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Heavy Metal Contents in Tea and Herb Leaves

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Abstract: The quality of tea brands and herbs available in the retail market in the Kingdom of Saudi Arabia were assessed based on contents of heavy metals in their tissues. All tested brands of tea and herbs possess considerable amounts of the eight tested heavy metals, Mn, Fe, Zn, Cu, Ni, Co, Pb and Cd. The tested nine brands of tea as well as five herbs proved high variability ($P < 0.01$) in their contents of heavy metals. Among tested heavy metals, Mn was the most abundant one in tea leaves (390-900 $\mu g\ g^{-1}$) whereas Fe was the predominance one in herb leaves (326-1755 $\mu g\ g^{-1}$). Fortunately, toxic heavy metals, Pb and Cd, had the lowest contents in both tea and herb leaves. Among tested tea brands, Chinese green tea possesses the highest contents of heavy metals. Concentrations of tested heavy metals in tea and herb beverage were markedly lower than their total contents. The concentrations of toxic heavy metals, Pb and Cd were too low to be detected in beverage using the available analytical techniques. The solubility of studied heavy metals in both brew and infusion extracts varied widely and ranged from 0.0−48%. The lowest rates of solubility were listed for toxic heavy metals Pb and Cd. The amounts of heavy metals that one may take up through consumption of tea and herb beverages were found to match the acceptable daily intake that takes into account exposure from air, food and drinking water.

Key words: Herbs, tea, infusion, brew, heavy metals

Introduction

Tea is one of the most popular beverages all over the world. The 75% of the estimated 2.5 million metric tons of dried tea that are manufactured annually processed as black tea which consumed by many countries (Nas *et al.*, 1993). In the UK, one liter of tea, in average, is consumed per person per day (Nas *et al.*, 1993).

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; Anonymous, 1999). For example, aluminium reported in tea leaves to reach 23,000 ppm levels in tea leaves, which considered higher than other plants that do not normally exceed 200 ppm (Atta, 1995; Coriat and Gillard, 1986).

Human body requires both metallic and non-metallic elements for healthy growth and development within certain permissible limits. (The optimum concentration needed for this purpose varies widely from one element to another; from infant to childhood to adult and from male to female). The determination of these elements in beverages, water, food, plant and soil is thus of outermost important task. Tremendous development in research tolerance limits for daily intake of nearly all essential elements needed for healthy growth and sound physiological changes in human body have been cited by authorized specialists (Jackson and Lee, 1988; Marcos *et al.*, 1996). Precise warnings have also been declared to avoid hazardous toxic elements. Tea-drinking habit is worldwide spread and many countries cultivate different brands of tea to meet the increasing demands. These quite great diverse brands of tea are well known to consumers. Very recent researches were confirmed the positive and negative effects of drinking tea on the health. It was pointed out that some of the beneficial effects of drinking tea are prevention of chronic and cardiovascular disease, cancer, antioxidative detoxification and removal of cadmium in administered rates (Lee *et al.*, 1995). Studies were extended to include biochemical studies of tea-polyphenols found to inhibit every stage of the multi-carcinogenic process (Anonymous, 1990).

Several attempts have been made to assess tea quality by chemical analysis, usually with reference to pigmentation and the flavouring characteristics. However, to date, little work has been done to identify the metal containing components of tea due to the analytical difficulties associated with both the separation of such components and their quantitative measurements. Metallic constituents of tea leaves is normally different according to the type of tea (green or black) and geological source (Marcos *et al.*, 1996). In this study different tea brands imported to the Kingdom of Saudi Arabia were investigated for their contents of heavy metals in relation to the procedures adapted by people for making and cooking tea as a hot drink.

Materials and Methods

Nine marked brands of black tea and another five herbs, which commonly consumed in Saudi Arabia and other countries, were collected from local markets in March 2002. The study then was carried out at soil and water sciences laboratory at College of Agriculture and Veterinary Medicine, King Saud University, Al-Gassim. Tea, *Thea Sinensis* brands include Lipton (India), Tylos and Azizia (Sir-Lanka), Alwzah (Selan), Earl grey (England), Chinese green tea (China), Lord (Selan) and Bentleys (India). The collected herbs includes Babong (Spain), Maramyah, *Salvia deserti* (Syria), Mixed herbs (India) , Carcadeh, *Hibiscus Sabdariffa* (Sudan) and Mint, *Mentha longifolia* (Saudi Arabia). Stock of these brands in tea bags (2 g of each) or package of 225 g of either coarse or fine particles were collected from the retail market in Burydah city Al-Qassim, Saudi Arabia.

