according to the soil standard methods of analysis (Committee of Soil Standard Methods for Analyses and Measurements (ed), 1986). The pH was measured in the suspension having a soil:water ratio of 1:2.5. The organic carbon content was determined by the Tyurin method and multiplied by the coefficient of 1.724 to give the organic matter content. The total nitrogen content was determined by the Kjeldahl method. The total phosphorus content was determined by the ascorbic acid method after decomposition with perchloric acid. The soil was digested by the HF–HNO_3–$HClO_4$ acid treatment and analyzed for total potassium by an atomic absorption spectrophotometer.

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Determination of heavy metal concentration

Digestion with the HF–HNO_3–$HClO_4$ acid treatment was used to determine total concentrations of heavy metals (Committee of Soil Standard Methods for Analyses and Measurements (ed), 1986). One g of the pulverized air–dry sample was weighed accurately into a teflon beaker, and 5 mL of the conc. $HClO_4$ solution and 5 mL of the conc. HNO_3 solution were added to the sample. The beaker was covered and heated for 3 h on a hot plate, and then heated without cover until dryness. After cooling and gradual addition of 5 mL of the conc. $HClO_4$ solution and 10 mL of the HF solution in this sequence, the beaker was heated for 15 min. Heating was stopped and then continued until dryness after addition of another 10 mL of the HF solution. The beaker was cooled, added with 5 mL of 6 M HCl and 1 mL of the conc. HNO_3 solution, and heated with cover for 1 h. The beaker was filled–up to two–thirds of the volume with water, covered, and heated to boiling for 2 h. The solution was transferred into a 50–mL volumetric flask and made up to the mark with water after cooling. The solution was quickly transferred into a plastic bottle and analyzed for Cd, Cr, Cu, Pb and Zn by an atomic absorption spectrophotometer.

Ten g of the air–dry sample was weighed accurately into a 100–mL wide–mouth bottle, added with 50.0 mL of 0.1 M HCl, shaken for 1 h with keeping temperature at 30 °C, and centrifuged. The heavy metal concentration in the supernatant was measured by an atomic absorption spectrophotometer to determine the concentration of soluble heavy metals in soil.

Calculation equation of simple and overall pollution indexes

The simple pollution index by a heavy metal was calculated by the equation of (measured heavy metal concentration/soil environmental standard). The overall pollution index by heavy metals was calculated by the equation of (√ (square of the average of the simple pollution indexes＋square of the maximum simple pollution index)/ √ 2).

RESULTS

Tea cultivation

In tea cultivation in Shandong Province, compost of 10,000 kg/ha and chemical fertilizer over 600 kg/ha as N are applied as basal dressing.

Chemical properties of soils in the tea garden

Chemical properties of soils in the tea garden are shown in Table 1. The pH ranged from 4.7 to 5.5, and the soils showed acidity. Especially, the pH of A1 in Laoshan, Qingdao was 4.7 and was lower than the pH of the other samples. Soils in Shandong Province usually show the slightly acid to neutral reaction, and it is mentioned that acidification is advanced in the tea garden soils. Acidification of the tea garden soils seems to be mainly due to application of a large amount of chemical fertilizers.

The organic matter content ranged from 15.6 to 28.3 g kg^{-1}. Compared with 30 g kg^{-1} of the desired value for improvement in the upland field, the organic matter contents of the tea garden soils were low in both regions. The organic matter content ranged from 23.8 to 28.3 g kg^{-1} in the soils of Laoshan, Qingdao, while from 15.6 to 18.7 g kg^{-1} in the soils of Dahainanchun, Jimo; it was higher for the former than for the latter on the regional basis. Completely fermented organic fertilizer imported from Japan has been applied to the tea garden in Laoshan, Qingdao, whereas poultry manure produced on site has been mainly applied to the tea garden in Dahainanchun, Jimo. The difference in the organic matter content between the two regions is considered to be principally ascribed to such difference in plant nutrient management practice.

The total N content ranged from 0.50 to 0.71 g kg^{-1} and was a little higher for the soils in Laoshan, Qingdao (0.58～0.71 g kg^{-1}) than for the soils in Dahainanchun, Jimo (0.50～0.58 g kg^{-1}). The total P_2O_5 content ranged from 1.07 to 2.67 g kg^{-1} in the tea garden soils. It was 2.00 to 2.67 g kg^{-1} for the soils in Laoshan, Qingdao and 1.07 to 1.68 g kg^{-1} for the soils in Dahainanchun, Jimo and was higher for the former by 1.01 g kg^{-1} in average

