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Survey of Heavy Metal Pollution in Four Chinese Crude Drugs and Their Cultivated Soils

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Abstract A two-year survey on the residues of heavy metals in four Chinese crude drugs and their cultivated soils was conducted. Targeted heavy metals were copper (Cu), arsenic (As), lead (Pb), nickel (Ni), and cadmium (Cd). Herbs surveyed include White Peony Root (Radix Paeoniae Alba), Turmeric Root Tuber (Radix Curcumae), Thunberg Fritillary Bulb (Bulbus Fritillariae Thumbergii), and Tuber of Dwarf Lilyturf (Radix Ophiopogonis). Concentrations of all heavy metals were under the permitted levels except cadmium, which exceeded the permitted level in some samples of Thunberg Fritillary Bulb, White Peony Root, and Turmeric Root Tuber. Concentration coefficients were less than 1.0 for all heavy metals except cadmium. The concentration coefficient of cadmium in Turmeric Root Tuber was 14.0. Lower pH and high Zn concentration in the soil may facilitate the transfer of cadmium from cultivated soil into the herbs.

Keywords Heavy metal · Chinese crude drug · Medicinal herb · Contamination

Herbal medicines have been used in medical practice for thousands of years and recognized as a valuable and readily available resource. A World Health Organization report indicated that about 70–80% of the world's population relies on non-conventional medicine, mainly of herbal sources, for their primary healthcare (Akerele 1993). Traditionally, herbs and herbal products have been considered to be mild, non-toxic and even harmless due to their natural origin. The market value of herbal medicine products in year 2004 had reached 10 billion US dollars in China (Zhang 2005, http://www.enorth.com.cn). The current international market value of herbal medicines is around 30 billion US dollars. However, China only shared about 5.0% of this international market. Although the herbal medicine industry has been developing rapidly in recent decades in China, unstable quality and contamination of heavy metals and pesticide residues have significantly impacted its competitiveness and export market shares. With the ever-increasing market demand, more medicinal herbs are cultivated in agricultural fields and agricultural practices will inevitably affect the quality of the medicinal herbs.

Paeonia lactiflora Pall., *Curcuma wenyujin* Y.H.Chen et C.Ling, *Fritillaria thunbergii* Miq. and *Ophiopogon japonicus* (Thunb.) Ker-Gawl. are perennial medicinal herbs. Their roots, White Peony Root (Radix Paeoniae Alba), Turmeric Root Tuber (Radix Curcumae), Thunberg Fritillary Bulb (Bulbus Fritillariae Thumbergii) and Tuber of Dwarf Lilyturf (Radix Ophiopogonis) are important varieties of geo-authentic crude medicines in Zhejiang Province of China. Since the roots have to grow in the soil for several years before the harvest, it is always a concern that they may be contaminated by heavy metals in the soil.

The objective of this study was to investigate the concentrations of heavy metals in four medicinal herbs with significant market value in Zhejiang province. An attempt was also made to understand the accumulating behavior of heavy metals in those medicinal herbs by simultaneously analyzing heavy metals in their cultivated soils. The results

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of this study will be useful in assessing the safety and quality of those medicinal herbs.

Materials and Methods

All herb and soil samples were collected on family farms in a Chinese medicinal herb mass producing area in Zhejiang Province of China from 2004 to 2005. Samples of White Peony Root were collected from ten farms in six villages in Dongyang City, ten farms in three villages in Panan County and five farms in a village in Jinyun County. Samples of Turmeric Root Tuber were collected from five farms in Ruian City and samples of Thunberg Fritillary Bulb from ten farms in five villages in Ningbo City, five farms in Panan County and five farms in Yongkang City. Samples of Dwarf Lilyturf were collected from ten farms in a village in Cixi City. Herb and soil were randomly sampled from the planting fields. Soil was sampled in a depth of approximately 0–15 cm. The herb and soil samples were dried at room temperature, ground, passed through 20-mesh sieve and then stored in polyethylene bottles before analysis.

All reagents were analytical reagent grade. Sample extraction followed the Germany standard method. Detecting light source: ICAP; ICAP view: line selection; analysis maximum integration times (s) low WL range: axial 20, radial 20, high WL range: axial 15, radial 10; Nebulizer pump flush pump rate (rpm): 130, 2.4 mL/min; analysis pump rate (rpm): 130, 240 mL/min; pump relaxation time (s) 0; pump tubing type: Tygon-Orange; RF power: 1,150 W; Nebulizer flow: 0.7 lpm; auxiliary gas: 0.5 lpm.

Results and Discussion

Residues of most of the heavy metals in the four medicinal herbs were under the China's permitted levels (Ministry of Foreign Trade and Economic Cooperation, PRC 2005) (Table 1). Cadmium residues, however, were 0.341 mg/kg in Thunberg Fritillary Bulb from Ningbo, 0.417 mg/kg in Thunberg Fritillary Bulb from Panan, and 0.482 mg/kg in White Peony Root from Jinyun, and 1.723 mg/kg in Turmeric Root Tuber from Ruian (Table 1). They were exceeded the permitted level (0.3 mg/kg). There were different concentration coefficients of heavy metals in the four medicinal herbs (Table 2). The concentration coefficients for all heavy metals were less than 1.0, except cadmium. Cadmium was a main factor that affected the quality and safety of Thunberg Fritillary Bulb, White Peony Root and Turmeric Root Tuber. The concentration coefficient of cadmium in Turmeric Root Tuber was 14.0,

Table 1 Residues of heavy metal in four kinds of geo-authentic crude drugs and soils of Zhejiang Province (mg/kg)