Preparation of tea beverage: The two methods commonly used for preparation of tea and herb beverages were adopted for this study to assess the actual amount of heavy metal reach human body through drinking such beverages. These are following two methods:

Brew: In this method, 2 g of black tea particles or one tea bag was boiled with 100 ml of distilled water for 5 min. The mixture was held for 5 min at room temperature and then filtered. Then tea bag was removed to obtain the clear solution for analysis.

Infusion: In this method, 100 ml of hot distilled water was added to either 2 g of black tea particles or one tea bag. The mixture left to cool at room temperature for 5 min and then filtered to obtain the clear solution for further processing.

Total contents of metals: Portions of 0.5 g of each brand were digested using 10 ml of a mixture (2:1 v/v) of concentrated HNO_3 and HCl. The mixture was heated on sand bath until the solution turned white and gives out the white fumes. The digest was transferred into 100 ml volumetric flask and the volume was adjusted to the mark using distilled water. Concentrations of heavy metals (Fe, Zn, Mn, Cu, Pb, Co, Cd, Ni) were determined in the obtained clear solutions using ICP (XL Integra, GBC, Australia).

Statistical analysis: Statistical analysis of the obtained results was preformed according to paired-samples t-test employing the SPSS statistical programme. The results of statistical analysis showed that both Fe and Mn contents in all extracts (total, infusion and brew) varied significantly ($P<0.05$) between tested brands. Also, significant differences were recorded between the overall means of Mn, Cu and Ni in infusion and brew extracts using least significant different ($P < 0.05$) LSD.

Results and Discussion

Total contents of heavy metals: The results of total contents of the studied heavy metals in leaves of both tea and herb (Table 1) show the ability of these plants to accumulate heavy metals, particularly Mn and Fe, to a lesser extent Zn and Cu. The most abundant metal in tea leaves was Mn (390-900 $\mu g\ g^{-1}$), whereas in herb, Fe was the predominance one (326-1755 $\mu g\ g^{-1}$). On the other hand, Mn content in herbs and Fe in tea were relatively lower than the above mentioned values and ranged from 8.00-792.00 and 123.90-513.00 $\mu g\ g^{-1}$, respectively. These two metals are followed by Zn, then Cu. Their values varied from 34.17-56.78 and 26.69-53.89 for Zn, against 9.04-23.36 and 22.12-40.66 $\mu g\ g^{-1}$, for Cu in herb and tea leaves, respectively. Contents of non-nutrients heavy metals, Ni, Co, Pb and Cd in both tea and herb leaves were rather low. These metals could be arranged in descending order according to their contents in both tea and herb leaves as follows:

Ni (2.46-8.90) > Pb (0.03-14.84) > Co (Nil-2.35) > Cd (Nil-0.37)

The obtained results show that the contents of heavy metals varied widely among the tested brands of both tea and herbs. Among tested tea brands, Chinese one has the highest contents of studied heavy metals, Mn, Fe, Zn, Cu and Pb. The level of Pb (14.84 $\mu g\ g^{-1}$) in Chinese tea was too high which tea consumers should consider. Also, the results show that all tested herbs possess high contents of Fe.

Heavy metal concentrations in tea and herb beverage: The concentrations of toxic heavy metals, Pb and Cd were too low to be detected by ICP or graphite furnace atomic absorption spectrophotometer. Also, the concentrations of other two non-nutrients metals (Ni, Co) in the tested substrates were relatively low and ranged from Nil-5.30 and Nil-0.07 $\mu g\ g^{-1}$, respectively. On the other hand, the concentrations of nutrients heavy metal (Mn, Fe, Zn and Cu) in infusion extract are relatively high and varied widely between tea and herb. Except for Carcadeh, the concentrations of Mn and Zn in herb beverage were much lower (3.40-339.60 and 1.95-10.55) than those (68.60-305.60 and 8.55-28.60 $\mu g\ g^{-1}$) in tea leaves, respectively. Contradiction to these results, concentrations of Fe in herb leaves were much higher (62.00-185.00) than those (19.00-135.00 $\mu g\ g^{-1}$) in tea leaves, whereas the concentrations of Cu were more or less similar in both tea and herb (Table 2).
General view on contents of heavy metals in brew extract

Table 1: Total contents of heavy metals (μg g^{-1}) in both black tea and herb leaves

