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DETERMINATION OF MULTI ELEMENT LEVELS IN LEAVES AND HERBAL TEAS FROM TURKEY BY ICP-OES

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ABSTRACT. We have determined the amount of some heavy metals (Al, Ba, Cr, Cu, Fe, Mn, Ni, Sr and Zn) contained in 18 herbal tea samples that had been bought from local markets where different parts of plants (leaves or flowers) are consumed especially for medical purposes in Turkey by ICP-OES. The validity of the analytical procedure was checked by analyzing INCT-TL-1 CRM certified reference material. The results from the analysis of CRM were all within the 95% reliability limit. The experiment results comply with the values reported in the literature. The wet decomposition method was used to solubilize the metals. The amount of the heavy metal that had passed to the tea via infusion has also been determined. The lowest and highest amounts of the metals found are: Al 268.0±0.6 and 2164.0±0.6 µg/g, Ba 3.0±0.1 and 101.0±0.6 µg/g, Cr 2.0±0.1 and 8.0±0.1 µg/g, Cu 5.0±0.2 and 64.0±0.8 µg/g, Fe 18.0±0.1 and 1471.0±0.3 µg/g, Mn 121.0±0.4 and 657.0±0.7 µg/g, Ni 2.0±0.1 and 17.0±0.3 µg/g, Sr 6.0±0.2 and 82.0±0.2 µg/g, Zn 15.0±0.1 and 103.0±0.4 µg/g. Cd, Pb and Co were not detected in the tea samples studied.

KEY WORDS: Herbal tea, ICP-OES, Multi element, Turkey

INTRODUCTION

Tea is one of the most popular beverages throughout the world [1]. Teas are classified into three major categories according to the degree of fermentation: unfermented green teas, partially fermented oolong and paochong teas, and fully fermented black and pu-erh teas [2]. Drying and roasting the leaves produces green tea; black tea is obtained after a fermentation process [3].

Herbal tea has been imbibed for as long as written history exists. Herbal tea is an infusion made from anything other than the leaves of the *Camellia sinensis*; typically, it is the combination of boiling water and dried fruits, flowers or herbs [4]. Herbal teas can be made with both green and dried flowers, leaves, seeds or roots by pouring boiling water onto the plant parts and letting them steep for a few minutes. Seeds and roots can also be boiled on a stove. Herbal teas are generally consumed for their physical or medicinal effects, especially for their stimulant, relaxant or calming features [5, 6].

In addition to modern medicine for the treatment of diseases, there is a great demand for herbal drugs. Different parts of the plants are consumed as tea with the help of a solution. Infusion and decoction methods are being used to prepare herbal teas. Several plants can also be used as herbal tea [7].

Elements that plants need to survive are called "plant nutrients". In the analysis of plant tissues, it is possible to see almost all the elements found in nature. Although the plants are selective about taking nutrient ions, as the rate of nutritious form of nutrient elements found in the growth medium increases, some heavy metals that can pass into the body of plants by passive means can enter involved in the food chain [8]. As a result, this can toxically affect plants and the humans and animals who feed on these plants. Plants absorb the elements found

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in the growth medium whether they need them or not. However 16 of these elements (C, H, O, N, P, K, S, Ca, Mg, Fe, Zn, Mn, Cu, B, Cr, Mo) are essential nutrients for all plants. Another 6 elements (Co, Al, Na, Si, Ni and V) are useful elements that are known to be necessary for only some of the plants or processes [9-13].

Al, Cu, Fe, Mn, Sr and Zn are multi-elements which have major significance for human health [14]. Especially in recent years, teas derived from plant leaves or flowers are consumed a great deal in alternative medicine treatments. There are studies on the popular teas such as black tea, green tea and chamomile tea [2, 3, 5, 7]. However, studies like ours that handle these plants are rare. For this reason, in our study we decided to examine the heavy metals in herbal teas that are consumed extensively in Turkey, about which there is very little literature.

