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Lead and Copper Levels in Tea Samples Marketed in Beijing, China

F. Qin · W. Chen

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Next to water, tea is the most widely consumed beverage (Marcos et al., 1998) because of its taste aroma and health benefits. On the basis of extensive animal experiments and some epidemiologic data, tea is considered to have numerous beneficial effects on the prevention of many diseases, including skin cancer (Katharine, 2001), Parkinson's disease (Richard, 2001), myocardial infarction (Cheng, 2003), and coronary artery disease (Hirano et al., 2003).

Tea is produced from the leaves of the tropical evergreen *Camellia sinensis*, which is indigenous to China. In China, tea sipping is an integral part of the culture. In the world, China is a major tea producer, supplying 23.3% of the world's tea. However, the rapid industrialization of China over the past two decades has caused increased contamination to the environment, which inevitably has brought contamination to tea.

During the growth period of the tea plant and during tea processing, tea itself can be contaminated by heavy metals, such as lead and copper, which might increase the metal body burden in humans. Lead is a physiologic and neurologic toxin that can affect almost every organ and system in the human body. It can reduce cognitive development and intellectual performance in children and damage kidneys and the reproductive system. Copper is an essential element, with both deficiencies and excesses associated with impaired health. Copper deficiency is known to cause various physiologic disorders such as anemia and bone abnormalities (Uauy et al., 1998) resulting from decreased activity of the copper-requiring enzymes. Copper excess can cause hepatic and kidney damage, hemolytic anemia (Evans, 1973), and methaemoglobinemia (Chugh et al., 1975). Therefore, after the pesticide contamination issue, heavy metal contamination in teas has become another noteworthy issue because it is related directly to heath and disease.

The current study aimed to determine the lead and copper concentrations in tea samples collected from the Beijing market, and to give an overview of the current safety situation of teas marketed in Beijing, host of the Olympic Games in the year 2008.

Materials and Methods

For this study, 57 commercial tea samples were randomly purchased at the local market in Beijing, China. The samples, produced by different provinces of China, included four main types of tea: green, black, Oolong, and scented teas. For each sample, about 1.5 g of ground tea sample was accurately weighed and digested with 10 ml of $HNO_3/HClO_4$ (1 + 4) mixture at 130° until the solution became transparent. Then the residue was filtered through a Whatman No.42 filter paper to remove any turbidity or suspended matter. The filtrate was diluted to 10 ml with 2% HNO_3, and the solutions were subsequently analyzed for lead and copper using GFAAS (PerkinElmer Annalyst 700, Inc., Shelton, USA) with the standard calibration technique. All measurements were performed at 283.3 nm (slit 0.7 nm) for lead and at 324.8 nm (slit 0.7 nm) for copper using PerkinElmer hollow cathode lamps (). The heating programs used for lead and copper determination are given in Table 1. Argon 99.96% was used as the protective gas

F. Qin (✉) · W. Chen
Beijing Key Laboratory of Bioactive Substances and Functional Foods, Beijing Union University, 197 Beituchengxi Street, Beijing 100083, China
e-mail: qinfeilll@hotmail.com

Table 1 Heating programs for lead and copper determinations in tea samples

Step	Temperature (°C)	Ramp/s	Hold/s	Ar flow rate (mL/min)
1	100	5	20	250
2	140	15	15	250
3	700,[a] 1,000[b]	10	20	250
4	1,800,[a] 2,300[b]	0	5	0
5	2,600	1	3	250

[a] For copper

[b] For lead

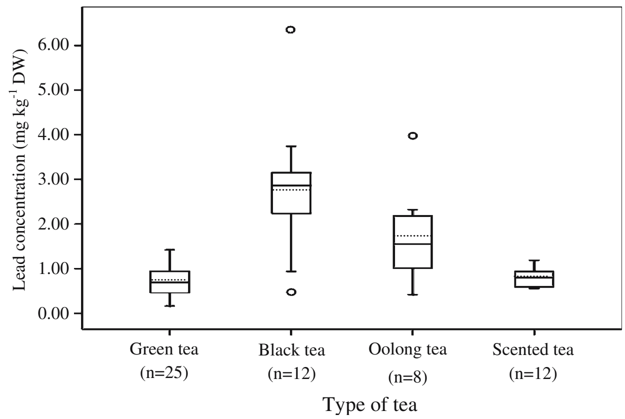

Fig. 1 Box plots of lead concentration in different tea types among the studied tea samples. The central solid line within each box is the median, and the bottom and top of each box represent the 25th and 75th percentiles, respectively. The dotted line is the mean. The whiskers represent the 10th and 90th percentiles, respectively, and the values outside this range are plotted as individual outliers (O)

throughout. The digestion of all tea samples was triplicated, and only the means were reported afterward.

