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DETERMINATION OF HEAVY METALS IN IRANIAN AND IMPORTED BLACK TEA

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ABSTRACT

Tea is the most popular beverage in the world and contains several essential nutrients, which are beneficial for human health. The contamination of tea leaves by heavy metals may pose a serious threat to human, because they are not biodegradable and remain in environment and pass to food chain. In this study, the concentration of heavy metals of Cd, Pb, Ni, and Al and macro-elements of Fe, Zn, Cu, and Mn were determined by atomic absorption spectrometry on 30 samples of black tea cultivated in Iran and compared with the results for 30 samples of imported black tea in 2006. The results of analysis showed that the mean level of Al was 699.2±172.7mg/kg for Iranian and 388.3±98.3mg/kg for imported black tea. However, the values for Cd, Pb, and Ni were non-detectable. The most abundant nutritive metal was manganese with 155.2-214.2mg/kg and 96.7-332.9mg/kg in Iranian and imported black tea, respectively. The average contents of detectable heavy metals were significantly ($p<0.05$) higher in Iranian black tea. According to the results of this study, it is justifiable to set maximum residue level for heavy metals in tea, such as Al which appears to be very high in concentration.

Key words: Black tea, heavy metal, aluminum, Iran

INTRODUCTION

Tea (*camellia sinesis*) is one of the most popular beverages all over the world. It has originated from China long time ago. About 75% of the estimated 2.5 million metric tons of dried tea that are manufactured annually are processed as black tea which are consumed by many countries (Nas *et al*., 1993). Currently, consumption of tea as a soft drink is very popular in Iran and its production in 2006 was about 78,000 tons.

Various reports have discussed the potential health implications of trace metals in tea, particularly since the tea bush is known to accumulate trace metals (Bosque *et al*., 1990 and Anonymous, 1999), cadmium, lead, nickel, and aluminum (Al), hereinafter referred to as toxic heavy metals and may pose a serious threat to plant, animal, human and environmental health, because they are not degradable by bio-process and remain in environment and passes to food chain. It is globally accepted that some heavy metals such as Fe, zinc, copper, and manganese (Mn); hereinafter referred to as macro-elements; are essential for healthy growth and development within certain permissible limits. Macro-elements are of great biological importance despite their small contribution to the body weight, although excess intake of them can cause chronic toxicity in human. The determination of these elements in beverages, water, food, plant and soil is thus of outermost important tasks. One of the major food sources of these metals is green leafy vegetable (McLaren, 1991). Several attempts have been made to assess tea quality by chemical analysis. However, to date, few works has been performed to determine the metal contents of tea due to the analytical difficulties associated with both the separation of such components and their quantitative measurements. Metallic constituents of tea leaves are normally different according to the type of tea (green or black) and geological source (Marcos *et al*., 1996).

As the issue was not much studied in Iran, the main objective of this study was set to determine the quantity of heavy metals (Cd, Pb, Ni, Al, Fe, Zn, Mn, and Cu) in black tea cultivated in Iran

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and imported. Finally, the results are compared with the outcomes of other related publications in this respect.

MATERIALS AND METHODS

Samples and chemicals

The black tea cultivated in north of Iran and also imported black teas from other countries were brought to the packaging factory of tea collected in July 2006. The study was then carried out at water science laboratory, at Food and Agriculture Department, Institute of Standard and Industrial Research. Thirty samples of each group (Iranian black tea and imported black tea) were examined. Samples were measured for their toxic (Cd, Pb, Al, and Ni) and macro-elements (Fe, Zn, Cu, and Mn). All chemicals were of analytical grade with Merch Chemical Company, Darmstadt, Germany and were used without further purification.

Analytical measurements

All heavy metals contents were determined by drying 10-20g of test portion to nearest 0.001g on a water-bath at 100°C and then ashed at 450°C under a gradual increase ($\leq$ 50°C/h) in temperature. Five mL hydrochloric acid 6M (1+1) was added and the solution was evaporated in water bath to dryness. The residue was dissolved in 10.0-30.0mL nitric acid 0.1M. Blanks were treated in the same way as products and the analys were determined by atomic absorption spectrometry, Varian-Plus 10 instrument. Wavelength, gas mixture, temperature program and other instrumental parameters that are most appropriate for each metal were provided with manual of instrument. Also, calibration curve of each heavy metal was drawn with working standard solution before testing (AOAC, 2000).

