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Arsenic, Cadmium, Chromium, Cobalt, and Copper in Different Types of Chinese Tea

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Tea was originated from China and Chinese tea plays a vital role in world's production and market. The total production and exportation in 2002 were 745,374 and 252,273 tonnes, respectively (International Tea Committee, 2003). Tea is one of the most popular and effective healthy drinks in the world. Many scientific researches have concluded that tea is beneficial in against cardiovascular disease, cancer, immune-system diseases, degenerative diseases and ageing (Modder et al. 2002). However, some undesirable elements, such as Al, F and Pb, are a concern of tea consumers. Mature tea leaves may contain up to 2-3% Al (Coriat et al. 1986; Matsumoto et al. 1976) and 0.28% F (Fung et al. 1999), and Al and F concentrations are significantly higher in brick tea than any other teas (Wong et al. 2003; Shu et al. 2003). It is understandable since tea (*Camellia sinensis*) has the nature to accumulate Al and F. Rapid industrialization and urbanization in China over the last two decades has resulted in heavy metal burden to the environment. There are an increasing number of cases of tea products exceeding the maximum permissible concentration for Pb of 2 mg kg^{-1} dry weight (Song et al. 2000; Zhang et al. 2000; Lin et al. 2002). However, there are few reports on the As, Cd, Co, Cr and Cu concentrations in tea. As, Cd and Cr are toxic to humans. Co and Cu are essential, while their ingestion at excessive levels have the toxicological effects on the body. Therefore, it is important to investigate the concentrations of these heavy metals in tea.

In this study we conducted a comprehensive survey on the As, Cd, Co, Cr and Cu concentrations in different types of Chinese made tea. The purpose is to understand the magnitude of these heavy metals contamination and possible reasons.

MATERIALS AND METHODS

Total 811 tea samples were collected from 17 tea producing provinces in China during 1999-2001. Samples were collected in two ways: 1) representative tea producers including tea farms and tea processing companies were pre-selected and requested to send their samples for analysis; and 2) tea producers sent in their samples for quality check prior to sale or export. The samples included three main types of tea produced in China: green, black and oolong teas. These types of tea

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differ in the varieties of tea plants and the way tea leaves are plucked and processed.

After collection, tea samples were dried again in 105℃ for 2 h in an oven and ground into a powder with a stainless steel grinder. Tea samples were digested in a microwave digestion oven (CEM Mars5, CEM Corp, Matthews, NC). Approximately 0.5 g of each sample was weighed into a Tefon coated tube and 5 ml of concentrated HNO_3 (high purity grade) was added. The samples were allowed to predigest for 15 min at room temperature and then transferred to the microwave oven. Digestion was carried out at 170℃ for 10 min at a maximal pressure of 800 psi. Samples were cooled to room temperature and diluted to 25 ml with high quality deionized water (>18 MΩ specific resistance). As, Cd, Co, Cr and Cu concentrations were determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES) (TJA, IRIS/AP, Thermo Jarrell Ash Corporation, Franklin, USA). The detection limit for As, Cd, Co, Cr and Cu were 0.07, 0.0005, 0.001, 0.001 and 0.0013 mg l^{-1}, respectively. Blanks and a reference tea material (GB07605-GSV-4, Institute of Geophysical and Geochemical Sciences, Chinese Academy of Geological Sciences, Langfang, Hubei, China) were included for quality control. The recovery rate for externally added As, Cd, Co, Cr and Cu were 96.5, 92.8, 100.8, 97.8 and 100.7%, respectively (Table 1). The result of the reference tea material analyzed by this method coincided with the certified concentration (Table 2). Fifteen percent of samples were replicated during determination.

Table 1. Recoveries of different elements determined.

Element	Concentration of tea sample (mg kg^{-1})	Concentration of element added (mg l^{-1})	Determined concentration (mg l^{-1})	Recovery (%)
As	0.13	2.0	2.06	96.5
Cd	0.03	2.0	1.88	92.8
Co	0.20	2.0	2.22	100.8
Cr	0.16	10.0	9.94	97.8
Cu	6.36	10.0	16.43	100.7

Table 2. Result of reference tea determined by ICP-AES method.

Element	Concentration of reference tea (mg kg^{-1})	Determined value (mg kg^{-1})	RSD (n=5) (%)
As	0.28±0.03	0.28	5.11
Cd	0.057±0.008	0.056	7.40
Co	0.18±0.02	0.20	0.73
Cr	0.80±0.02	0.84	5.31
Cu	17.3±1.0	17.7	0.72

Data are expressed on a dry weight (DW) basis. Statistical analyses of mean, median, 95^{th} % value and one-way ANOVA were performed using SPSS 10.0 for Windows.

RESULTS AND DISCUSSION

Arsenic concentration of total 800 tea samples ranged from below the detection limit to 4.43 mg kg^{-1} DW, with a mean and median value of 0.26 and 0.00 mg kg^{-1} DW, respectively (Table 3). Among the varied types of tea, black tea was the highest and statistically significantly ($p<0.001$) different with other types of tea. Next is other tea, which including brick tea, Pu-er tea, white tea and mixed tea with other materials, such as roast rice, ginseng, and rose flower. Green tea was the lowest and the difference was significant ($p<0.001$) compared with black and other teas except for the oolong tea.

Table 3. Arsenic concentrations in different types of Chinese tea.

Types of tea	No. of samples	Range (mg kg^{-1})	Mean* (mg kg^{-1})	Median (mg kg^{-1})	95th % value (mg kg^{-1})
Green tea	536	Tr.-1.66	0.14 a	0.00	0.78
Black tea	148	Tr.-4.43	0.69 b	0.63	1.85
Oolong tea	81	Tr.-1.72	0.20 a	0.00	1.27
Other tea	35	Tr.-1.68	0.37 c	0.29	1.45
Total	800	Tr.-4.43	0.26	0.00	1.09

* The different letters in mean column implicate significant difference ($p<0.05$).

