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Cadmium and Lead Content in Several Brands of Black Tea (*Camellia sinensis*) in Iran

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ABSTRACT: Tea is one of the most popular beverages in the world and contains several important essential micro nutrients, that are beneficial to human health. The contamination of tea leaves by heavy metals may pose serious problems to human health, because they are not biodegradable and remain in the environment and might enter the food chain. In this study, the concentration of heavy metals; Cd and Pb was measured by atomic absorption spectrometry (GFAAS) according to AOAC method of analysis. Ten samples of black tea cultivated in Iran were compared with 10 samples of imported black tea in 2011. The results of analysis showed that the mean level of Pb was 497.5 ± 175 ppb for Iranian and 293.3 ± 269.5 ppb for imported black tea. However, the value for Cd was 45 ±17ppb for Iranian and 9.14 ±7 ppb for imported black tea. The average contents of detectable heavy metals were significantly ($P<0.05$) higher in Iranian black tea. Cadmium and Lead concentrations in the sampled brands of black tea were lower in comparison with their upper limits (0.2 and 2 ppm for Cd and Pb, respectively), therefore, it might be concluded that there are not any health problems concerned with tea consumption, for these two elements.

Keywords: *Black Tea, Cadmium, Iran, Lead.*

Introduction

Tea (*Camellia sinensis*) is one of the most popular non-alcoholic beverages in the world because of its taste, aroma and low caffeine content (Mondal *et al.,* 2004). It has been frequently reported that drinking tea is beneficial to human health due to tea's antimutagenic, anticarcinogenic and antioxidant activities (Khokhar and Magnusdottir, 2002). However, various reports have noted the potential health implications of trace metals in tea, particularly since the tea bush is known to accumulate such metals (Bosque *et al.*, 1990; Wong *et al.*, 2003). For example, some researchers found that Chinese green tea possessed the highest contents of heavy metals among tested tea brands (Al-Oud, 2003). Another investigation revealed that among 57 tested tea samples marketed in Beijing (China), the concentration of lead varied from 0.198 to 6.345 mg/kg dry weight (Qin and Chen, 2006).

Cadmium (Cd) and lead (Pb) are widely distributed throughout the environment naturally or anthropogenically by agricultural and industrial utilizations. Power stations, metalworking industries, incineration of wastes, combustion of fossil fuels and the application of chemical fertilizers such as super phosphate, every year deposits considerable amounts of Cd in farmlands, thus increasing its levels in soil. Additionally, acid rain might increase the amount of Cd in the soil and expand the concentration of metals in agricultural products. In the case of Pb, the tetraethyl lead combustion products in gasoline are related into the atmosphere, becoming the

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major factors responsible for high Pb concentrations in air, water, soil and plants (Cui *et al.,* 2005; Singh *et al.,* 2010; Hongbin *et al.,* 2010; Tanmoy and Bhagat, 2010). Once ingested or absorbed by humans, they are known to be persistent in the human body with long excretion half-lives for decades and therefore classified as potentially toxic elements. Food consumption had been identified as the major pathway of human exposure to heavy metals, compared with other ways of exposure such as inhalation and dermal contact. Therefore the accumulation of heavy metals in the environment was of increasing concern due to food safety issues and potential health risks (McLaughlin *et al.,* 1999; Zwicker *et al.,* 2010).

Tea is one of the most important cash crops worldwide, particularly in Asian countries. However, the rapid industrialization of Asian countries over the past two decades has caused increased contamination of the environment by heavy metals, especially Pb and Cd, that has inevitably led to increased contamination of tea plants. However, current research is mostly concerned with the concentration of Pb and Cd in tea. There are few studies on the distribution of Pb and Cd in different varieties of tea plant or on the differences among tea varieties regarding their uptake and accumulation of Pb and Cd from the soil. Many factors might influence the concentration of Pb, Cd and other metals in both tea plants and final tea products, including the variety of tea plant, soil/atmospheric conditions, the maturity of raw materials used and the processing of leaves (Tanmoy and Bhagat, 2010; Jin *et al.,* 2005). The purpose of this study was to determine Cd and Pb concentrations in the several brands of Black Tea consumed in Iran.

Materials and Methods

All the reagents were of analytical grade unless otherwise stated. Double distilled water was used for the preparation of solutions. All the plastic and glass ware were soaked in nitric acid for 15 min and rinsed with deionized water before use. The stock solutions of metals (1000 mg/l) were obtained by dissolving appropriate salts of the corresponding metals (Merck) and further dilutions were carried out prior to use.

- Sampling

A total of 20 random samples of black tea cultivated in Iran (10 samples) and imported (10 samples) were purchased from markets in Iran in 2011. All the samples were collected in separate polyethylene containers, transported to the laboratory and stored for a short time at 4 °C until they were analyzed.