	Sample name	pH	Cu	As	Pb	Ni	Cd	Zn	Mn
Chinese crude drugs	Thunberg Fritillary Bulb (Ningbo)		2.88 ± 0.36	0.35 ± 0.17	1.36 ± 0.60	0.78 ± 0.17	0.341 ± 0.094	41.00 ± 4.67	18.34 ± 2.47
	Thunberg Fritillary Bulb (Panan)		2.37 ± 0.81	0.32 ± 0.08	0.56 ± 0.13	0.81 ± 0.04	0.417 ± 0.121	37.55 ± 3.39	17.68 ± 1.58
	Dwarf Lilyturf (Cixi)		6.93 ± 1.08	0.13 ± 0.09	0.35 ± 0.12	1.23 ± 0.74	0.110 ± 0.013	9.47 ± 1.05	7.06 ± 0.38
	White Peony Root (Jinyun)		2.77 ± 0.71	0.27 ± 0.09	0.52 ± 0.13	1.28 ± 0.09	0.482 ± 0.078	48.64 ± 11.39	74.10 ± 73.52
	White Peony Root (Panan)		2.28 ± 0.32	0.25 ± 0.09	0.57 ± 0.08	2.06 ± 0.52	0.291 ± 0.168	38.23 ± 1.40	38.08 ± 8.93
	Turmeric Root Tuber (Ruian)		7.77 ± 0.61	0.41 ± 0.08	4.96 ± 1.77	2.89 ± 0.24	1.723 ± 0.790	130.72 ± 38.19	644.90 ± 249.15
Soils	Thunberg Fritillary Bulb (Ningbo)	5.57 ± 0.49	36.87 ± 1.68	6.56 ± 3.03	30.93 ± 7.09	9.22 ± 0.72	0.424 ± 0.203	106.20 ± 18.44	431.70 ± 187.90
	Thunberg Fritillary Bulb (Panan)	4.64 ± 0.85	7.49 ± 1.28	3.68 ± 0.28	24.40 ± 0.10	3.60 ± 0.50	0.099 ± 0.013	50.73 ± 3.63	506.90 ± 30.30
	Dwarf Lilyturf (Cixi)	7.33 ± 0.03	18.63 ± 2.64	4.55 ± 0.16	17.63 ± 0.38	18.65 ± 0.43	0.174 ± 0.013	55.82 ± 3.76	347.70 ± 7.30
	White Peony Root (Jinyun)	4.04 ± 0.36	10.44 ± 1.36	4.85 ± 0.25	35.47 ± 2.50	3.66 ± 0.60	0.183 ± 0.059	53.66 ± 4.84	495.80 ± 82.40
	White Peony Root (Panan)	4.05 ± 0.53	14.68 ± 6.88	3.66 ± 0.41	34.00 ± 1.87	7.36 ± 2.81	0.148 ± 0.022	68.86 ± 16.99	338.20 ± 98.80
	Turmeric Root Tuber (Ruian)	4.95 ± 0.89	10.65 ± 1.23	3.96 ± 0.28	25.63 ± 0.90	12.96 ± 1.38	0.123 ± 0.008	60.31 ± 2.67	470.30 ± 58.40

Table 2 Concentration coefficients of heavy metals in geo-authentic crude drugs

Sample name	Cu	As	Pb	Ni	Cd	Zn	Mn
Thunberg Fritillary Bulb (Ningbo)	0.08	0.05	0.04	0.08	0.80	0.39	0.04
Thunberg Fritillary Bulb (Panan)	0.32	0.09	0.02	0.23	4.21	0.74	0.03
Dwarf Lilyturf (Cixi)	0.37	0.03	0.02	0.07	0.66	0.17	0.02
White Peony Root (Jinyun)	0.27	0.06	0.01	0.35	2.63	0.91	0.15
White Peony Root (Panan)	0.16	0.07	0.02	0.28	1.97	0.56	0.11
Turmeric Root Tuber (Ruian)	0.73	0.10	0.19	0.22	14.0	2.17	1.37

the highest among all heavy metal and herb combinations. This may explain why the residue of cadmium in Turmeric Root Tuber reached to 1.723 mg/kg, greatly exceeding the permitted level (0.3 mg/kg), although its concentration in the soil was only 0.123 mg/kg. Sauve et al. (1998) argued that the toxicological significance of the predicted free metal activities was comparable to that of the total metal contents. In most cases, estimated free metal activity improves the prediction of toxic effects on crops, soil organisms, or soil microbial processes. There were many factors influencing free metal activities in soil, such as pH, organic carbon content, Zn element, etc. (Singh et al. 1998; Hua et al. 2002a, b; Hart et al. 2002). The concentrations of cadmium in Thunberg Fritillary Bulb of Ningbo (0.341 mg/kg) and Panan (0.417 mg/kg) were similar, although there was a significant difference in cadmium concentration in their cultivated soils (0.424 mg/kg for Ningbo samples and 0.099 mg/kg for Panan samples). It indicated that heavy metal residues in Thunberg Fritillary Bulb were not closely related to those in its cultivated soils. However, low pH and Zn concentration in the soil seems to facilitate the transfer of cadmium from soil into the herbs by comparing the pH and Zn concentrations between these two soil samples. This result was agreed with Singh and Hua's experimental results (Singh et al. 1998; Hua et al. 2002).

This study provided evidence that most heavy metal residues in the four studied Chinese medicinal herbs were under the permitted levels. However, cadmium in Thunberg Fritillary Bulb, White Peony Root and Turmeric Root Tuber exceeded the permitted level (0.3 mg/kg). The cadmium concentrations in the herbs were related to pH values and Zn element contents in the soils. Low pH and Zn concentration in the soil will enhance the transfer of cadmium from soil to herbs. Turmeric Root Tuber has the highest concentration coefficient among the four Chinese medicinal herbs, which may also facilitate the accumulation of cadmium.

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