

Determination of Heavy Metal Contents and Some Basic Aspects of Widely Used Herbal Teas in Turkey

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Sage (Salvia aramiensis L.), rosemary (Rosmarinus officinalis L.), hibiscus (Hibiscus sabdariffa L.), linden (Tilia argentea Desf.), senna (Cassia angustifolia Vahl.), green tea (Camellia sinensis L.) and zahter (Thymbra spicata L.) are the top selling and locally most widely used herbal teas in Turkey. Proximate analyses and heavy metals (Al, Zn, Cu, Fe, Cd, Pb) contents were determined for seven herbal teas with 12 replicates (from different 12 sailors). The moisture content ranged between 5.49% (senna)–7.39% (hibiscus) while the total ash content were found between 4.86% (green tea)-10.54% (zahter). Crude fiber content ranged between 31.65-26.76% (rosemary-linden), acid soluble content found highest in hibiscus (1.02), lowest in linden (0.37) and water insoluble content ranged between 2.72-8.16% (green tea-senna). The highest heavy metals values in the experiment; for Al 1716.900 ppm (senna), for Cu 17.250 ppm (senna), for Zn 51.975 ppm (rosemary), for Fe 431.925 ppm (rosemary) were found. Cd was not determined in all samples. The highest average Pb content were obtained from sage with 4.350 ppm. Barely no Pb was found in the samples of green tea, hibiscus, linden and senna.

Keywords: heavy metals, medicinal plants, ICP-AES, ash content, crude fiber

Medicinal and aromatic plants are used for the prevention of diseases, sustainability of health and to cure diseases in traditional and modern medicine. At the same time, utilized as a nutritional supplements, spice, herbal tea and taste condiment in nutrition as well as in various fields of industry (food, perfumery, cosmetics, polishes, pesticides) [1].

Today, the number of medicinal and aromatic plants used in the world is around 20,000 according to the World Health Organization (WHO) and 4,000 of these drugs are still used widely in the World [2].

The heavy metals that involved and accumulate in the soil, can affect microbial activity, soil fertility, biodiversity and yield loss, even through the food chain can cause warm-blooded poisoning, lead to the emergence of many environmental and human health problems. The heavy metals also affect vegetative and generative organs of plants and the all living things maintain their active life in nature [3, 4].

The expansion of consumption of herbal plants has seen a significant increase in the use of herbal medicine due to their minimal side effects, availability and acceptability to the majority of the population, so medicinal plants play an important and vital role in traditional medicine and are widely consumed as a home remedies (It is therefore major interest to established the levels of some elements in common herbal plants because, at elevated levels, these elements can also be dangerous and toxic [5, 6]. Because of these concerns, many researchers study to detect moisture, ash, crude fiber and heavy metal contents of these plants. Their source is either edible wild plants or the plants sold in local markets [7-16].

Copper is an important element due to the incorporate the plant enzyme activation, play a part in carbohydrate and lipid metabolism [17]. Zinc directly effects the quantity and quality of the product due to the having an important

role in the protein and carbohydrate synthesis, enzyme activation, photosynthesis, respiration and biological membrane stability [18].

Cadmium involve and expansion in to the agricultural land is via industrial activities, phosphoric fertilizers, sewage and atmospheric deposits [19]. Including cadmium Majority of the plants and soil which include cadmium causes by precipitation of dust particles. Cadmium causes many physiological changes in plants due to the change of nitrogen and carbohydrate metabolism [20].

Lead is a common element in environment due to the widely usage in industrial and agricultural activities. Lead reach soils by the automobile industry, as an additive to the battery and gas and use of pesticides reach the ground with the use of pesticides [21].

It is important to have a good quality control for medicinal herbs in order to protect consumers from contamination. There are no standards for medical raw plant materials, which establish a permissible level of metals in such materials. WHO reports maximum permissible levels in raw materials for arsenic, cadmium and lead (1.0, 0.3 and 10 mg/kg, respectively) and European Pharmacope reports for mercury [8, 22, 23].