Table 1. Chemical properties of soils in the tea garden

Region/City	Sample No	Depth (cm)	pH	Organic matter	Total N	Total P_2O_5	Total K_2O
				(g kg^{-1})			
Laoshan/ Qingdao	A1	0–20	4.7	28.1	0.60	2.57	1.76
	A2	0–20	5.3	28.3	0.71	2.00	1.76
	A3	0–20	5.5	26.5	0.60	2.38	1.68
	A4	0–20	5.4	27.2	0.68	2.18	1.73
	A5	0–20	5.2	26.7	0.60	2.67	1.72
	A6	0–20	5.3	23.8	0.58	2.26	1.70
Dahainanchun/ Jimo	B1	0–20	5.4	16.6	0.52	1.42	2.30
	B2	0–20	5.4	18.4	0.58	1.68	2.12
	B3	0–20	5.5	15.6	0.50	1.56	2.23
	B4	0–20	5.3	17.8	0.56	1.12	2.27
	B5	0–20	5.4	18.6	0.57	1.07	2.24
	B6	0–20	5.0	18.7	0.58	1.10	2.23

than for the latter. The total K_2O content ranged from 1.68 to 2.30 g kg^{-1} in the tea garden soils and was higher for the soils in Dahainanchun, Jimo (2.12 to 2.30 g kg^{-1}) than for the soils in Laoshan, Qingdao (1.68 to 1.76 g kg^{-1}), in contrast to the total N and P_2O_5 contents. The regional difference in the chemical composition of the tea garden soils probably reflects the difference in the chemical composition of the organic amendments used in the two regions, as stated in the organic matter content.

Total heavy metal concentrations of soils in the tea garden

Total heavy metal concentrations of soils in the tea garden are shown in Table 2. The total Cu concentration varied from 10.99 to 26.18 mg kg^{-1} in the tea garden soils. It varied from 22.43 to 26.18 mg kg^{-1} in the soils of Laoshan, Qingdao and from10.99 to 20.48 mg kg^{-1} in the soils of Dahainanchun, Jimo and higher for the former by 5.88 mg kg^{-1} in average than for the latter. The total Zn concentration ranged from 13.79 to 28.29 mg kg^{-1} in the tea garden soils, and was higher for the soils in Laoshan, Qingdao (26.10 to 28.29 mg kg^{-1}) by 9.79 mg kg^{-1} in average than for the soils in Dahainanchun, Jimo (13.79 to19.68 mg kg^{-1}). The total Cr concentration was in a range between 13.13 and 26.56 mg kg^{-1} in the tea garden soils. It was in a range between 13.31 and 24.31 mg kg^{-1} in the soils of Laoshan, Qingdao and between 13.13 and 26.56 mg kg^{-1} in the soils of Dahainanchun, Jimo, and there was not a large difference between them. The total Pb concentration ranged from 4.09 to 6.23 mg kg^{-1} in the tea garden soils, and was 4.09 to 6.08 mg kg^{-1} in the soils of Laoshan, Qingdao and 4.25 to 6.23 mg kg^{-1} in the soils of Dahainanchun, Jimo, showing no essential difference between them. The total Cd concentration was in a range of 0.32 to 0.81 mg kg^{-1} in the tea garden soils. It was in a range of 0.54 to 0.81 mg kg^{-1} in the soils of Laoshan, Qingdao and of 0.32 to 0.63 mg kg^{-1} in the soils of Dahainanchun, Jimo and higher for the former by 0.10 mg kg^{-1} in average than for the latter.

Soluble heavy metal concentrations of soils in the tea garden

Soluble heavy metal concentrations of soils in the tea garden are shown in Table 3. The soluble Cu con-

Table 2. Total heavy metal concentrations of soils in the tea garden (mg kg^{-1})

Region/City	Sample No	Cu	Zn	Cr	Pb	Cd
Laoshan/ Qingdao	A1	23.21	27.52	13.31	6.08	0.54
	A2	26.18	28.29	14.78	5.01	0.58
	A3	23.22	26.86	21.68	4.09	0.81
	A4	25.13	27.16	22.67	5.18	0.71
	A5	23.56	26.10	24.31	5.26	0.56
	A6	22.43	27.48	19.47	4.41	0.54
	Average±SD	23.95±0.57	27.23±0.30	19.37±1.82	5.00±0.28	0.62±0.05
Dahainanchun/ Jimo	B1	20.11	15.88	14.27	6.23	0.63
	B2	20.48	13.79	26.51	5.96	0.32
	B3	20.18	17.74	26.56	4.25	0.57
	B4	18.86	19.68	25.83	4.65	0.53
	B5	17.81	18.67	13.13	4.52	0.53
	B6	10.99	18.86	15.41	4.92	0.53
	Average±SD	18.07±1.48	17.44±0.90	20.28±2.71	5.09±0.33	0.52±0.04