Statistical analysis

Statistical analysis of the obtained results was performed according to paired-samples t-test employing the SPSS statistical program. Means of duplicate analysis from 30 samples of each region were reported for the studied parameters.

RESULTS

As shown in Table 1 the descriptive statistical parameters are presented for Iranian black tea. It should be noted that the concentrations of heavy metals, such as Ni, Pb, and Cd were not in the detectable ranges. However, the average concentration of Al was determined as 699.2 mg/kg which appears to be quite high. The descriptive statistical parameters for imported black tea are presented in Table 2. Similar to what was mentioned for Iranian black tea earlier, the concentrations of heavy metals, such as Ni, Pb, and Cd were not in the detectable ranges. The average concentration of Al was determined as 388.3mg/kg which is much lower than the Al content of Iranian black tea.

Table 1: Concentration of heavy metals in Iranian black tea, mg/kg

Statistical analysis*	Fe	Cu	Zn	Mn	Ni	Pb	Cd	Al
Min.	17.2	19.6	34.1	155.2	ND**	ND	ND	405.0
Max.	194.0	36.7	47.4	214.2	ND	ND	ND	1013.0
Mean	92.6	29.3	40.3	182.9	ND	ND	ND	699.2
SD	45.0	4.9	3.8	15.0	ND	ND	ND	172.8
Median	83.1	30.8	39.1	183.4	ND	ND	ND	699.3

* No.=30 **ND: Non-detectable

Table 2: Concentration of heavy metals in black tea imported, mg/kg

Statistical analysis*	Fe	Cu	Zn	Mn	Ni	Pb	Cd	Al
Min.	17.6	0.6	21.6	96.7	ND**	ND	ND	266.7
Max.	322.8	20.1	46.7	332.9	ND	ND	ND	622.0
Mean	73.0	8.9	35.8	146.6	ND	ND	ND	388.3
SD	72.2	5.7	7.4	59.6	ND	ND	ND	98.3
Median	45.3	8.6	38.5	124.9	ND	ND	ND	352.0

* No.=30 **ND: Non-detectable

The average values of all determined heavy metals for both groups of tea are illustrated in Fig. 1. As this figure indicates the concentrations of heavy metals are presented in a descending sorted style. According to the performed t-test, the concentrations of all studied heavy metals in Iranian black tea were significantly higher ($p<0.05$). The variation of heavy metals concentrations is shown in Fig. 2. As it is shown, the Fe and Mn contents varied widely among the samples of both groups.

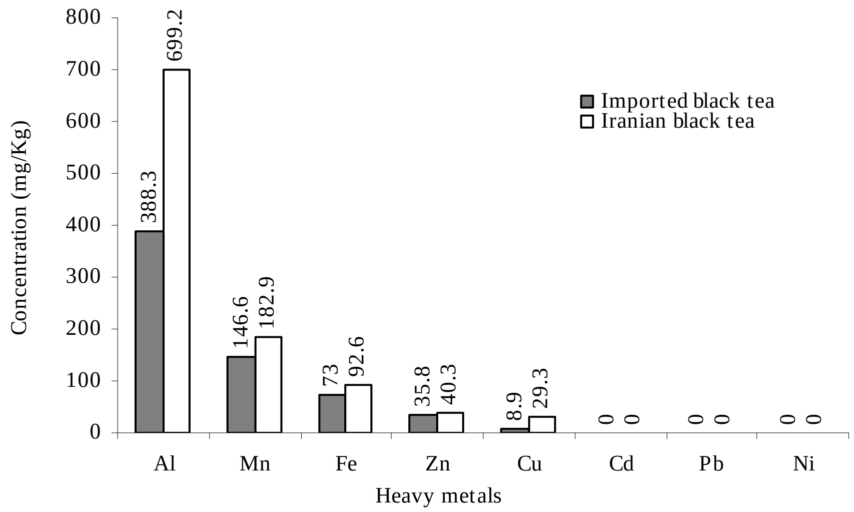

Fig. 1: Comparative concentrations of heavy metals in Iranian and imported black tea samples

