

## LEVELS OF ESSENTIAL HEAVY METALS IN HERBAL TEA COLLECTED FROM SABAC

Kosana Popović<sup>1</sup>, MSc; Mirjana Antonijević Nikolić<sup>1</sup>, PhD; Đorđe Nikolić<sup>2</sup>

<sup>1</sup>Higher Medical and Business-technological School of Applied Studies, Šabac, Serbia, [kosanal@gmail.com](mailto:kosanal@gmail.com)

<sup>1</sup>Higher Medical and Business-technological School of Applied Studies, Šabac, Serbia, [nikanto@ptt.rs](mailto:nikanto@ptt.rs)

<sup>2</sup>HBIS GROUP Serbia Iron & Steel doo. Beograd, Šabac, Serbia

**Abstract:** The chemical components in tea have received great interest because they are related to health. In this study, the content of four essential heavy metals including Cu, Fe, Mn and Zn were determined by atomic absorption spectrometry on samples of herbal teas (*Matricariae flos*, *Thymy herba*, *Menthae piperitae folium* and *Betulae folium*) collected from Sabac market, Serbia. The sample preparation has been performed using dry digestion procedure and dissolving the ash the first in 6M HCl and then in 0.1 M HNO<sub>3</sub>. Herbal teas showed the concentration of the heavy metals Cu, Fe, Mn and Zn in the range: 11.0-13.4 mg/kg, 166.5-755.5 mg/kg, 43.5-561.0 mg/kg and 16.5-242.5 mg/kg, respectively. The level of copper in all samples was uniform. The highest content of Fe was in *Thymy herba*, while Mn and Zn were in *Betulae folium*. The herbal tea samples analysed contain essential heavy metals (Cu, Fe, Mn and Zn) and could contribute to the daily dietary requirements.

**Keywords:** heavy metals, essential elements, herbal tea, AAS.

### 1. INTRODUCTION

Medicinal plants have a long history of therapeutical use through out the world and still represent an important part of traditional medicine [1]. A report of World Health Organization (WHO) displayed that about 70–80% of the world population applies non-conventional medicine, mainly of herbal origin in their primary healthcare [2]. The use of herbal teas have spread in our country as a complementary way to treat and prevent illness.

Herbal teas have a complex chemical composition and chemical substances which they are contained can have a biological activity in humans [1]. Consequently, herbal teas represent sources of various organic and inorganic components that can affect on human health. Tea is a rich source of minerals and trace elements that are essential to human health [3, 4, 5] and drinking of tea could be an important source of some essential minerals [6].

Essential minerals, including the trace elements, are inorganic elements that have a physiological function within the body. These must be provided in the diet and vary from grams per day for the major minerals through milligrams to micrograms per day for the trace elements [7].

Minerals, important for humans, have been divided into the next groups: (1) essential or major: calcium, magnesium, phosphorus, sodium, potassium, chlorine; (2) essential in traces: iron, copper, chrome, zinc, iodine, selenium, fluorine, cobalt, manganese, molybdenum; (3) probably essential in traces: silicon, nickel, tin, vanadium and (4) non-essential: arsenic, mercury, lead, aluminum, boron, gold, silver, titanium, lithium, strontium, germanium, cadmium, bismuth, rubidium, bromine [8].

In report of World Health Organization, trace elements have been classified into three groups from the point of view of their nutritional importance in humans, in the following way: (1) essential elements; (2) elements which are probably essential; and (3) potentially toxic elements, some of which may nevertheless have some essential functions at low levels [9].

Herbal teas have both essential (Fe, Cu, Mn, Zn) and toxic elements (As, Cd, Hg, Pb). A negative influence on human health can have the disadvantage, as well as, increased concentrations of essential elements [1, 7, 10]. Therefore, determining the content of heavy metals in herbal teas is very important. The physiological effects of heavy metals in living systems strongly depend on their concentration [11]. Thus, measuring heavy metal concentrations is important not only from a nutritional point of view but also for the assessment of their quality and safety [10] and should be carefully controlled, especially when herbal teas are used in human medicine.