- Lead and Cadmium measurements

A Varian Model 240 FS atomic absorption spectrometer, equipped with a GTA 120 Graphite furnace, was used. Pyrolytic-coated graphite tubes with a platform were used and signals were measured at peak areas. The instrument settings and furnace programmers for analysis of trace elements are described in Table 1.

The samples were solubilized using high-pressure decomposition vessels, commonly known as a digestion bomb. A sample (1g) was placed in a Teflon container and 5 ml of concentrated HNO3 was added. The system was heated to 130 °C for 90 min and finally diluted to 25 ml with deionized water. A blank solution was prepared in the same way as the samples. Saturated solution of methyl isobutyl ketone (MIBK) was prepared. 2.0 g of ammonium pyrrolidine dithiocarbamate (APDC) was dissolved in 50 ml of distilled water. 20 ml of water-saturated MIBK was then added and shaken and allowed to separate. Extraction was repeated with 20 ml of MIBK and then 4 ml of water-saturated MIBK was added. The solution was allowed to stand for 5 minutes and then centrifuged for 3 minutes at 3000 rpm. The organic phase was used for determinations. All the metals were determined against prepared standards.

The detection limit is defined as the concentration corresponding to three times the standard deviation of ten blanks. Detection limit values of these elements as microgram per liter was found to be, 0.20 for Pb, 0.03 for Cd in GFAAS. The recovery rates for cadmium and lead were 98.4%, 97.8% respectively. The recovery values were nearly quantitative for wet digestion method. The relative standard deviations were less than 10% for all investigated elements. All samples were prepared and analysed in triplicate. All the results were expressed on a dry weight basis. The means and standard deviations (SD) were calculated using Microsoft Office Excel 2007. Statistical analysis was carried out using SPSS ver.18. Analysis of variance (ANOVA) was applied to detect significant differences.

Results and Discussion

The results of cadmium and lead contents in 20 samples of black tea brands both Iranian and Imported are shown in Table 2.

- Cadmium

The results indicated that the mean value of Cd concentration in Iranian black tea was 45.17 ± 18.5 (SD) ppb on dry weight basis and the range was from 16.7 to 69.85 ppb dry weight and the mean value of Cd concentration in imported black tea was 9.175 ± 6.75 (SD) ppb on dry weight basis and the range was from 0.05 to 19.05 ppb. Significant differences ($P < 0.05$) were found between the mean values of the Iranian and imported teas. The food sanitary standard of Cd in black tea is 0.2 ppm (Ashraf and Mian, 2008; Commission Regulation, 2006), therefore according to the current maximum allowable concentrations (MAC) the findings were much lower than the MAC in black tea.

Various reports have discussed the potential health implications of Cd in tea (Tsushida and Takeo 1977; Natesan and Ranganathan, 1990; Tsushida and Takeo, 1977; Shi *et al.,* 2008) and reported that the concentration of Cd in green tea in Japan was found to be 0.11 to 1.93 mg/kg with the mean value of 0.49 mg/ kg (Tsushida and Takeo, 1977). AL-Oud (2003) has reported the level of Cd in tea samples marketed in Pakistan to be within the range from below the detectable limit to 0.18 mg/kg.

Table1. Instrument settings and furnace programs for analysis of trace elements by AAS

Working conditions	Pb	Cd
Wavelength (nm)	283.3	228.8
Slit width (nm)	0.5	0.7
Lamp current (mA)	15	8
Air Flow (ml/min)	250	250
Injection volume (µl)	20	20
Heating program temperature °C	[ramp time (s) , hold time (s)]	
Drying 1	125(1,20)	115(1,20)
Drying 2	150(5,30)	140(5,30)
Pyrolysis	900(15,10)	950(15,10)
Atomization	2150(0,5)	2000(0,5)
Cleaning	2400(1,2)	2300(1,2)

Table 2. Cadmium and Lead contents of black tea (ppb, on dry weight basis)

Metals	Type of black tea	n	Mean	Std. Deviation	Min	Max
Cadmium	Iranian	10	45.17	18.5	16.7	69.85
	Imported	10	9.175	6.75	0.05	19.05
Lead	Iranian	10	447	222.2	57	852
	Imported	10	289.3	159.5	108	615

Seenivasan *et al*. (2008) analyzed one hundred black tea samples collected from the tea-growing regions of south India for Cd . The results showed that the level of Cd in black tea was between 0.05 and 0.38 mg/kg. Analysing the tea samples collected from various countries of the world, Ferrara *et al*. (2001) found that Cd in tea was below the detectable limit. However, Narin *et al*. (2004) reported that Cd level in fourteen Turkish black tea samples were 1 to 3.0 mg/kg with the mean value of 2.3 ± 0.4 mg/kg.