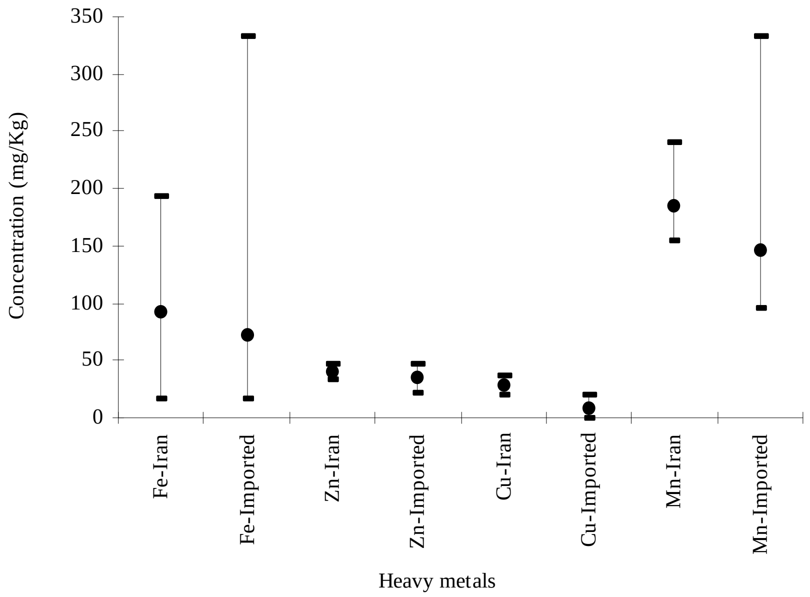

Fig. 2: Variation of heavy metals concentrations in Iranian and imported black tea samples

The correlations between heavy metals for Iranian black tea and imported black tea were determined which are presented in Table 3 and Table 4, respectively. According to the correlation analysis, there were significant relationship between Cu-Al and Zn-Al in all of the tea samples. Also, significant relationships were shown between Fe-Mn and Cu-Zn in imported black teas.

Table 3: Correlations between heavy metals for thirty samples of Iranian black tea

		Fe	Cu	Zn	Mn	Ni	Pb	Cd	Al
Fe	Pearson Correlation	1	-0.285	-0.185	-0.141	-0.037	.(a)	.(a)	-0.347
	Sig. (2-tailed)	.	0.223	0.436	0.553	0.876	.	.	0.133
Cu	Pearson Correlation	-0.285	1	0.347	0.400	-0.395	.(a)	.(a)	0.864**
	Sig. (2-tailed)	0.223	.	0.134	0.081	0.085	.	.	0.000
Zn	Pearson Correlation	-0.185	0.347	1	-0.304	0.057	.(a)	.(a)	0.645**
	Sig. (2-tailed)	0.436	0.134	.	0.193	0.813	.	.	0.002
Mn	Pearson Correlation	-0.141	0.400	-0.304	1	-0.259	.(a)	.(a)	0.202
	Sig. (2-tailed)	0.553	0.081	0.193	.	0.270	.	.	0.394
Ni	Pearson Correlation	-0.037	-0.395	0.057	-0.259	1	.(a)	.(a)	-0.323
	Sig. (2-tailed)	0.876	0.085	0.813	0.270	.	.	.	0.164
Pb	Pearson Correlation	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)
	Sig. (2-tailed)	.	.	.	.	.	.	.	.
Cd	Pearson Correlation	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)
	Sig. (2-tailed)	.	.	.	.	.	.	.	.
Al	Pearson Correlation	-0.347	0.864**	0.645**	0.202	-0.323	.(a)	.(a)	1
	Sig. (2-tailed)	0.133	0.000	0.002	0.394	0.164	.	.	.