Herbal teas plants are good sources of mineral elements [12] and could be a valuable source of nutrient elements for the human diet [13]. In particular, because iron (Fe) zinc (Zn), manganese (Mn) and copper (Cu) are vital for numerous physiological functions, their intake is considered good for human health. Iron acts as a catalytic center for a broad

spectrum of metabolic functions [7]. Zinc affects as a catalyst, coactive, or structural unit for some enzymes and is a cofactor of metalloenzymes. Deficiency of zinc can obstruct normal growth and development, reproduction, and immune function [14]. Manganese is an essential element that is incorporated into a number of metalloenzymes (Mn-metalloenzymes) [15]. Copper acts as a co-factor in cuproenzymes, which are essential enzymes for normal functioning of the body [14].

Contents of heavy metals in herbal teas vary depending on various factors. Among these factors are geoclimatic conditions, geochemical characteristics of the soil, anthropogenic activities (e.g., chemical industries in the vicinity), plant species (some can selectively accumulate heavy elements) and the part of the plant used for herbal tea [1]. The distribution of the heavy metals among plant organs is selective and depended on the part of the plant, surface characteristics of the plant organ, and the element that was examined [16].

Due to the importance of essential heavy metals in herbal teas, a number of studies have been carried out to determine their levels by using atomic absorption spectrometry (AAS), inductively coupled plasma mass spectrometry (ICP-MS), inductively coupled plasma-atomic emission spectrometry (ICP-AES) [5, 12, 13, 17-21]. These techniques are highly sensitive spectroscopic techniques and generally require the destruction of the sample matrix to render a solution of the analyte ready for analysis.

Lately, in our country, there have been more and more health food stores where herbal drugs can be bought in bulk cargo, which can be used as herbal teas alone or by combining two or more to prepare herbal tea blends. Their use in the population is very popular. The aim of this study was to examine the level selected essential heavy metals (Fe, Zn, Mn and Cu) by using AAS in samples of the herbal teas in bulk cargo from health food stores in Sabac. Due to the great application this tea for medical purposes and they are related to health, determining the content of metals is very important for controlling the quality of these beverages.

## 2. EXPERIMENTAL PART

### 2.1. Sampling and sample preparation

A total of four herbal tea samples were collected from local health food stores in Sabac, Serbia. The samples were herbal teas sold in bulk: *Matricariae flos*, *Thymy herba*, *Menthae piperitae folium* and *Betulae folium*. The dried tea samples were ground using a grinder and passed through a 0.5 mm sieve. The powdered tea samples were stored at ambient temperature and used for the analysis of metals in herbal tea.

### 2.1. Analytical measurements

The contents of essential metals were determined by drying 3 g samples (measured on an analytical balance with accuracy to the fourth decimal) and then ashed at 450 °C under for 8 h, a gradual increase ( $\leq 50$  °C/h) in temperature. Five ml hydrochloric acid 6M (1:1/ v:v) was added and the solution was evaporated in water bath to dryness. The residue was dissolved in ~10.0 ml nitric acid ( $c = 0.1$  mol/dm<sup>3</sup>), filtered, washed with deionised water, and received in 50.0 cm<sup>3</sup> volumetric flasks [22, 23].

Essential heavy metals (Cu, Fe, Mn and Zn) concentrations were determined by atomic absorption (AAS) method, using a Perkin-Elmer spectrophotometer AAS-5100/PC. Blanks were treated in the same way as products.

## 3. RESULTS AND DISCUSSION

Medicinal plants are the raw material for many herbal formulations and popular supplements. The use of herbal medicines has been on the rise in recent years due to their low prices. The last few decades have witnessed a rapid development in the diet studies focused on the determination of trace elements, which reflect their role in human health and nutrition. Deficiency, excess or imbalance of trace element intake into human body may result in various diseases.

In our work the content of essential elements, Fe, Zn, Cu and Mn, was determined in the four investigated herbal teas by using Atomic absorption spectroscopy (AAS). Herbal tea samples were collected from the local health food markets in the territory of Sabac, Serbia. Analyzed samples were in bulk, without adequate evidence of their quality.

Figure 1 shows the essential trace elements in marketed herbal teas measured in this study. The content of studied metal is expressed in mg/kg of dry matter. All these elements are essential to humans for growth, metabolism, and hormone balancing. The highest concentration of metal in the herbal teas was that of iron. It ranged from 166.3 to 755.5 mg/kg. The content of Cu is the lowest of all the elements in the analyzed herbal teas (11-13.4 mg/kg).