In this study, 18 different popular herbal teas commercially available in the local markets in Turkey were used. The determination of multi elements in the samples was performed using ICP-OES.

EXPERIMENTAL

Apparatus

ICP-OES (Spectro Analytical Instruments, Kleve, Germany) was used for the determination of elements. The operating parameters of ICP-OES were set as recommended by the manufacturer. The ICP-OES operating conditions are given in Table 1.

Table 1. The operating parameters of determination of elements by ICP-OES.

Instrumental parameter	Value
Viewing height (mm)	12
Wavelength	nm
RF Power (W)	1450
Spray chamber	Cyclonic
Nebulizer	Modified Lichte
Nebulizer flow (L/min)	0.8
Plasma torch	Quartz, fixed, 3.0 mm injector tube
Plasma gas flow (L/min)	13
Auxiliary gas flow (mL/min)	0.7
Sample aspiration rate (mL/min)	2.0
Sample pump rate (rpm)	25

Chemicals

All glassware and polyethylene bottles were cleaned by soaking overnight in 10% HNO_3 and 65% perchloric acid, $HClO_4$ (E. Merck, Darmstadt, Germany) and rinsed five times with distilled water before use. These solutions were diluted carefully to the necessary concentrations with ultra-distilled water (Milli-Q Millipore 18.2 MΩ.cm). All the chemicals used during the experiments were of analytical-reagent grade (E. Merck, Darmstadt, Germany).

Samples

In this study, 18 different herbal tea samples (leaves and flowers) that are traditionally used for medical purposes across Turkey were collected in open packs of 50 g from local markets.

Wet decomposition method

Eighteen processed herbal tea samples that were obtained from the local stores were classified and washed with purified water. The samples were then dried in an oven for 4 h at 105 °C. A sample of 0.25 g of each extract was digested in a 10 mL mixture of nitric acid and perchloric acid (7:3 v/v) in a polyethylene plugged bottle. This mixture was heated at 130 °C for 1 h until the sample dissolved. The samples were filtered with black filter paper and then the volume adjusted up to 25 mL with deionized water. The samples were then kept in a refrigerator at 4 °C until analysis. The different digests of tea samples were analysed for heavy metal content using ICP-OES. All the standard solutions (1000 mg/L) for Al, Ba, Cr, Cu, Fe, Mn, Ni, Sr and Zn were spectroscopic grade and obtained from Merck (Darmstadt, Germany). These solutions were diluted carefully to the required concentrations with UHQ water. The ranges of the calibration curves (6 points) were selected to match the expected concentrations (10-320 ng/mL) for all the elements.

Infusion methods

Herbal tea (5.0 g) was added to 200 mL of boiling distilled water and allowed to infuse for 2 min [15]. After filtering under vacuum, the solution was collected as a first infusion. The same tea samples were used for making a second and then a third infusion, using 200 mL of boiling distilled water each time and allowing infusing for a further 5 and 10 min, respectively. Liquors from the first, second and third infusions were analysed for Al, Ba, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Sr, Zn content by ICP-OES.

Quality control

The quality of the analytical procedures was checked using Polish Certified Reference Material Tea Leaves (INCT-TL-1). Each part of the (sample of dry powder) prepared for the study of recovery and analytical reproducibility was assessed using sample duplicates, blanks and certified standards. The analysis of certified reference material (CRM) allowed an evaluation of accuracy and precision over a wide range of element concentrations. The results from the analysis of CRM were all within the 95% reliability limit. The results are given in Table 2.

Table 2. Certificate for the values of the standard reference material and the results of tea leaves (INCY-TL-1).