Mengel and Kirby (1987) noted that certain plants accumulated Cd without exhibiting any toxic symptoms. Further, they claimed that the availability of cadmium in soils was influenced positively by the addition of rock phosphate because rock phosphate is applied annually in many tea soils. Franklin *et al*. (2005) reported 4.9 to 5.5 mg/g of Cd in phosphatic fertilizers and 11.8 to 50.9 mg/kg in zinc sources and these are major sources of Cd in tea. Han *et al*. (2005) reported the Cd concentrations in 798 tea samples collected from the main tea producing provinces in China. The Cd concentrations in the tea samples ranged from below the detection limit to 1.07 mg/kg on dry weight basis with a mean value of 0.02 mg/kg. In another study, Zhang and Fang (2007) reported that Cd concentration in green tea leaves in China was at the lowest concentration among different trace metals, ranging from 0.012 to 0.057 mg/kg. From this study, it was concluded that the dissolved organic matter (DOM) in soil and acidification increased the bioavailablity of Cd in soil towards tea plants. Cadmium levels in black tea samples in Saudi Arabia were found to be in the range of 0.32 to 2.17 mg/kg with mean cadmium value of 1.1 mg/kg (Ashraf and Mian, 2008). These results indicate that there were substantial differences in Cd levels in black tea consumed in various areas in Asia and elsewhere.

- Lead

The results indicated that the mean value of Pb concentration in Iranian black tea was 447 ± 222.5 (SD) ppb on dry weight basis where the range was from 57 to 852 ppb and the mean value of Pb concentration in imported black tea was 289.3 ± 159.55 (SD) ppb on dry weight basis where the range was from 108 to 615 ppb. There were not significant differences ($P < 0.05$) between the mean values of the Iranian and imported black tea. The food sanitary standard of Pb in black tea is 2 ppm (Ashraf and Mian, 2008; Commission Regulation, 2006), therefore according to the current maximum allowable concentrations (MAC), 2 ppm was much lower than the MAC in black tea. Comparable results by Karimi *et al*. (2008), reported a mean level of 0.01 ppb Pb in tea infusion in Iran. Achudume and Owoeye (2010), Al-Oud (2003) and Yemane *et al.* (2008) reported that Pb concentrations were too low in tea samples but some others namely Han *et al.* (2006) who carried out analysis on 1225 tea samples collected in china reported higher levels of Pb. The Pb concentrations varied from 0.2 to 97.9 mg/kg on dry weight basis (DW) of tea samples in china, where 32% of the samples exceeded the national maximum permissible concentration (MPC) of Pb. Qin and Chen (2007) reported that the concentrations of Pb in the 57 tea samples marketed in Beijing (China) varied from 0.198 to 6.345 mg/kg based on dry weight. Ashraf and Mian (2008) in a study on black tea in Saudi Arabia found that the level of Pb in the 17 tea versions ranged between 0.3 and 2.2 mg/kg . Narin *et al.* (2004) found maximum Pb in Turkish tea samples at the level of 27.3±0.1 mg/ kg. It was indicated that the higher levels of Pb in tea samples could be attributed to the dust particle entry during tea processing and solder being used in packaging. Tsushida and Takeo (1977) reported that the concentration of Pb in green tea in Japan was found to be 0.11 to

1.93 mg/ kg. It was concluded that the green tea produced in some districts of Japan near the metropolis areas contained the highest concentration of lead. Among the 41 tea plant varieties, Chen *et al.* (2010) further reported that the ratio of Pb concentration in mature leaves to that in young leaves ranged from 1.7 to 6.5 where the minimum concentration of Pb in young leaf was 1 mg/ kg and the maximum was 19.8 mg/kg. The result of other researchers indicates that many factors and variables might have some effects on the absorption and storage of Pb in the tea plant varieties. The phenomenon of self-cleansing has an effect on the concentration of Pb in the river because a few of these elements are absorbed by the sediments and the organic and mineral compounds of the water, therefore, this lowers their level of pollution and concentration in the water. Washing stage in tea leaf processing in factories and pH are important factors that affect the concentration of Pb. An increase in pH causes a decrease in the solubility of the Pb compounds (Shi *et al.,* 2008; Seenivasan *et al.,* 2008; Ferrara *et al.,* 2010; Narin *et al.,* 2004; Mengel and Kirby, 1987). Other factors including the ionic exchange capacity and the competitive effect of the other metallic cations on each other might have effects on the rate of absorption of Pb in tea plant varieties.

Conclusion

This study indicated that several brands of Black Tea had Cd and Pb contents below the food sanitary standards of 0.2 ppm and 2 ppm for Cd and Pb, respectively. In conclusion, this work has shown that several brands of Black Tea produced, consumed and imported to Iran is safe in respect of cadmium and lead contents.

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