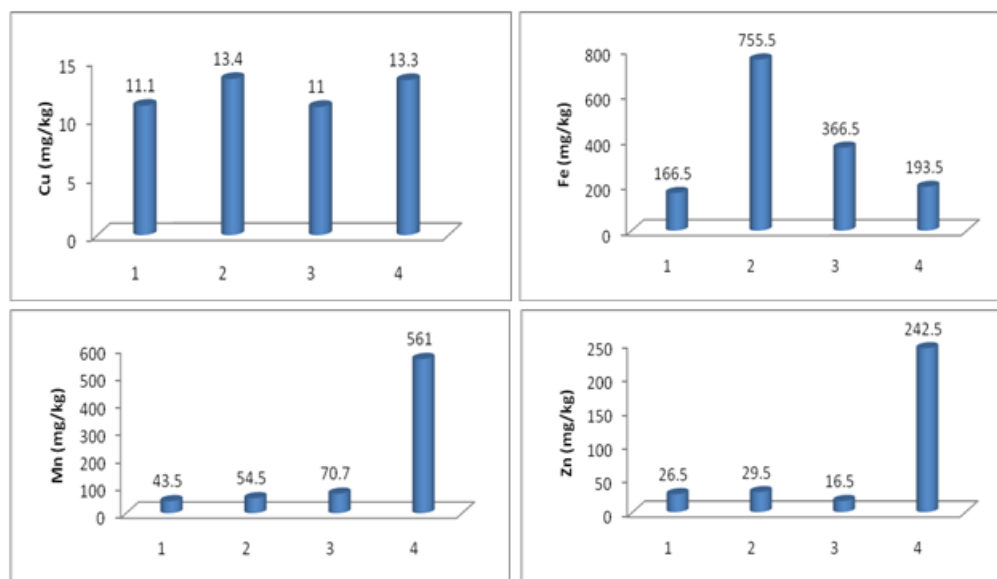
### 3.1. Iron

Iron is an essential element for growth of animals and plants. Its deficiency can hinder metabolism. The analyzed herbal tea samples contain large amounts of Fe from 166.5 to 755.5 mg/kg. The highest content is in the *Thymy herba* sample, 755.5 mg/kg. Iron is an essential element and for human growth and development, too and an essential component of

hemoglobin. It facilitates the oxidation of carbohydrates, proteins and fats to control body weight, which is a very important factor is diabetes management. Iron is necessary for the formation of hemoglobin and also plays an important role in oxygen and electron transfer in the human body. Low iron content causes gastrointestinal infection, nose bleeding and myocardial infection [24]. The WHO limit for iron in medicinal herbs has not been established yet. The results of the current study show a wide variation of iron in different herb samples. These levels are relatively high compared to iron in some medicinal plants for the treatment of anemia. A conducted study on selected medical herbs indicated iron concentrations within the range 6.67-223 mg/kg [25]. Based on these investigations and the total content of Fe in the analyzed herbal teas, it can be concluded that these herbal teas can contribute to the daily dietary requirements (15 mg/day (10-28 mg/day) Table 1) [15, 17]. Especially, it can be recommended herbal tea *Thymy herba* to use for increased daily intake Fe.

### 3.2. Zinc

The concentration of Zn in the analysed plant samples was between 26.5-242.5 mg/kg (Fig 1). The highest concentration was recorded in *Betulae folium* (242.5 mg/kg) while the remaining three sample contents are much smaller (16.5-29.5 mg/kg). Zinc is essential trace element necessary for proper growth, blood clotting, thyroid function and protein and DNA synthesis. Zinc intake beyond permissible limits produces toxic effects on the immune system of the body and disrupts copper levels [26]. The FAO/WHO permissible limit set for zinc in herbal medicines is 100 mg/kg [27]. The obtained results indicate a significantly higher content of Zn in the medical plant *Betulae folium* than the permissible limit. Whereas the recommended daily dietary intake of zinc stands at about 15 mg (Table 1) [15, 17].