Element	Certified values (µg/g)	Determined values (µg/g)	Recovery (%)
Al (%)	0.23±0.03	0.20±0.01	86.03
Ba	43.2±3.9	43.5±2.4	100.7
Cr	1.9±0.2	1.7±0.1	86.39
Cu	20.4±1.5	20.0±1.1	98.04
Fe	432	460	106.5
Mn (%)	0.16±0.01	0.142±0.081	90.45
Ni	6.1±0.5	6.0±0.4	98.04
Sr	21 ±1.7	21.0±1.4	100.9
Zn	35±2.7	34±2.3	98.27

RESULTS AND DISCUSSION

The accumulation of metals, whatever the reason may be, is one of the most important environmental problems [16]. Most of the metals accumulate in the biological environment. The pollution is then accumulated within the body of organisms. These metals are toxic materials when they are in excessive doses [17]. The heavy metals (Al, Ba, Cr, Cu, Fe, Mn, Ni, Sr and Zn) content in 18 herbal teas (some consumed as leaves or as flowers) purchased from local markets were determined by ICP-OES. The results are given in Table 3. Co, Cd and Pb were not detectable.

Table 3.Trace element concentrations in herbal tea samples (mean + RSD %).

Samples	Element concentrations (µg/g) in herbal tea sample								
Tea leaves	Al	Ba	Cr	Cu	Fe	Mn	Ni	Sr	Zn
Olea europaea	488.0±0.7	8.0±0.3	-	32.0±0.6	520.0±0.5	361.0±0.3	-	35.0±0.5	18.0±0.2
Laurus nobiles	696.0±0.7	3.0±0.1	-	36.0±0.5	352.0±0.2	367.0±0.5	-	10.0±0.4	24.0±0.3
Crataegus	1232.0±0.6	67.0±0.4	8.0±0.1	31.0±0.5	368.0±0.3	302.0±0.4	2.0±0.1	45.0±0.1	27.0±0.7
Myrtus communis L.	464.0±0.5	11.0±0.2	-	43.0±0.4	185.0±0.4	297.0±0.9	-	23.0±0.3	14.0±0.3
Calluna vulgaris	888.0±0.4	25.0±0.6	-	30.0±0.3	194.0±0.2	328.0±0.5	-	6.0±0.2	24.0±0.4
Viscum album L.	2164.0±0.6	8.0±0.2	3.0±0.1	34.0±0.2	752.0±0.4	277.0±0.5	-	57.0±0.8	25.0±0.6
Flowers	Al	Ba	Cr	Cu	Fe	Mn	Ni	Sr	Zn
Achillea millefolium	442.0±0.5	4.0±0.1	-	13.0±0.3	100.0±0.5	-	3.0±0.4	3.03±0.1	15.0±0.1
Marsdeniaerecta	1160.0±0.7	5.0±0.4	2.0±0.1	41.0±0.5	870.0±0.6	605.0±0.5	17.0±0.3	59.0±0.2	34.0±0.2
Cichorium intybus	1180.0±0.3	30.0±0.7	-	35.0±0.3	566.0±0.7	399.0±0.7	-	82.0±0.2	103.0±0.4
Althaea officinals	956.0±0.6	26.0±0.6	2.0±0.1	22.0±0.2	952.0±0.3	-	4.0±0.3	28.0±0.5	22.0±0.3
Plantago	4064.0±0.8	20.0±0.5	2.0±0.1	42.0±0.4	1223.0±0.2	657.0±0.7	-	31.0±0.6	38.0±0.8
Alchemilla vulgaris L.	764.0±0.6	-	-	15.0±0.1	260.0±0.5	169.0±0.4	-	38.0±0.2	20.0±0.4
Veronica officinals	564.0±0.6	16.0±0.9	5.0±0.3	11.0±0.3	1471.0±0.3	450.0±0.8	3.0±0.1	22.0±0.6	18.0±0.1
Pimpirella anisum L.	940.0±0.4	10.0±0.3	2.0±0.1	5.0±0.2	227.0±0.3	135.0±0.2	4.0±0.1	44.0±0.2	5.0±0.2
Hibuscus sabdariffa L.	328.0±0.6	101.0±0.6	-	40.0±0.6	172.0±0.4	268.0±0.7	2.0±0.1	62.0±0.6	41.0±0.4
Lamium L.	412.0±0.3	12.0±0.2	-	64.0±0.8	190.0±0.4	154.0±0.3	-	10.0±0.1	53.0±0.4
Malva vulgaris	456.0±0.5	8.0±0.3	-	14.0±0.2	278.0±0.5	318.0±0.5	-	51.0±0.5	33.0±0.2
Famaria officinals	268.0±0.6	19.0±0.4	-	17.0±0.1	18.0±0.1	121.0±0.4	-	53.0±0.7	40.0±0.7

- Below detection limit (BDL).