Table 3. Soluble heavy metal concentrations of soils in the tea garden (mg kg^{-1})

Region/City	Sample No	Cu	Zn	Cr	Pb	Cd
Laoshan/ Qingdao	A1	0.39 (1.7)[1)]	0.66 (2.4)	tr[2)]	0.29 (4.8)	tr
	A2	0.48 (1.8)	0.87 (3.1)	tr	0.33 (6.6)	tr
	A3	0.39 (1.7)	1.22 (4.5)	tr	0.57 (13.9)	tr
	A4	0.46 (1.8)	0.95 (3.5)	tr	0.36 (6.9)	tr
	A5	0.43 (1.8)	1.29 (4.9)	tr	0.49 (9.3)	tr
	A6	0.32 (1.4)	1.30 (4.7)	tr	0.63 (14.3)	tr
	Average±SD	0.41±0.02	1.05±0.11		0.45±0.06	
Dahainanchun/ Jimo	B1	0.19 (0.9)	0.37 (2.3)	tr	0.78 (12.5)	tr
	B2	0.26 (1.3)	0.35 (2.5)	tr	0.54 (9.1)	tr
	B3	0.30 (1.5)	0.31 (1.7)	tr	0.56 (13.2)	tr
	B4	0.21 (1.1)	0.42 (2.1)	tr	0.59 (12.7)	tr
	B5	0.38 (2.1)	0.47 (2.5)	tr	0.47 (10.4)	tr
	B6	0.28 (2.5)	0.38 (2.0)	tr	0.58 (11.8)	tr
	Average±SD	0.27±0.03	0.38±0.02		0.59±0.04	

[1)] The ratio to the total concentration (%).
[2)] tr: detected in a trace amount.

centration varied from 0.19 to 0.48 mg kg^{-1} in the tea garden soils, and was higher for the soils in Laoshan, Qingdao by 0.14 mg kg^{-1} in average than for the soils in Dahainanchun, Jimo. The ratio of soluble to total Cu concentrations was in a range of 0.9 to 2.5%. The soluble Zn concentration ranged from 0.31 to 1.30 mg kg^{-1} in the tea garden soils, and was higher for the soils in Laoshan, Qingdao by about 3 times than for the soils in Dahainanchun, Jimo. The ratio of soluble to total Zn concentrations was in a range of 1.7 to 4.9%. The soluble Pb concentration ranged from 0.29 to 0.78 mg kg^{-1} in the tea garden soils, and was somewhat lower for the soils in Laoshan, Qingdao than for the soils in Dahainanchun, Jimo. The ratio of soluble to total Pb concentrations was in a range of 4.8 to 14.3%. The soluble Cr and Cd were detected in a trace amount.

DISCUSSION

A tendency of acidification was noticed to the soils in the tea garden in Laoshan region of Qingdao City and in Dahainanchun region of Jimo City in Shandong Province. The acidification of the tea garden soils was ascribed to application of a large amount of chemical fertilizers. A regional difference was recognized to the organic matter content of the tea garden soils, which is probably caused by the difference in plant nutrient management practice, but the content itself was generally low. The total N and K_2O contents in both regions and the total P_2O_5 content in the Dahainanchun region were also found to be low. These rather poor properties of the tea garden soils suggest the necessity of fertility management of tea garden soil based on the soil diagnosis.

T–test was carried out to analyze significance in the difference of total heavy metal concentrations between the Laoshan and Dahainanchun regions. The results are shown in Table 4. As shown in Table 4, a significant difference at the 1% level was observed to the total concentrations of Cu and Zn. T–test was again carried out to analyze significance in the difference of soluble heavy metal concentrations between the two regions. The results are shown in Table 5, and a significant difference at the 1% level was found to the soluble concentrations of Cu and Zn, similar to the results on the total concentration.

As stated already, completely fermented organic fertilizer imported from Japan has been applied to the tea garden in Laoshan region of Qingdao City, whereas poultry manure produced on site has been mainly applied to the tea garden in Dahainanchun region of Jimo City. The completely fermented organic fertilizer imported from Japan is surely higher in the Cu and Zn concentrations, and the difference in the organic amendments used in the two regions is considered to explain well a significant difference observed in the total and soluble concentrations of Cu and Zn between the Laoshan and Dahainanchun regions.