1 = *Matricariae flos*, 2 = *Thymy herba*, 3 = *Menthae piperitae folium*, 4 = *Betulae folium*

**Figure 1.** Content of essential elements determined by AAS in the herbal tea samples, mg/kg dry weight.

### 3.3. Manganese

The content of Mn in herbal teas samples ranges from 43.5 to 561.0 mg/kg. A very high content was measured in the *Betulae folium* tea (561.0 mg/kg). Manganese (Mn) is a trace element necessary for plant, animal and human as enzyme cofactor [28]. However, the deficiency of manganese in humans may lead to immunodeficiency disorder, rheumatic arthritis in adults, disorder of bony cartilaginous growth in infants, as well as myocardial infarction and other cardiovascular diseases [29]. The adequate intake for adult men and women is 2.3 and 1.8 mg/day, respectively. A tolerable upper intake level of 11 mg/day was set for adults based on a no-observed-adverse-effect level [30]. The permissible level set by FAO/WHO in medicinal plants is 200 mg/kg [31]. In the present study, except for Mn levels of *Betulae folium*, Mn concentration levels in the studied herbal teas herbs were below the permissible limit set by FAO/WHO in medicinal plants. The leaves of *Betulae folium* contain higher levels than the permissible limits set for Mn in plants. Hence, they contain unsafe levels of Mn and might be detrimental to consumers' health.

### 3.4. Copper

The level of copper in all samples was uniform and in the range 11.0-13.4 mg/kg. Copper is an essential enzymatic element and is necessary for normal biological activities of amino oxides and tyrosinase enzyme. Tyrosinase is the

enzyme that is required for catalytic conversion of tyrosine to melanin, which is a vital pigment located beneath the skin, and thus protects the skin from dangerous radiations [32]. Copper is essential to the human body since it forms a component of many enzyme systems, such as cytochrome oxidase, lysyl oxidase and ceruloplasmin, an iron-oxidizing enzyme in blood. Copper deficiency results in anemia and congenital inability to excrete copper resulting in Wilson’s disease. However, copper could be toxic depending on the dose and duration of exposure [33, 34]. The regulatory limits of the WHO/FAO have not been established yet for the copper in herbal medicines. The recommended daily intake for adults of Cu is 2-3 mg/day [15, 17, 30].

Contents of microelements in medicinal plants are influenced by genetically-determined properties of a plant as well as by external factors, including geographic location, soil type and profile, fertilization, availability of water, pollution by pesticides or dusts, and gases. The differences can be a consequence the specific methods of the production of certain sorts of tea, including type, age of the herbal material, season of harvesting and manner of maturing and storing [21, 35].

**Table 1.** Contents of heavy metals (mg/kg) in herbal tea (in the original package) from Serbia described from the literature and recommended daily intakes (mg/day).

Tea	Concentration, mg/kg			
	Cu	Fe	Mn	Zn
<i>Matricariae flos</i> [36]	14.25	130.26	76.14	34.67
<i>Thymy herba</i> [36]	8.94	445.78	127.06	44.26
<i>Mentha x piperita</i> [36]	17.15	443.90	111.97	26.86
Recommended daily intakes [15, 17]	mg/day			
	2.5 (2-3)	15 (10-28)	2.8 (1.8-5)	15 (8-18)

Mihaljev et al. are analyzed by AAS the same or related herbal tea samples in the original package collected from the retail shops on the territory of Novi Sad, Serbia, Table 1 [36]. For species *Matricariae flos*, *Thymy herba*, and *Menthae piperitae folium*, comparison the content of certain elements, good agreement is observed. This may indicate that these three types of herbal tea are of approximate quality in terms of certain essential elements, regardless of the way they are placed on the market (original package with declaration or bulk).

The essential heavy metals concentrations in the herbal teas were given for 1 kg of the corresponding herb. These herbal teas are generally drunk as infusions in a quantity of 150 ml per cup of tea, usually three times during the day, and 2-3 g of herbal drug are used to prepare one cup of tea [37]. The transfer of essential heavy metals from the herbal drug to infusion according to literal data ranges from 0 to 88,86 % [38]. Taking into account these facts, herbal teas can contribute to daily intake of essential elements, without fear that their daily intake will be exceeded. This particularly refers to manganese, which has an increased level in the test sample of *Betulae folium*.

#### 4. CONCLUSIONS

The analysis of chosen herbal tea (*Matricariae flos*, *Thymy herba*, *Menthae piperitae folium* and *Betulae folium*) from Sabac, Serbia showed the presence of essential elements: Fe, Cu, Zn and Mn. The iron concentration in the plants was rather high and varied from 166.3 to 755.5 mg/kg. Content Mn and Zn in *Betulae folium* was high 561 and 242.5 mg/kg, respectively. The level of copper in all samples was uniform and in range 11.0-13.4 mg/kg.

The herbal tea samples analysed contain essential heavy metals (Fe, Cu, Zn and Mn) and could contribute to the daily dietary requirements.

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