The whole data was subjected to a statistical analysis and correlation matrices were produced to examine the inter-relationships between the trace metal concentrations. Students' t-test was employed to predict the importance of t-test values used in this study ($p < 0.05$).

These results demonstrated that the solubility of metals in the second infusion was considerably higher than that which was seen in the 5 min infusion. The infusion that was seen in 2 min was more than that seen in 5 min. The solubility of metals in the 5 min infusions was also significantly higher than in the 10 min. The experimental results are in agreement with the values reported in the literature [2, 7, 18]. The concentration of elements in tea infusions is given in Table 4.

Al was found to be the highest in herbal tea *Plantago* (4064.0±0.8 µg/g) and the lowest in herbal tea *Famaria officinals* (268.0±0.6 µg/g). Ba was found to be the highest in *Hibuscus sabdariffa* L. (101.0±0.6 µg/g) and the lowest in *Alchillea millefolium* (BDL). Cr was found to be the highest in herbal tea *Crataegus* (8.0±0.1 µg/g) and the lowest in herbal tea; *Olea europaea*, *Laurus nobiles*, *Myrtus communis* L., *Calluna vulgaris*, *Achillea millefolium*, *Cichorium intybus*, *Alchemilla vulgaris* L., *Hibuscus* s*abdariffa* L., *Lamium* L., *Malva vulgaris*, were non-detectable. Cu was the highest in herbal tea *Lamium* L. (64.0±0.8 µg/g) and the lowest in herbal tea *Pimpirella anisum* L. (5.0±0.2 µg/g). Fe was the highest in herbal tea *Verenico officinals* and the lowest in herbal tea *Famaria officinals.* Mn was the highest in herbal tea *Plantago* and the lowest in herbal tea *Achillea millefolium* and *Althaea officinals* (BDL). Ni was the highest in herbal tea *Marsdenia erecta* (17.0±0.3 µg/g) and lowest in herbal tea *Olea europaea*, *Laurus nobiles*, *Myrtu scommunis* L., *Calluna vulgaris*, *Viscum album* L., *Cichorium intybus*, *Plantago*, *Alchemilla vulgaris* L., *Lamium* L., *Malva vulgaris* and *Famaria officinals* were non-detectable. Sr was the highest in herbal tea *Cichorium intybus* (82.0±0.2 µg/g) and the lowest in herbal tea *Calluna vulgaris* (6.0±0.2 µg/g). Zn was the highest in herbal tea

Cichorium intybus (103.0±0.4 μg/g) and the lowest in herbal tea *Pimpirella anisum* L. (5.0±0.2 μg/g).

Table 4A. Concentration of elements (μg/g) in tea leaves infusions at 2, 5 and 10 min.