Tea	Cu	Mn	Fe	Zn	Pb	Co	Ni	Cd
Alwzah	26.08	390	123.90	28.22	1.56	N.D	3.50	0.02
Azizia tea	34.16	644	155.80	26.69	0.47	1.05	5.60	N.D
Lipton (lose)	24.08	610	194.70	31.96	0.35	0.25	4.80	N.D
Chinese green tea	40.66	900	513.30	53.89	14.84	N.D	5.60	0.18
Earl grey	26.46	878	262.20	35.36	1.95	0.05	6.48	0.01
Tylos	28.42	546	137.80	32.81	0.29	2.35	5.70	0.04
Lipton (Bag)	22.12	866	209.20	33.15	0.21	0.04	4.74	0.11
Lord	24.52	686	259.40	39.10	0.03	N.D	5.44	N.D
Bentleys	31.40	642	188.70	30.43	0.63	0.10	4.80	0.10
Herb								
Babong	19.30	68	1253.80	43.86	0.45	N.D	3.30	0.12
Maramyah	9.34	60	521.00	47.26	2.08	0.11	3.80	N.D
Mixed herbs	23.36	8.00	557.10	56.78	1.31	0.55	2.46	0.05
Carcadeh	9.04	792	326.40	34.17	2.42	0.29	6.50	0.05
Mint	26.00	130	1755.60	55.93	1.40	0.15	8.90	0.37

N.D. = Not detected

Table 2: Heavy metal contents (μg g^{-1}) in infusion of both tea and herb leaves

Tea	Cu	Mn	Fe	Zn	Co	Ni
Alwzah	4.42	108.10	50.98	11.70	N.D	2.52
Azizia tea	3.85	227.60	32.83	8.55	0.03	3.50
Lipton (lose)	3.85	211.10	56.00	14.25	N.D	1.50
Chinese green Tea	17.48	160.60	135.00	28.60	N.D	2.40
Earl grey	4.50	287.60	32.00	17.85	N.D	5.30
Tylos	11.30	133.10	23.00	10.15	N.D	N.D
Lipton (Bag)	3.33	305.60	19.00	11.90	0.04	2.50
Lord	0.33	121.60	23.20	9.90	N.D	1.75
Bentleys	6.17	68.60	30.00	14.10	0.07	0.55
Herb						
Babong	18.98	8.15	95.00	10.55	N.D	2.48
Maramyah	3.40	3.40	109.00	9.55	0.07	1.15
Mixed herbs	2.70	6.55	88.00	7.30	0.05	1.95
Carcadeh	7.60	339.60	62.00	30.35	0.02	3.86
Mint	0.40	39.60	185.00	1.95	0.01	2.64

Table3: Heavy metal contents (μg g^{-1}) in brew of both tea and herb leaves

Tea	Cu	Mn	Fe	Zn	Ni
Alwzah	5.40	66.25	49.15	25.22	0.00
Azizia tea	2.40	110.25	26.15	14.62	0.00
Lipton (lose)	0.30	158.00	42.63	11.87	0.80
Chinese green Tea	6.95	173.45	103.00	14.27	0.00
Earl grey	3.90	197.95	20.77	16.27	0.00
Tylos	N.D	105.65	30.58	13.27	0.00
Lipton (Bag)	0.35	237.25	13.20	13.72	0.00
Lord	1.85	141.25	16.41	13.27	0.07
Bentleys	0.70	81.30	81.39	5.72	0.60
Herb					
Babong	5.35	7.03	54.00	8.72	0.95
Maramyah	1.55	2.81	52.50	6.62	0.00
Mixed herbs	4.45	6.30	91.00	5.47	1.75
Carcadeh	0.50	361.80	79.00	19.72	1.41
Mint	2.75	11.65	82.50	13.02	0.55

Table 4: Overall means of heavy metals in brew and infusion extracts as well as total content ($\mu g\ g^{-1}$) in tea and herbs

		Brew		Infusion		Intake ($\mu g\ day^{-1}$)	
Heavy metals	Total	($\mu g\ g^{-1}$)	Solubility (%)	($\mu g\ g^{-1}$)	Solubility (%)	Brew	Infusion
Tea							
Cu	28.66	2.43	8.47	6.14	21.43	.01	0.04
Mn	684.67	141.26	20.63	180.43	26.35	0.85	1.08
Fe	227.22	42.59	18.74	44.67	19.66	0.26	0.3
Zn	34.62	14.25	41.15	14.11	40.76	0.09	0.08
Pb	2.26	N.D	0.00	N.D	0.00	0.0	0.0
Co	0.43	0.16	0.00	0.02	3.65	0.0	0.0
Ni	5.18	0.16	3.15	2.22	42.91	0.0	0.01
Cd	0.05	N.D	0.00	N.D	0.00	0.0	0.0
Herb							
Cu	17.41	2.92	16.77	6.62	38.01	0.02	0.04
Mn	211.60	77.92	36.82	79.46	37.55	0.47	0.48
Fe	882.78	71.80	8.13	107.80	12.21	0.43	0.65
Zn	47.60	10.71	22.50	11.94	25.08	0.06	0.07
Pb	1.53	N.D	0.00	N.D	0.00	0.0	0.0
Co	0.22	N.D	0.00	0.03	13.64	0.0	0.0
Ni	4.99	0.93	18.67	2.42	48.40	0.0	0.02
Cd	0.12	N.D	0.00	N.D	0.00	0.0	0.0

* These amounts calculated based on the concentration of metal in brew and infusion extracts and on the assumption that the average consumption of tea beverage for single person is three cups a day with one packet of 2 g (for each), i.e. 6 g of tea particles day^{-1}.