All the reagents were of guaranteed reagent grade. Blanks and a certified reference material for tea (GBW 07605; Institute of Geophysical and Geochemical Exploration, Langfang, China) with certified lead (4.4 mg/kg) and copper (17 mg/kg) concentrations were included for quality control. Repeated analysis of the reference material gave a mean lead concentration of 4.4 ± 0.05 mg/kg (n = 6) and a mean copper concentration of 17 ± 0.13 mg/kg (n = 6). A recovery test of the total analytical procedure was performed for selected samples by spiking analyzed samples with metal standards according to the original concentrations of the spiked samples. Acceptable recoveries of 96.2% to 103.5% and 97.6% to 106.4% were obtained for externally added lead and copper, respectively.

Data are expressed on a dry weight basis. Statistical analysis of variance (ANOVA) was performed using SPSS (Inc., Chicago, USA) 14.0 for Windows.

Results and Discussion

The concentrations of lead in the 57 tea samples marketed in Beijing (China) varied from 0.198 to 6.345 mg/kg dry weight (median, 0.879 mg/kg; mean, 1.320 mg/kg). The highest level of lead was found in Qimen Black tea (Anhui Province), and the lowest level was found in Longjing green tea (Zhejiang Province). A previous report showed that the lead concentrations in 85 Chinese teas varied from 0.15 to 11.61 mg/kg dry weight (Tang et al., 2003), which compared well with our results. The average amount of lead in the all samples was below the maximum permissible concentration (MPC) of 5 mg/kg dry weight (Chinese Ministry of Health, 2005). Only one tea sample exceeded the MPC, accounting for 1.75% of the total analyzed samples. In the survey of lead concentration in 1,225 Chinese teas collected from 1999 to 2001, Han et al. (2006b) found that 32% of the samples exceeded the MPC. However, the authors used the MPC of 2 mg/kg dry weight (Chinese Ministry of Health, 1988). According to this MPC, 13 of all the investigated samples in our study exceeded this permissible level, accounting for 22.8% of the total analyzed samples. Thus our results were comparable.

The main sources of lead in tea samples are their growth media, such as soils. Lead contamination in the soil usually can be attributed to the following three main processes: industrial activities such as mining and smelting processes, agricultural activities such as application of insecticide and municipal sewage sludges, and urban activities such as combustion of gasoline. Tea plants normally are grown in highly acidic soils, where lead is potentially more bioavailable for root uptake (Han et al., 2006b). Deposits from the polluted air onto the leaves of the tea plant can be another source of lead contamination of tea. Tea plants have a large leaf area, which is conducive to foliar deposition or uptake of lead from the atmosphere. The levels of lead in the majority of the tea samples analyzed were higher than those reported worldwide for many other food categories, such as mushroom (Demirbaş, 2000), honey (Muñoz and Palmero, 2006), and vegetables (Bahemuka and Mubofum, 1999).

Figure 1 shows the box plots for lead concentrations in different tea types. An ANOVA of log-transformed data showed significant differences (p < 0.05) in the lead concentrations between different tea types. As can be seen from Fig. 1, green and scented teas had lower median and mean lead concentrations than black and Oolong teas. The higher exceedance of lead concentrations in black and Oolong teas than in green and scented teas could be explained by the different types of leaves picked for different teas. Usually more and older leaves are harvested for the