Element	Infusion time	Herbal tea sample (Tea leaves)					
		Olea europaea	*Laurus nobiles*	*Crataegus*	*Myrtu scommunis* L.	*Calluna vulgaris*	*Viscum album* L.
Al	2 min	232.0±0.1	332.0±0.2	676.0±0.4	242.0±0.4	564.0±0.6	589.0±0.5
	5 min	154.0±0.6	284.0±0.4	524.0±0.6	85.0±0.6	245.0±0..3	472.0±0.7
	10 min	97.0±0.5	200.0±0.8	452.0±0.6	74.0±0.5	132.0±0.4	323.0±0.4
Ba	2 min	5.0±0.2	3.0±0.2	12.0±0.5	6.0±0.3	14.0±0.4	7.0±0.2
	5 min	4.0±0.3	2.0±0.1	10.0±0.5	2.0±0.1	12.0±0.2	5.0±0.3
	10 min	2.0±0.1	1.0±0.1	7.0±0.4	1.0±0.1	6.0±0.2	4.0±0.3
Cu	2 min	5.0±0.4	4.0±0.3	8.0±0.2	7.0±0.1	18.0±0.3	5.0±0.2
	5 min	3.0±0.1	3.0±0.2	5.0±0.3	5.0±0.1	15.0±0.2	3.0±0.1
	10 min	2.0±0.1	2.0±0.1	2.0±0.1	2.0±0.1	4.0±0.3	2.0±0.2
Fe	2 min	6.0±0.5	32.0±0.3	9.0±0.5	5.0±0.3	132.0±0.6	165.0±0.4
	5 min	5.0±0.3	30.0±0.7	5.0±0.3	4.0±0.3	58.0±0.6	43.0±0.2
	10 min	2.0±0.1	28.0±0.2	2.0±0.1	2.0±0.1	14.0±0.1	38.0±0.2
Mn	2 min	54.0±0.4	45.0±0.1	50.0±0.2	4.0±0.2	154.0±0.2	148.0±0.8
	5 min	23.0±0.2	25.0±0.5	45.0±0.3	2.0±0.1	86.0±0.6	85.0±0.5
	10 min	16.0±0.3	17.0±0.1	21.0±0.2	1.0±0.1	24.0±0.3	54.0±0.3
Sr	2 min	30.0±0.3	8.0±0.2	8.0±0.3	8.0±0.2	5.0±0.3	27.0±0.1
	5 min	24.0±0.4	7.0±0.3	7.0±0.1	5.0±0.2	4.0±0.3	17.0±0.7
	10 min	15.0±0.5	4.0±0.2	6.0±0.2	2.0±0.1	2.0±0.2	13.0±0..3
Zn	2 min	17.0±0.8	6.0±0.2	9.0±0.3	5.0±0.3	15.0±0.5	17.0±0.7
	5 min	14.0±0.2	5.0±0.3	5.0±0.3	2.0±0.1	12.0±0.2	13.0±0.3
	10 min	5.0±0.4	2.0±0.1	4.0±0.3	1.0±0.1	3.0±0.2	12.0±0.4

Table 4B. Concentration of elements (μg/g) in tea flowers infusions at 2, 5 and 10 min.