**. Correlation is significant at the 0.01 level (2-tailed)
a. Cannot be computed because at least one of the variables is constant

Table 4: Correlations between heavy metals for thirty samples of imported black tea

		Fe	Cu	Zn	Mn	Ni	Pb	Cd	Al
Fe	Pearson Correlation	1	-0.164	-0.088	0.592**	.(a)	.(a)	.(a)	0.290
	Sig. (2-tailed)	.	0.489	0.712	0.006	.	.	.	0.216
Cu	Pearson Correlation	-0.164	1	0.525*	-0.186	.(a)	.(a)	.(a)	-0.445*
	Sig. (2-tailed)	0.489	.	0.018	0.432	.	.	.	0.049
Zn	Pearson Correlation	-0.088	0.525*	1	0.011	.(a)	.(a)	.(a)	-0.484*
	Sig. (2-tailed)	0.712	0.018	.	0.965	.	.	.	0.030
Mn	Pearson Correlation	0.592**	-0.186	0.011	1	.(a)	.(a)	.(a)	0.195
	Sig. (2-tailed)	0.006	0.432	0.965	.	.	.	.	0.410
Ni	Pearson Correlation	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)
	Sig. (2-tailed)	.	.	.	.	.	.	.	.
Pb	Pearson Correlation	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)
	Sig. (2-tailed)	.	.	.	.	.	.	.	.
Cd	Pearson Correlation	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)	.(a)
	Sig. (2-tailed)	.	.	.	.	.	.	.	.
Al	Pearson Correlation	0.290	-0.445*	-0.484*	0.195	.(a)	.(a)	.(a)	1
	Sig. (2-tailed)	0.216	0.049	0.030	0.410	.	.	.	.

**. Correlation is significant at the 0.01 level (2-tailed)
*. Correlation is significant at the 0.05 level (2-tailed)
a. Cannot be computed because at least one of the variables is constant

DISCUSSION

The results of total contents of the studied heavy metals (Al, Pb, Cd, and Ni) in black tea of both Iranian region and imported tea showed the ability of these plants in accumulating Al. The average value of Al in Iranian black tea and imported black tea were 699.2 and 388.3mg/kg and their concentrations varied from 405.0 to 1013.0 and 266.7 to 622.0mg/kg, respectively. Furthermore, it has been indicated that Al could be accumulated in tea leaves up to 23,000mg/kg which considered much higher than the other plants accumulation which do not normally exceed 200mg/kg (Coriat and Gillard, 1989; Atta, 1995).

The comparison of Al in two groups showed significant differences ($p<0.05$). Zhang, M. *et al*., (2006) also reported that there were relationship between extractable metals (Al, Cu, Cd, Pb, and Zn) in acid soil and metals taken up by tea plants. Results of their study indicated that long-term plantation of tea can cause soil acidification and elevated concentrations of bioavailable heavy metals in the soil, hence, enhance the risk of heavy metals accumulation in tea leaves. Long-term plantation of Iranian black tea might be considered as a cause of high Al content which needs to be further studied. As the Al content of tea is dependent to its concentration in soil, applying best agricultural practice (BAP) would be considered as an alternative to control the Al contents in tea leaves.

The concentrations of other toxic metals, Pb, Cd, and Ni were not detectable using the available analytical techniques. AL-Oud (2003) reported too low concentrations of Pb and Cd in tea leaves. But, comparing with results of Seenivasan *et al*., (2007), the average values recorded for Pb, Cd, and Ni were generally higher than those obtained in our study.