(Table 3) showed that the concentrations of toxic heavy metals, Pb, Cd, as well as Co were too low to be detected using the available analytical techniques. That is besides significant decrease ($P<0.05$) in Mn, Cu and Ni concentrations in brew extract comparing with their corresponding values in infusion. This drop could be attributed to chelation of these metals with tannic acid and tannins which exudates during the boiling of tea particles. Precipitation of these chelates leads to the noticed decrease in metal concentration in brew extract. Similar to the remark of the results of infusion extract, except for Carcadeh, concentrations of Mn and Zn metals in herb beverage were generally lower (2.81-11.65 and 5.47-13.20) than those (66.25-237.25 and 5.72-25.22 $\mu g\ g^{-1}$) recorded for tea ones. Concentrations of Cu and Fe in herb brew were approximately matching those of tea ones. The studied heavy metals could be arranged in descending order according to their contents in tea brew as the following:

Mn > Fe > Zn > Cu > Ni

In herb brew, Fe was the predominance metal.

Comparing with the results of Atta *et al.* (2000), the overall means recorded for some heavy metals in various extracts (total, infusion, brew) were generally higher than those obtained for this study (Table 4). Levels of Cu, Fe, Zn and Ni in the brew extract were approximately 3, 2, 2.5 and 24 folds of this study. For infusion extract, except for Cu, levels of Fe, Zn and Ni were 1.5, 2 and 1.5 time of this study. Also, total contents of Fe, Zn and Pb were roughly 4, 3 and 5.5 times of this study.

Intake and solubility of heavy metals: Due to the lack of information, naming the maximum allowable levels of heavy metals in tea leaves, the discussion will be extended to the acceptable daily intake that taken into account exposure from air, food and drinking water. For instance, the expected calculated intake (Table 4) of Mn of the present study was 0.85 and 1.08 for tea brew and infusion, respectively, against 0.47 and 0.48 mg day^{-1} for herb ones. These values are much higher than intake from food (0.008) and at the same time much lower than that (3.8 mg day^{-1}) from drinking water according to the US environmental protection agency, Anonymous (1997). The calculated overall mean of Ni for tea and herb brew were zero and those for infusion were 0.01 and 0.02 μg day^{-1}, respectively. These values were much lower than human requirements (50 $\mu g\ day^{-1}$) of this element and rather lower than the population average intake in the UK (0.13 mg day^{-1}) recorded by total diet study (Anonymous, 1999). This connection (Anonymous, 1998) recorded that tea beverage and vegetables are known to have considerable amounts of Ni that could significantly contributed to daily intake. With respect to Cu, calculated intake for brew of tea and herbs were 0.01 and 0.02, respectively against 0.04 and 0.04 $\mu g\ day^{-1}$ for tea and herb infusion, respectively (Table 4). These values were much lower than those (7.8 and 1.2 mg day^{-1}) set by UK and WHO (1998a, b).

Solubility of the studied metals (Table 4) that calculated as a ratio between brew and infusion extractable metal and total content, varied widely between tested metals and ranged from 0.0−48%. The obtained results show that toxic metals, Cd and Pb were insoluble (solubility = 0%) in both brew and infusion extracts. Solubility of Co in brew beverage was 0%, while in infusion was relatively low from 3.65-13.64%. Also, solubility of Ni varied widely between infusion and brew, whereas the solubility of Ni in brew was relatively low (3.15-18.67%), it was markedly high in infusion (42.91-48.4%), for tea and herbs, respectively. The solubility criterions of the other four metals were intermediate and could be arranged in ascending order as follows:

Fe (8.13-19.66%) < Cu (8.47-38.01%) < Mn (20.63-37.55)< Zn (22.5-41.15%).

These results show that only small part of heavy metal content of both tea and herb leaves may brought into beverage, while the higher contents remain insoluble in the solid particles during beverage preparation. Thereby, much care should be paid to how to get red of the residual particles to avoid wide spreading of these metals in the environment.

All tea and herb brands available in markets contain considerable contents of the studied heavy metals. Fortunately, the concentrations of toxic metals i.e., Pb and Cd, in tea and herb beverage prepared using their leaves are too low. On the other hand, the considerable concentrations recorded for other non-toxic metals lie within the acceptable daily intake from air, food and water of these elements.

Due to the lack of information specifying the acceptable contents of heavy metals in tea leave, the maximum allowable and safe concentration of each metal in tea leaves is urgently needed. Occasional check and frequent analyze foodstuff is also with intention to avoid many risk that arise from intake beyond the tolerance limits standards.

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