Ele-ment	Infusion time	Herbal tea sample (Tea flowers)											
		Achillea millefolium	*Marsdenia erecta*	*Cichorium intybus*	*Althaea officinals*	*Plantago*	*Alchemilla vulgaris* L.	*Veronica officinals*	*Pimpirella anisum* L.	*Hibuscus sabdariffa* L.	*Lamium* L.	*Malva vulgaris*	*Famaria officinals*
Al	2 min	384.0±0.5	354.0±0.7	986.0±0.9	856.0±0.5	1000.0±0.5	325.0±0.5	356.0±0.6	856.0±0.6	235.0±0.5	69.0±0.4	285.0±0.5	184.0±0.4
	5 min	296.0±0.6	124.0±0.4	456.0±0.6	562.0±0.6	800.0±0.4	350.0±0.6	286.0±0.5	456.0±0.4	156.0±0.3	48.0±0.6	56.0±0.6	116.0±0.5
	10 min	208.0±0.7	56.0±0.6	231.0±0.3	256.0±0.5	231.0±0.1	276.0±0.7	232.0±0.7	321.0±0.1	89.0±0.6	36.0±0.4	24.0±0.4	160.0±0.2
Ba	2 min	4.0±0.4	3.0±0.2	20.0±0.7	16.0±0.5	13.0±0.5	23.0±0.3	12.0±0.4	8.0±0.5	96.0±0.7	4.0±0.3	8.0±0.3	13.0±0.2
	5 min	3.0±0.1	2.0±0.1	13.0±0.3	14.0±0.3	10.0±0.6	14.0±0.5	10.0±0.7	7.0±0.6	78.0±0.8	3.0±0.2	5.0±0.2	12.0±0.2
	10 min	2.0±0.1	2.0±0.1	7.0±0.1	5.0±0.3	8.0±0.5	10.0±0.3	8.0±0.5	4.0±0.3	56.0±0.1	2.0±0.1	4.0±0.2	10.0±0.8
Cu	2 min	3.0±0.2	24.0±0.4	23.0±0.3	15.0±0.5	32.0±0.2	13.0±0.3	8.0±0.1	3.0±0.2	30.0±0.3	7.0±0.5	8.0±0.2	3.0±0.2
	5 min	2.0±0.1	12.0±0.2	14.0±0.4	10.0±0.5	25.0±0.5	7.0±0.8	7.0±0.4	2.0±0.1	20.0±0.4	6.0±0.4	4.0±0.2	2.0±0.1
	10 min	2.0±0.1	6.0±0.3	10.0±0.6	3.0±0.2	14.0±0.4	5.0±0.6	4.0±0.2	1.0±0.1	10.0±0.3	2.0±0.1	3.0±0.1	2.0±0.1
Fe	2 min	70.0±0.7	23.0±0.3	456.0±0.5	568.0±0.6	869.0±0.7	22.0±0.2	856.0±0.5	123.0±0.7	100.0±0.4	56.0±0.4	256.0±0.3	10.0±0.2
	5 min	50.0±0.3	10.0±0.7	253.0±0.4	456.0±0.5	564.0±0.7	20.0±0.5	126.0±0.7	45.0±0.5	86.0±0.6	23.0±0.3	125.0±0.4	8.0±0.1
	10 min	20.0±0.1	5.0±0.3	124.0±0.5	323.0±0.1	235.0±0.2	18.0±0.5	27.0±0.4	23.0±0.3	45.0±0.5	6.0±0.4	70.0±0.2	6.0±0.3
Mn	2 min	232.0±0.6	352.0±0.5	256.0±0.8	5.0±0.2	356.0±0.6	100.0±0.8	356.0±0.6	100.0±0.6	245.0±01	100.0±0.4	236.0±0.2	100.0±0.6
	5 min	85.0±0.5	276.0±0.7	132.0±0.3	4.0±0.3	254.0±0.4	85.0±0.5	245.0±06	85.0±0.8	123.0±0.2	85.0±0.5	145.0±0.5	85.0±0.7
	10 min	24.0±0.6	132.0±0.2	45.0±0.5	3.0±0.1	123.0±0.3	24.0±0.4	123.0±0.6	25.0±0.6	81.0±0.1	56.0±0.6	52.0±0.2	52.0±0.3
Sr	2 min	10.0±0.3	40.0±0.2	75.0±0.2	24.0±0.4	25.0±0.5	10.0±0.3	12.0±0.2	36.0±0.8	56.0±0.6	7.0±0.3	40.0±0.4	48.0±0.2
	5 min	4.0±0.1	25.0±0.5	62.0±0.6	12.0±0.3	13.0±0.3	8.0±0.8	10.0±0.2	25.0±0.5	42.0±0.2	5.0±0.1	25.0±0.5	35.0±0.5
	10 min	3.0±0.1	21.0±0.1	13.0±0.2	7.0±0.4	10.0±0.2	6.0±0.1	8.0±0.4	12.0±0.4	12.0±0.2	3.0±0.2	21.0±0.2	10.0±0.2
Zn	2 min	13.0±0.1	9.0±0.3	90.0±0.4	18.0±0.1	25.0±0.3	18.0±0.3	8.0±0.2	2.0±0.1	35.0±0.5	14.0±0.4	25.0±0.3	28.0±0.8
	5 min	12.0±0.2	8.0±0.5	56.0±0.6	14.0±0.4	20.0±0.2	13.0±0.3	5.0±0.3	1.0±0.1	24.0±0.2	7.0±0.5	14.0±0.3	10.0±0.2
	10 min	10.0±0.6	4.0±0.2	32.0±0.5	7.0±0.4	12.0±0.1	10.0±0.4	2.0±0.1	1.0±0.1	13.0±0.3	3.0±0.2	8.0±0.3	8.0±0.7

Al occurs in several areas. It can get into the body in various ways and has gastrointestinal, hematologic, skeletal and nervous system effects [19]. It can produce encephalopathy in renal-impaired humans and cognitive defects in young children, a low-turnover bone illness, and a microcytic hypochromic anemia (UK Spelling) [20]. Controversially, it has been implicated as an environmental factor that may contribute to neurodegenerative diseases, including Alzheimer's disease [19].