As it is indicated in Fig. 1, Mn was the most abundant metal in Iranian and imported black tea (excluding Al). This result confirms the outcomes of similar study which was conducted by AL-Oud (2003). The quantitative order of studied heavy metals in both Iranian and imported black tea as mg/Kg is presented as follow:

For Iranian black tea:

Mn (155.2-214.2)>Fe (17.2-194.0)>Zn (34.1-47.4)>Cu (19.6-36.7)

For imported black tea:

Mn (96.7-332.9)>Fe(17.6-322.8)>Zn(21.6-46.7)>Cu (0.6-20.1)

The average contents of macro-elements (Mn, Fe, Zn, and Cu) in Iranian black tea were 182.9, 92.6, 40.3, and 29.3mg/kg, respectively. These contents significantly ($p<0.05$) were higher than those for imported black teas with the average values of 146.6, 73.0, 35.8, and 8.9mg/kg. Accordingly, the nutritional values of cultivated teas in Iran were better than the others which were imported to Iran and included in our study. As a result, tea could be considered as an important source of Mn.

Although toxic effects of heavy metals such as Cu, Fe, Zn and Mn have been sufficiently discussed by Mahindru, (2004), beneficial effects of them in a fairly narrow concentration range between the essential and the toxic levels are also described.

Fe and Mn content varied widely among the tea samples of both groups. The variations were higher in imported tea than the other group. Seenivasan *et al*., (2007) reported that there were wide variations in the heavy metal content of black teas collected from the different regions of south India for the different agro-climatic regions. The wide variations of Fe and Mn in the present study might be due to the different agro-climatic regions as well.

Inter-relationships between the investigated metal concentrations showed that the correlation between Cu-Al and Zn-Al (in two groups of teas) and Fe-Mn and Cu-Zn in imported black teas were significant. The other correlations of metal concentrations of the samples were not significant. Due to the lack of information specifying the acceptable contents of heavy metals in black tea, regulatic on the maximum allowable and safe concentration of each metal in black tea are urgently needed. Routine check and frequent analysis of foodstuff is also required with intention to avoid the risk of exceeding the intake beyond the tolerance limits standards.

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REFERENCES

AL-Oud, S. S., (2003). Heavy metal contents in tea and herb leaves. Pak. J. Biol.Sci., **6** (3):208-212.

Anonymous, (1999).Total diet study: aluminum, arsenic, cadmium, chromium, copper, lead, mercury, nickel, selenium, tin, and zinc. Food Surveillance Information, Sheet No.191.

AOAC., (2000). Association of Official Agricultural Chemists, official method 999.11, .Determination of Lead, Cadmium, Copper, Iron and Zinc in foods. Ch. 9, 19.

Atta, M. B., (1995). Aluminum content in dust black tea leaves and its beverages. Menofiya. J. Agri. Res., **20**: 137-150.

Bosque, M. A., Schuhmacher, M., Domingo, J. L., (1990). Concentration of lead and cadmium in edible vegetables from Tarragona province, Spain. Sci. Total Environ., **95**: 61-70.

Coriat, A. M., Gillard, R. D., (1996). Beware the cups that cheer. Nature., 321:570.

Mahindru, S. N., (2004). Pathways of heavy metals entrance in the foods. Food Contaminants- Origin, Propagation and Analysis. A. P. H. Publishing Corporation, New Delhi, India, Ch. **14**: 171-183.

Marcus, A., Fisher, A., Ree, J., Hill, S. J., (1996). Preliminary study using trace element concentrations and a chemometrics approach to determine the geological origin of tea. J. Agric. Atom. Spect., **113** : 521-525.

McLaren, D. S., Burman, D., and Belton, N. R., (1991). Textbook of Paediatric Nutrition. Ed. 3, Churchill Livingstone Co., London., 451-483.

Nas, S., Gokalp, H. Y., and Sahin, Y., (1993). K and Ca content of fresh green tea, black tea and the tea residue determined by x-ray fluorescence analysis technique. Zeitschrift fur Lebensmittel-Untersuchung und-Forschung., **196**: 32-37.

Seenivasan, S., Manikandan, N., Muraleedharan, N. N., and Selvasundaram, R., (2007). Heavy metal content of black teas from south India. http://www.elsevier.com/locate/foodcont

Zhang, M., Zhou, C., and Huang, C., (2006). Relationship between extractable metals in acid soils and metals taken up by tea planets. http://cat.inist.fr/?aModele=afficheN&cpsidt=17561212.