Turkey is one of the countries where the plants were collected from nature, sold in herbalists and local products heavily consumed. Therefore, the effect of environmental pollution on herbal tea sold extensively in the herbalists and hence the effects on human health are of great importance. Although importance of home remedies known, there are only a few literature studies addressing moisture, ash, crude fiber and heavy metal of herbs that are widely consumed as a tea.

In this scope, the present study not only examined the heavy metal content of the plants consumed as food but also discussed the total ash content, water insoluble ash, acid soluble ash, moisture and crude fiber of these plants.

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Gas flow rate (L/min)		
Ar	1.5	
N ₂	20	
Torch	Fused quartz (axial)	
Auto sampler	ASX 520	
Replicates	3	
Pump speed (rpm)	15	
Stabilization time (s)	15	
Uptake time (s)	15	
Elements monitored (wave length, nm)	Fe (259.940), Cd (228.802), Al (396.152),	Zn (213.857), Cu (324.754), Pb (405.781).

Table 1

ICP-AES OPERATING CONDITIONS FOR DETERMINATION OF SOME ELEMENTS IN THE HERBS AND THEIR INFUSIONS

Experimental part

Materials and methods

In this study, samples of rosemary (*Rosmarinus officinalis* L.), sage (*Salvia aramiensis* L.), mediterranean thyme (*Thymbra spicata* L.), senna (*Cassia angustifolia* Vahl.), green tea (*Camellia sinensis* L.), hibiscus (*Hibiscus sabdariffa* L.) and linden tea (*Tilia argentea* Desf.) samples supplied from local herbalists in Hatay. The most widely accepted and frequently consumed herbal tea were selected. The plants samples were analyzed to determine the moisture, ash, crude fiber and heavy metal contents.

Moisture content measurement of the plants was based on the principle of *removal of plant water at a certain temperature and detection of moisture content according to the resulting weight loss*. Approximately 5 g of plant material obtained from each specimen was transferred to metal cups and incubated for 7 hours at 105 °C. The specimens removed from the incubator were re-weighed at room temperature to determine the moisture content.

Total Ash Content of the Plant Samples (%) was based on the principle of *burning a certain amount of a specimen and converting it into ash*. 3 g of plant material obtained from each specimen was transferred to porcelain crucibles. Then, the tare of the porcelain crucible was weighed. The crucibles were placed in an ash furnace at 550-600 °C until the specimens turned into gray colour. The weight of the specimens that were removed from the furnace was re-measured at room temperature to determine the ash content.

Water Soluble Ash Content (%); 20 mL of distilled water (or at least equivalent purity water) is added in to total ash, heated until the boiling and filtered through filter paper. Warm distilled water (or an equivalent purity water) and the filtrate and washings are washed a total up to reach 60 mL. The filter paper and the contents are again transferred to the crucible. Carefully evaporate from the water on the steam bath and heated in a muffle furnace at 525 ± 25 °C, until it is free of carbon particles. Cool in a desiccator and weighed. Ash is heated again in the oven for 30 min, cooled and weighed. This process repeated until the difference between two consecutive repeats weighing less than 0.001 g.

Herbal Tea Samples	*ASA %	**WUA %	***TA %	Moisture %	Crude fiber %
Rosemary	0.65	5.83	7.53	5.98	31.65
Sage	0.53	5.15	7.42	6.34	28.02
Zahter	0.59	6.75	10.54	6.80	30.63
Senna	0.49	8.16	9.81	5.49	30.39
Green tea	0.35	2.72	4.86	6.18	29.49
Hibiscus	1.02	6.28	9.59	7.39	29.85
Linden	0.37	5.47	7.67	6.96	26.76

* ASA= Acid soluble ash, ** WSU= Water insoluble ash, *** TA= Total ash

Acid in-soluble (%); Total ash or water insoluble ash residue is used as a test sample. 25 mL of hydrochloric acid is added in to total ash, boiled in steam bath around 10 min and filtered through filter paper. Warm distilled water (or an equivalent purity water) and the filtrate and washings are washed. The filter paper and the contents are again transferred to the crucible. Carefully evaporate from the water on the steam bath and heated in a muffle furnace at 525 ± 25 °C, until it is free of carbon particles. Cool in a desiccator and weighed. Ash is heated again in the oven for 30 min, cooled and weighed. This process repeated until the difference between two consecutive repeats weighing less than 0.001 g.