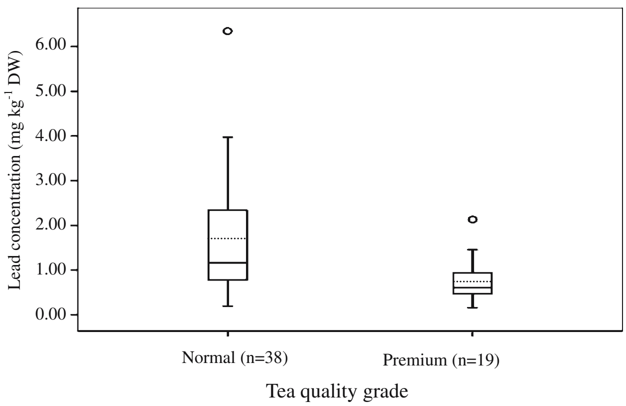

Fig. 2 Box plots of lead concentration in normal and premium grade teas. See the Fig. 1 legend for an explanation of the box plot

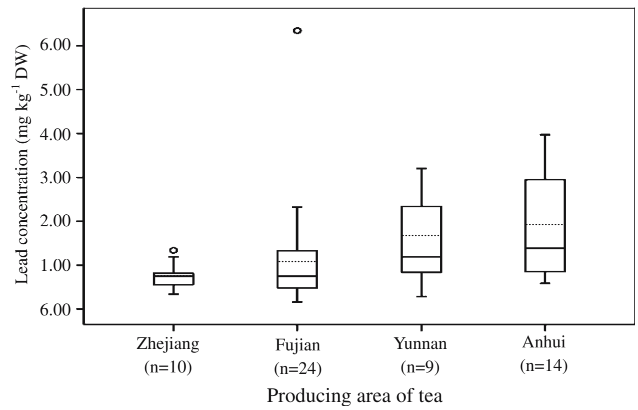

Fig. 3 Box plots of lead concentration in teas of different producing areas. See the Fig. 1 legend for an explanation of the box plot

production of black and Oolong teas than for green and scented teas. Older leaves tend to contain more lead than younger leaves (Han et al., 2006b). The different lead contents in different tea types could also be attributed to differences in the processing of these four tea types.

Lead contents in different tea samples of the same type also varied significantly. Lead concentrations, expressed in mg/kg of dry weight, varied from 0.162 to 1.421 for green tea (average, 0.736), from 0.554 to 1.188 for scented tea (average, 0.814), from 0.420 to 3.973 for Oolong tea (average, 1.736), and from 0.475 to 6.345 for black tea (average, 2.779). The lead contents in black tea and Oolong tea were in a much wider range than in green tea and scented tea. The lead contents in black tea and Oolong tea were in a much wider range than in green tea and scented tea. Previous studies demonstrated that the range for lead concentrations was less than 8 to 27.3 mg/kg in Turkey black tea samples and 0.11 to 1.93 mg/kg in 139 green tea samples in Japan (Narin et al., 2004; Tsushida and Takeo, 1977). The large variations of lead content in these studies were in accordance with our results.

Figure 2 shows box plots of lead concentration in normal and premium grade teas. Lead concentrations in 19 premium grade tea samples varied from 0.162 to 2.132 mg/kg of dry weight (median, 0.605 mg/kg; mean, 0.766 mg/kg). The lead concentrations in normal grade tea were found in a much wider range of 0.198 to 6.345 mg/kg of dry weight (median, 1.180 mg/kg; mean, 1.602 mg/kg). The median and mean lead concentrations were about 48% and 52% lower in the premium tea than in the normal tea, respectively. An ANOVA of log-transformed data showed a significant difference ($p < 0.05$) in lead concentrations between the two quality grades. This difference could be attributed to the different raw materials for processing of different grades of tea. The number of leaves per shoot picked for tea processing is the main difference between the normal and premium grades. Usually, premium grade tea is made from the unopened leaf bud and fewer leaves (1 to 2 in the case of green tea) than normal grade tea (3 to 4 in the case of green tea). Thus the leaves used to produce normal grade tea tend to be older than those used to produce premium grade tea.

Due to the accumulation of lead in the tea plants, lead concentrations tend to increase with age. It is reported that old leaves have 2- to 2.5-fold more lead than young leaves (Han et al., 2006b). Also, it is well known that premium grade tea usually is much better packed than normal grade tea, and thus has less potential to be contaminated by lead during transportation.