Cadmium concentration in total 798 tea samples collected in main tea producing provinces in China ranged from below the detection limit to 1.07 mg kg^{-1} DW, with a mean and median value of 0.02 mg kg^{-1} DW and under the detection limit, respectively (Table 4). Among the different types of tea, green tea was the highest. Next was oolong tea. Black and other tea was quite lower compared to green and oolong teas. There were significantly different ($p<0.05$) between green tea and black tea, black tea and oolong tea, oolong tea and other tea.

Table 4. Cadmium concentrations in different types of Chinese tea.

Types of tea	No. of samples	Range (mg kg^{-1})	Mean* (mg kg^{-1})	Median (mg kg^{-1})	95th % value (mg kg^{-1})
Green tea	535	Tr.-1.07	0.24 ab	0	0.13
Black tea	147	Tr.-0.09	0.01 c	0	0.04
Oolong tea	81	Tr.-0.23	0.03 a	0	0.16
Other tea	35	Tr.-0.16	0.01 bc	0	0.07
Total	798	Tr.-1.07	0.02	0	0.11

* The different letters in mean column implicate significant difference ($p<0.05$).

Cobalt concentration in total 801 tea samples ranged from below the detection limit to 3.56 mg kg^{-1} DW, with a mean and median value of 0.37 and 0.32 mg kg^{-1} DW (Table 5). Among the different types of tea, green tea was the highest. Other tea was the lowest, only accounting for approximate half Co concentration of green tea. There were significantly different ($p<0.05$) between green tea and oolong or other teas, between black tea and other tea.

Chromium concentration in total 801 tea samples collected from the main tea producing provinces in China ranged from below the detection limit to 16.1 mg kg^{-1} DW, with a mean and median value of 0.91 and 0.54 mg kg^{-1} DW, respectively (Table 6). The black tea was significantly higher (p<0.001), almost 2-4 fold that of other types of tea. Oolong tea was the lowest one. Green tea and other tea were similar in both mean and median values of Cr concentration.

Table 5. Cobalt concentrations in different types of Chinese tea.

Types of tea	No. of samples	Range (mg kg^{-1})	Mean* (mg kg^{-1})	Median (mg kg^{-1})	95^{th} % value (mg kg^{-1})
Green tea	537	Tr.-3.56	0.40 a	0.33	0.94
Black tea	148	Tr.-1.24	0.35 ab	0.35	0.64
Oolong tea	81	Tr.-2.77	0.28 bc	0.21	0.80
Other tea	35	Tr.-0.55	0.21 c	0.24	0.53
Total	801	Tr.-3.56	0.37	0.32	0.86

* The different letters in mean column implicate significant difference (p<0.05).

Table 6. Chromium concentrations in different types of Chinese tea.

Types of tea	No. of samples	Range (mg kg^{-1})	Mean* (mg kg^{-1})	Median (mg kg^{-1})	95^{th} % value (mg kg^{-1})
Green tea	537	Tr.-16.10	0.70 a	0.48	1.68
Black tea	148	Tr.-5.43	1.92 b	1.73	4.47
Oolong tea	81	Tr.-5.16	0.41 c	0.27	1.46
Other tea	35	Tr.-7.26	0.99 a	0.48	3.71
Total	801	Tr.-16.1	0.91	0.54	3.29

* The different letters in mean column implicate significant difference (p<0.05).

Copper concentration in total 811 tea samples ranged from 2.04 to 447.50 mg kg^{-1} DW, with a mean and median value of 18.33 and 15.24 mg kg^{-1} DW, respectively (Table 7). Similar to Cr concentration, the black tea was significantly higher (p<0.001), and almost 1-2 fold higher than that of other types of tea. The possible reason was that black tea were mainly produced in southern part of China, in particular in Guangdong, Yunnan, Guangxi and Hainan provinces, where Cu-bearing bordeaux mixture was extensively used to deal with disease problem. Oolong tea was the lowest one among the 4 types of tea. Green tea and other tea were no much different. This was due to different plucking standard for varied types of tea. The raw material of oolong tea was much older than that of other types of tea. Normally three to four leaves with a dormant bud were plucked for oolong tea and two to three leaves with an active bud for others. The older the tea leaf, the lower Cu concentration is.

The concentrations of these heavy metals in Chinese tea were compared with that of tea produced in Japan, Sri Lanka, India and Nigeria (Tsushida et al. 1977; Ramakrishna et al. 1987; Natesan et al. 1990; Onianwa et al. 1999). Cd concentration of Chinese tea was similar to that of Japanese green tea, but quite lower compared with that of Indian, Sri Lankan and Nigerian teas. Co concentration was lower than that of Indian and Nigerian teas. Cr content was similar to that of Nigerian tea, but generally lower compared with Indian tea. Cu concentration was a little higher than that of Japanese, Nigerian and Sri Lankan teas, but lower compared with that of Indian tea. From above comparison we could conclude that Chinese tea was generally not contaminated in terms of these heavy metals.

Table 7. Copper concentrations in different types of Chinese tea.

Types of tea	No. of samples	Range (mg kg^{-1})	Mean* (mg kg^{-1})	Median (mg kg^{-1})	95th % value (mg kg^{-1})
Green tea	547	2.06-239.02	15.68 a	14.95	22.34
Black tea	148	2.04-447.50	32.82 b	28.14	48.67
Oolong tea	81	3.44-29.82	11.28 c	10.41	23.18
Other tea	35	4.49-38.07	14.68 a	12.87	29.24
Total	811	2.04-447.5	18.33	15.24	36.61

* The different letters in mean column implicate significant difference (p<0.05).

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