Crude Fiber Content (%); Crude fiber analysis were exercised by using Ankom Fiber Analyzer (A200). 1 g dry sample was put into filter bag (Ankom F-57) and seal 0.5 cm distance from the opening. Bags placed in to the analyzer. Firstly 1900-2000 mL of 1.25 % sulfuric acid solution added and then heated 45 min. After heated 45 min samples cleaned out from the acid then 5 min twice washed with distilled water. After this step NaOH solution added and heated 45 min cleaned out, twice washed with distilled water. After the cleaning step filter bags remove from the machine, wringed out and filter bags placed in to the beaker and filled with acetone. 3-5 min later wringed out and after a while samples left in the oven for about 2-4 h at 105 °C. The material was transferred into crucible and muffle oven at 550°C for 2 h. After it, crucibles were placed in muffle oven for 12 h at 550°C, cooled in the desiccator and recorded the crucible weight.

Heavy metal concentrations were determined ICP-AES [24]. Agilent Technologies model inductively coupled plasma atomic emission spectrometry (ICP-AES) was used simultaneous multielement detection of Al, Cu, Zn, Fe, Cd and Pb. Instrument configuration and general experimental conditions for ICP-AES are given in table 1.

Results and discussions

In current study, moisture, acid soluble ash, water unsolvable ash, total ash content and crude fiber of the 7 herbal tea samples from 12 different local sellers were investigated. In table 2 results of ash, crude fiber and

Table 2
RESULTS OF ASA, WUA, TA, MOISTURE, CRUDE FIBER ANALYSIS

Elements	Sage	Rosemary	Hibiscus	Linden	Senna	Green Tea	Zahter
Aluminum	377.55	580.275	321.975	259.445	1716.9	245.7	95.625
Copper	9.675	8.475	11.55	5.645	17.25	6.463	14.4
Zinc	35.025	51.975	33.075	9.57	22.2	31.090	27.075
Iron	379.275	431.925	203.4	171.818	185.925	154.963	102.525
Cadmium	*ND	ND	ND	ND	ND	ND	ND
Lead	4.35	1.8	ND	ND	ND	ND	0.9

*ND = Not detected

moisture analysis have given table 3 Al, Cu, Zn, Fe, Cd and Pb contents of sage, rosemary, hibiscus, linden, senna, green tea and mediteranean thyme presented.

It was obtained that the moisture content was higher in hibiscus followed by linden and zahter (table 2). Total ash content was higher in zahter and senna. Water unsolvable ash content was higher in senna while significantly lower in green tea. Acid soluble ash content was higher in hibiscus (table 2). Crude fiber content was higher in rosemary and lower in linden (table 2). Moisture contents of linden (*Tilia platyphyllos*) and sage (*Salvia officinalis* L.) are found similar to some other studies while the total ash content was found significantly lower than current results [14].

Moisture content of tea is one of the most important quality factor. According to the Turkish Food Codex limit for the tea samples especially for sage, rosemary, and zahter maximum moisture content should be 10% [25, 26]. In this study moisture content of the herbal tea samples range lower than 8%.