Our study showed that Al in herbal tea leaf levels was found in the range of 464.0±0.5 and 2164.0±0.6 µg/g in plant material and 74 and 676 µg/g in the infusion and in flowers as 268.0±0.6 and 4064.0±0.8 µg/g in the digestion and 24 to 1000 µg/g in the infusion. Wong *et al.* has determined that Al concentration in various tea types growing in a tea plantation in Hong Kong was found to be 2500 to 13000 µg/g [21]. Recently, it has been demonstrated that young leaves of the Kenyan variety had a significantly high Al level of 2152 µg/g, and the young leaves of the Chinese diversity had the lowest Al of 381 µg/g when plants are grown in Lantau Peak located on Lantau Island in Hong Kong [22].

In the literature there is a very limited number of studies about Ba levels in herbal tea with reported range from 2 to 42 µg/g [23]. Our study on herbal tea leaves indicated Ba as 3.0±0.1 and 67.0±0.4 µg/g in the digestion and from 1.0±0.1 to 14.0±0.4 µg/g in the infusion. Whereas Ba in flowers were BDL and 101.0±0.6 µg/g in the digestion and from 2.0±0.1 to 96.0±0.7 µg/g in the infusion.

It is known that heavy metals such as Cu, proteins and enzymes (as catalytic and structural components) are necessary as cofactor for plants to grow up. These micro-nutrients and cadmium have a toxic effect on plants due to the large amount of heavy metals such as nickel and lead [24]. Therefore the Cu content in water, food and beverages must be traced and controlled on a daily basis [25]. Our study on the herbal tea leaves levels determined Cu as 30.0±0.3 and 43.0±0.4 µg/g in the digestion and from 3 to18 µg/g in the infusion whereas in flowers the results were 5.0±0.2 and 64.0±0.8 µg/g in the digestion and from 2 to 35 µg/g in the infusion. In the literature, Cu levels in tea samples consumed have been reported in the range of 15.7 and 34 µg/g [20]. The range of Cu concentration in the black tea samples has been reported as 24.07±2.25 µg/g [17] and a range from 10.089 and 30.178 µg/g in children's herbal tea.

Cr in tea may have not only useful but also opposite effects on human health [26]. Cr(III) plays a crucial role in human physiology by stimulating glucose metabolism, controlling blood cholesterol levels, stimulating the synthesis of protein, increasing resistance to pain and suppressing hunger pain [14, 26]. Our study on the herbal tea leaves determined Cr levels 3.0±0.1 and 8.0±0.1 µg/g whereas, in flowers were 2.0±0.1 and 5.0±0.3µg/g, reported values ranged from 0.33 to 2.43 µg/g Cr in herbal tea samples [27] and 0.45 to 0.99 µg/g Cr in green tea [28] and reported range from 2.95 to 7.6 µg/g in black tea samples from South India.

Fe can be widely found in nature and is one of the most important elements in environmental and biological systems as many kinds of compounds [29]. Fe is a necessary and useful element for organisms and a significant part of tissue and blood in animal and human bodies [8]. Fe is essentially distributed in hemachrome that occupies 60-70% of the total Fe in a body [29]. Our study on the herbal tea leaves Fe levels found to be 185.0±0.4 and 752.0±0.4 µg/g in the digestion and from 2.0±0.1 to 165.0±0.4 µg/g in the infusion and 18.0±0.1 and 1471.0±0.3 µg/g in the digestion and from 5.0±0.3 to 869.0±0.7 µg/g in the infusion. Fe is the second highest element in our study. Similarly, Fe concentration ranged from 119.2 to 368.85 µg/g in the digestion. Fe is the second highest element found in tea samples in the study of Gorur et al. (2001) [30].