Figure 3 shows the box plots for lead concentrations in teas from different producing areas. An ANOVA of log-transformed data showed that only the lead concentrations in the teas from Anhui province significantly differed ($p < 0.05$) from that of the teas from the other three tea-producing areas. The average lead concentration in tea samples from different producing areas showed the following order: Anhui > Yunnan > Fujian > Zhejiang, which is consistent with the results of the Tea Research Institute, the Chinese Academy of Agricultural Sciences (Ding and Fan, 2005). This geographic difference in lead concentrations of tea could be attributed to the fact that the metal content of tea is influenced by soil composition and local environmental factors. Kumar et al. (2005) also found large variations in concentrations for many elements in teas from India and the United States.

The copper content in the investigated teas ranged widely from 1.790 to 48.19 mg/kg of dry weight (median, 15.69 mg/kg; mean, 17.13 mg/kg). The copper content in teas was higher by an order of magnitude, as compared with lead. Moreover, the copper content of all the tea samples in the current study were below the upper limits imposed on tea by various countries: China (60 mg/kg), Japan (100 mg/kg), and Australia, the United Kingdom, and the United States (150 mg/kg).

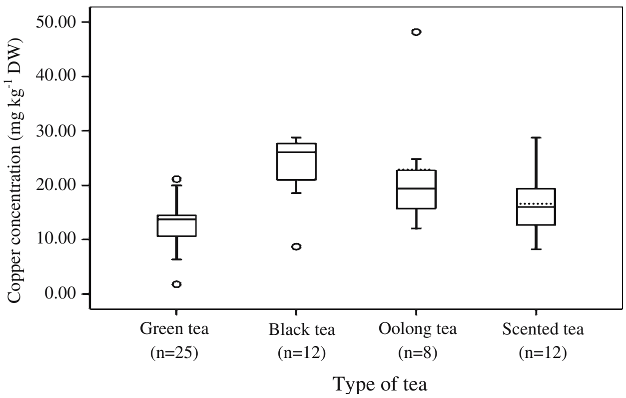

Fig. 4 Box plots of copper concentration in different tea types. See the Fig. 1 legend for an explanation of the box plot

Our results compared well with the literature values reported for tea samples from Turkey, India, China, and the United States (Kumar et al., 2005; Narin et al., 2004; Wang et al., 1993).The large variation of copper content in the investigated teas could be attributed to the different types, grades, and producing areas of the teas. However, no definite trend was observed for tea copper content between normal and premium grades and between different producing areas (data not shown). The pollution sources of copper were quite different from those of lead. Copper pollution was mainly from the rolling machine, whereas lead pollution was mainly from growth media and dust (Han et al., 2006a). Differences in tea copper concentration between different tea types are shown in Fig. 4. An ANOVA of log-transformed data showed significant differences ($p < 0.05$) in the copper concentrations between the different tea types. As shown in Fig. 4, the copper content in the different types of teas showed a trend similar to that found in lead.

The copper contents in different tea samples of the same type also varied significantly. Copper concentrations varied from 1.74 to 21.15 mg/kg of dry weight (average, 12.63 mg/kg) for green tea, 8.22 to 28.71 mg/kg (average, 16.35 mg/kg) for scented tea, 12.10 to 48.19 mg/kg (average, 22.02 mg/kg) for Oolong tea, and 8.72 to 28.78 mg/kg (average, 23.88 mg/kg) for black tea. The differences in methods used for processing and storage as well as the different types of leaves used could be contributory factors to this difference.

The current study showed that the lead and copper contents in teas marketed in Beijing were generally within the Chinese MPC, with a small proportion (1.75%) of the samples exceeding the Chinese MPC for lead and no sample exceeding the Chinese MPC for copper. The safety situation of teas marketed in Beijing was optimistic. The lead content varied among different types, grades, and producing areas. Scented and green teas contained less lead than black and Oolong teas, as did the premium grade as compared with the normal grade. Teas from Anhui had higher lead than teas from the other regions. However, no definite trend was observed for the copper content of tea between normal and premium grades or between different producing areas. The differences in copper concentrations between different tea types showed a trend similar to that for lead.

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