The content of aluminum in herbal teas ranged from 95.625-1716.9 ppm. Aluminum concentration, compare to the other tea samples, significantly higher in senna and lower in zahter. Different aluminum levels are found for sage, linden and senna in different studies [8,14]

Copper and zinc are among physiological elements essential for the correct functioning of living organisms but, in high concentration, can be also harmful to organisms. Copper contents found higher in senna (17.25 ppm), zahter (14.4 ppm) and hibiscus (11.55 ppm). In the previous studies, copper content for linden, sage and senna found to be respectively 9.64, 35.8 and 3.92 ppm [8]. It was reported Cu content for *Valeriana officinalis* 57.4 ppm [10] and Cu contents of *Tilia platyphyllos* and *Salvia officinalis* 2.08 and 4.60 respectively [14]. Similar results were obtained for *Salvia officinalis* [27] and lower results were obtained for *Salvia aramiensis* in some other studies [28] which comparative to our Cu contents in *Salvia aramiensis*. Copper content in *Hibiscus cannabinus* L. was found to be respectively 65.39 ppm [29] while Cu content for *Hibiscus sabdariffa* was lower and found to be 11.55 ppm in this study. Copper content of *Thymbra spicata* was found to be 13.639 ppm [11] which is similar to the result of this study for zahter (14.4 ppm).

Concentration of zinc in the samples ranged between 51.975-9.57 ppm and found higher in rosemary and lower in linden. In the study zinc contents found higher in rosemary (51.975 ppm) and lower in linden tea samples (9.57 ppm). Zinc content of *S. aramiensis* with 35.025 ppm was found to be higher than the Zn content of *S. officinalis* and *S. aramiensis* [27, 28]. Zinc content of *Hibiscus cannabinus* L. was found to be 15.78 ppm [29] while the Zn content of hibiscus was found to be 33.075 ppm in this study. Zinc content of *T. spicata* with 27.075 was close to the previous result 36.7792 ppm [11]. Sage (*S. aramiensis*) zinc content was found to be 35.025 ppm and showed similarities reported Zn contents of *S. viridis*, *S. verticillata* L. subsp.

amasiaca, *S. sericeo-tomentosa* and *S. tomentosa* respectively 31.5548, 21.0104, 24.4992, 29.0816 ppm [11].

Iron contents of the samples were higher in rosemary (431.925 ppm) and sage (379.275 ppm) compare to the other tea samples. Fe content found in hibiscus 203.4 ppm, and similar results were obtained from some other studies on different hibiscus species [29]. Fe content of *T. spicata* was found 102.525 ppm, but it was reported that on Fe content for *T. spicata* 882.6 ppm. in some other studies [11]. Fe levels for *Salvia aramiensis* were found 379.275 ppm. Similar results were reported for Fe contents for *Salvia viridis*, *Salvia tomentosa* and *Salvia aramiensis* [8, 28]. Fe content for Linden tea samples in the present work we found 171.818 ppm. Different and similar results were obtained in some other studies [8].

Lead and cadmium are on the list of the most dangerous metals that cause acute and chronic environmental contamination. Cadmium was not detected in all herbal tea samples that were analyzed. The lead was found in sage (4.35 ppm), rosemary (1.8 ppm) and zahter (0.9 ppm) samples and did not detect in hibiscus, linden and senna tea samples. Pb level from sage samples was high but WHO mentions maximum permissible levels for lead which amount to 10 mg/kg [8,22].

It was reported that Pb (0.12 ppm) and Cd (0.02 ppm) were found some *Tilia platyphyllos* plants [14]. Heavy metal levels for some *Salvia* species and *Thymbra spicata* investigated in some other studies and Pb only detected in *Salvia verticillata* L. subsp. *amasiaca* with 0.3326 ppm and Cd contents were detected between 0.0389-1.023 ppm for *Salvia* species. Cd and Pb were not detected for *Thymbra spicata* [11]. Cd and Pb contents were also reported for *Hibiscus cannabinus* L. 1.26 and 2.55 ppm, respectively [29].

Conclusions

Medicinal and aromatic plants have widely traditional used in folks. Day by day interest in alternative medicines are increasing. The most important problem in the use of medicinal and aromatic plants is these plants are provided from the nature an uncontrolled manner. It was difficult to accurately comment on the obtained results, because of the lack of information and regulations about acceptable content of heavy metals in medicinal and aromatic herbs.

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Table 3
HEAVY METAL CONTENTS OF
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