In the literature Mn was found to be in the range of 300 and 1147 µg/g for tea samples. Mn is the only element found in significant amounts in tea, specifically in black tea which has reported levels between 730 and 880 µg/g [18, 31]. It has a reported range from 190±12 to 6376±756 in ten types of commercially available tea Manganese is an important co-factor for many enzymes and plays an essential role in the body's functions [26]. However, high doses of dietary manganese can be associated with long-term toxicity. Therefore, an estimated safe and sufficient daily dietary intake is 2-5 mg [18]. Our study on the herbal tea leaves found Mn levels in the range of 277.0±0.5 and 367.0±0.5 µg/g in the digestion and from 1.0±0.1 to 154±0.2 µg/g in the infusion and flowers were 121.0±0.4 and 657.0±0.7 µg/g in the digestion and from 4.0±0.3 to 356.0±0.6 µg/g in the infusion. Mn was found to be in the range of 300 and 1147 µg/g for tea samples. Mn is the only element with a significant amount in tea, specifically in black tea has reported between 730 and 880 µg/g [18, 31].

Although Ni is necessary for plants at low concentrations, it is toxic at higher concentrations [32]. Our study on the herbal tea leaves Ni levels were BDL and 2.0±0.1 µg/g whereas in flowers levels were 2.0±0.1 and 17.0±0.3 µg/g. Ni in tea was found in the range of 1.1 to 5.3 µg/g. The lowest Ni content was found in Nilgiris teas and the highest in tea from Vandiperiyar and Permedu [27,28] and the range of Ni concentration is reported in the samples as 2.9 and 22.6 µg/g. These results are similar to our study.

Sr is an element that can be found in substantial concentrations in nature, as a result of natural mineral degradation or consecutively on polluting anthropic activities [33]. The transfer of such a contaminant from the environment to plants is an important first step in the contamination of the food pathway. Sr is a naturally occurring mineral in the same mineral family as Ca and Mg; it has been shown to promote bone growth in both human and animal studies [34]. Our study on the herbal tea leaves Sr levels were found in the range of 6.0±0.2 and 57.0±0.8 µg/g in the digestion and 2.0±0.1 to 30.0±0.3 µg/g in the infusion and 10.0±0.1 and 82.0±0.2 µg/g in the digestion and 3.0±0.1 to 75.0±0.2 µg/g in the infusion. Sr was found between 14.4±0.3 and 74.2 ± 3.6 µg/g in the study that Leśniewicz *et al.* made in 2006 [8].

Zn is a necessary trace element for humans, animals and plants [35]. It is vital for many biological functions and plays a very important role in more than 300 enzymes in the human organism [17]. In the present study on the herbal tea leaves Zn levels were found to be 14.0±0.3 and 27.0±0.7 µg/g in the digestion and 1.0±0.1 to 17.0±0.8 µg/g and 5.0±0.2 and 103.0±0.4 µg/g in the digestion and 1.0±0.1 to 90.0±0.4 µg/g in the infusion. Görür *et al.* reported in 2011 Zn levels of 23.6 to 120.5 µg/g in the digestion and from 2.8 to 18 µg/g in the infusion of the Turkish tea market. Haban *et al.* have reported ranges from 36.6 to 71.7 µg/g Zn concentrations in children's herbal tea samples [17, 30].

CONCLUSIONS

While in many parts of the world tea is quite popular, nutritious and beneficial for humans, it is also important to monitor the concentration of heavy metals regularly as they could be possible reasons for serious health risks. The contamination of tea leaves by heavy metals may pose a serious threat to humans since the toxins are not biodegradable; they remain in nature and pass along the food chain. However, the absence of Cd and Pb in the tea leaves makes them consumable.

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