The total concentration (Table 2) was compared with its soil environmental standard (Table 6) in different heavy metals. Total concentrations of Cu, Zn, Cr and Pb were within the soil environmental standards in the second standard, and the tea garden soils were judged to be not polluted by these heavy metals. The ratio of the total concentration to the soil environmental standard was 5～13, 6～11, 7～13 and 3～4% for Cu, Zn, Cr and Pb, respectively. In contrast to them, the total Cd concentration exceeded the soil environmental standard, and pollution of the tea garden soils by Cd was recognized. The ratio of the total concentration to the soil environmental standard was in a range of 106 to 270%, and the maximum total Cd concentration amounted to 2.7 times of the soil environmental standard. However, the soluble Cd concentration was below the detection level, and little possibility of pollution of tea plants by Cd is expected.

The simple and overall pollution indexes by the total heavy metal concentrations were evaluated for the environmental impact assessment of the soils in the tea garden. The simple pollution index is given in Table 6. The simple pollution index was low to Cu, Zn, Cr and Pb, and was 0.14, 0.11, 0.13 and 0.02, respectively. The simple pollution index for Cd was 1.90, and pollution of the tea garden soils by Cd was clearly indicated. The

Table 4. T–test on significance in the difference of total heavy metal concnetrations between the two regions

Heavy metal	t	$t_{0.01,10}$	Significance
Cu	3.72	3.17	**
Zn	10.32	3.17	**
Cr	0.28	3.17	(o)
Pb	0.18	3.17	(o)
Cd	1.59	3.17	(o)

*: significance at the 1% level.
(o): no significance.

Table 5. T–test on significance in the difference of soluble heavy metal concnetrations between the two regions

Heavy metal	t	$t_{0.01,10}$	Significance
Cu	3.83	3.17	**
Zn	6.16	3.17	**
Pb	1.97	3.17	(o)

*: significance at the 1% level.
(o): no significance.

Table 6. The simple pollution index of the tea garden soils by the total heavy metal concentration for different heavy metals

Heavy metal	Average±SD (mg kg^{-1})	Soil environmental standard (mg kg^{-1})	Simple pollution index
Cu	21.01±1.16	150	0.14
Zn	22.34±1.54	200	0.11
Cr	19.83±1.56	150	0.13
Pb	5.05±0.21	250	0.02
Cd	0.57±0.03	0.30	1.90

Table 7. Classifiication of the soil pollution level by heavy metals based on the overall pollution index

Pollution level	Overall pollution index (P)	Status of pollution	Effect on soil and crops
1	$P \leqq 0.7$	Hardly polluted	Nothing
2	$0.7 < P \leqq 1.0$	Near the pollution level	Scarcely affected
3	$1.0 < P \leqq 2.0$	Slightly polluted	Some crops start to be suffered from pollution
4	$2.0 < P \leqq 3.0$	Fairly polluted	Soil and crops are suffered from the middle degree of pollution
5	$P > 3.0$	Severely polluted	Soil and crops are suffered from severe pollution

overall pollution index for the tea garden soils examined was calculated to be 1.38, and they were designated as the pollution level of "3" or "slightly polluted" according to the classification of the soil pollution level based on the overall pollution index (Table 7). This pollution level of the tea garden soils was in a similar level to the orchard soils (Han *et al.*, 2007a) but was low compared with the soils in the vegetable field (Han *et al.*, 2007b) in Shandong Province. Contribution of Cd is great to the overall pollution index of 1.38. However, absorption of Cd by tea plants would hardly occur, because the soluble Cd concentration was detected in a trace amount.

Based on the above discussion, it is mentioned that status of soil pollution is more worsened without proper and scientific field management practice. It will adversely affect quality of tea and retard the sustainable tea production. Since all tea garden soils examined exceeded the soil environmental standard, it is necessary and important to take measures for remediation of Cd pollution on the practice of field management in the tea garden. In addition, it is desirable to improve the fertilizer use efficiency, to reduce the amount of fertilizer applied and to use organic fertilizer of the environmental preservation type, on the fertility management in the tea garden. Use of biological control agent is recommended, which exerts little pollution to the environment, when pesticide is applied.

CONCLUSIONS AND PROPOSAL

The total Cd concentration exceeded the soil environmental standard in the soils of the tea garden in Shandong Province, China, and its simple pollution index was highest among the five heavy metals of Cu, Zn, Cr, Pb and Cd examined. The total Cu and Zn concentrations were affected by the type and amount of organic amendments applied to the tea gardens, and they showed a cumulative tendency by consecutive application of organic amendments. Since quality of tea is greatly influenced by the Cd concentration, it has a high possibility to become a limiting factor of sustainable tea production. Although it is supposed that there is little risk of pollution of tea plants by Cd in the present, field management practice based on the scientific data is proposed to keep sustainable tea production in Shandong Province.

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