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Original article

Determination of arsenic content in tea samples available on Republic of Srpska market by atomic absorption spectrophotometry

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Summary

Introduction. Arsenic exists in various forms in nature and living organisms. Toxic elements, including arsenic, which are present in some plants, can severely damage haemopoietic, immune, nervous and reproductive systems. For this reason, a content of heavy metals is one of the criteria for the assessment of the safe use of plant material in the production of traditional medicines and herbal infusions. This instigates the need for constant and organized safety control of plants that are used as raw materials in pharmaceutical industry.

The aim of this study is to determine the arsenic content in selected teas which are available on the market of the Republic of Srpska.

Methods. The 10 g samples of 13 herbal and 3 fruit teas were mineralized by dry ashing and arsenic contents were determined by the atomic absorption spectrophotometer Agilent Technologies Series 200 with an air-acetylene burner and D2 background correction.

Results. Mean arsenic concentrations in the herbal tea samples ranged from 0.009 to 0.145 mg/kg. The lowest arsenic concentration in a single sample of 0.007 mg/kg was found in Chamomile tea and Uva ursi collected as a wild plant at elevation above 1200 m. The highest arsenic concentration was found in the sample of Sambucus nigra tea (0.145 mg/kg). In fruit teas, the arsenic concentration ranged from 0.014 mg/kg (Cranberry) to 0.027 mg/kg (Fruit mix).

Conclusion. Arsenic content in all analyzed tea samples is below the value stipulated by the national legislation.

Keywords: arsenic, herbal tea, fruit tea, atomic absorption spectroscopy

Introduction

Metals can be considered as one of the oldest known poisons. In the periodic table of the elements, there are about 80 metal elements among which about 30 elements induce toxic effects in humans. In the past, metal toxicology mostly followed effects of accute exposure, but today, concentration of metals and other poisons is intensively monitored in living and working environment. If values are higher than the permitted ones, measures are undertaken to reduce health risks of persons living or staying for longer periods of time in these areas. [1]. The term „heavy metal" refers to any metal chemical complex with a relatively high density, i. e. greater than 5 g/cm^3, which is toxic or poisonous in low concentrations [2].

Primarily, plants absorb heavy metals through the root, from the soil, and, to a lesser extent, through over ground organs from the atmosphere. Atmospheric heavy metals can be deposited on plant surfaces, especially leaf surfaces, and mostly on surface soil. In this way, metals enter plants, and upon their consumption, they enter humans and animals. In such a way, they are introduced to the food chain. Under normal conditions, there are three ways of how metals can come into the body: through the skin, gastro-intestinal tract and respiratory tract. The International Agency for Research on Cancer classifies arsenic into the group of compounds for which there is enough evidence of carcinogenicity in humans. Arsenic belongs to the group A carcinogenic. Cases of skin cancer in people exposed to arsenic compounds were described in the nineteenth century. All types of skin cancer can appear, i. e. basal cell carcinoma and squamous cell carcinoma, which can develop into local and distal metastases [1, 2].

In nature, arsenic can be found in organic and inorganic compounds and arsine (AsH3) is also a significant compound. Main inorganic arsenic compounds are arsenic trioxide, sodium arsenite, and arsenic trichloride. Inorganic compounds of pentavalent arsenic are arsenic pentoxide, arsenic acid, and lead and calcium arsenate. Organic compounds are considered harmless for their poor cell absorption. Inorganic arsenic compounds are highly reactive, influence a series of intracellular reactions, and are used as pesticides [1]. Pollution of the environment by arsenic also occurs as a result of natural phenomena such as volcanic eruptions and fires. The application of intensive agro-technical measures in modern agricultural production, proximity to industrial plants, mines, roads, inevitably leads to contamination of soil and plants with pesticides and heavy metals (arsenic, lead).

Consumption of food grown on arsenic-contaminated areas is one of the sources of chronic arsenic exposure. Arsenic compounds are found not only in food of plant origin such as tea, but also in food of animal origin such as fish and shells, which contain organic arsenic compounds. Food can be an important way of arsenic exposure and such an exposure might have long-term negative effects on people. Symptoms of arsenic poisoning are fever, vomiting, diarrhea, difficulty breathing and swallowing, enlarged liver, cyanosis, arrhythmia, coma, oliguria, and tubular necrosis [2].

Arsenic intoxication is detected by the determination of its content in blood, urine, hair and nails. Bearing in mind short-term retention in blood (half-retention is four days), blood determination might be useful only few days after poisoning. Arsenic concentration in urine is significant in case of chronic poisoning. It remains in hair and nails for years after poisoning [1].

Plants represent direct or indirect sources of minerals in human nutrition. Plant species for production of mono-component tea or tea blends, which have been applied in traditional medicine, are of special significance. Plantation and self-seeding plants serve as a source of medicinal plant raw materials [3]. Its large production and consumption indicate that this drink is very popular in our region, whether black, green or herbal [4]. Tea (Camellia sinensis) is the most popular beverage in the world after water. Due to acidophilic nature of tea plant, it has an inherent tendency to uptake metals/metalloids including the toxic ones from the soil which is of great concern worldwide [5].

The objective of the study was to determine arsenic content in the samples of various herbal and fruit teas taken from pharmacies in order to gain some insight into safety of these products, bearing in mind their extensive use.

Methods

Arsenic content was examined in tea preparations which can be found in the Republic of Srpska and are available in pharmacies. Tested samples covered 16 different tea products (13 herbal teas and 3 fruit teas) from 10 different tea producers. Green tea, black tea, uva ursi, thyme, urtica dioica, hypericum perforatum, anise, sambucus nigra, milfoil, chamomile, mint, and rose hip were tested. The presence and content of arsenic were determined by the atomic absorption spectroscopy method, on the atomic absorption spectrometer Agilent 200 Series AA Systems, using the hydride technique. The sample is ho-

Table 1. The names of medicinal plants and plant parts used in the study [4,6]

Commercial name	Latin name	English name	Part of the plant
Zeleni[R]	Tea siensis	Green	folium
Crni[R]	Tea siensis	Black	folium
Kamilica[R]	Matricaria chamomilla	Chamomile	flos
Nana[R]	Mentha piperita	Mint	folium
Majčina dušica[R]	Thymus serpyllum	Thyme	folium, flos and stems
Anis[R]	Anis	Anisi	fructus
Zova[R]	Sambuci flos	Sambucus nigra	flos
Uva[R]	Uvae ursi	Uva ursi	folium
Sljez[R]	Althaea officinalis	Althea	radix
Kopriva[R]	Urtica dioica	Urtica dioica	folium
Kantarion[R]	Hypericum perforatum	Hypericum perforatum	folium, flos and stems
Šipak[R]	Rosa canina	Rose hip	fructus
Hajdučka trava[R]	Achillea millefolium	Milfoil	folium, flos and stems

mogenized in accordance with the standard EN 13804. Samples (10 g) treated with acid and ash residues are steamed until dried, and then mineralized at 425ºC, gradually increasing temperature. The process is repeated until the sample totally burns. An ash has to be white/gray or a little bit coloured. The ash is then dissolved in hydrochloric acid; and if necessary, the sample is filtered before dissolving to the certain volume. Solutions for determination of arsenic were pre-reduced (As^{5+} to As^{3+}) prior to analysis. This was achieved by adding a reduced solution containing 5% (w/v) potassium iodide (KI) and 5% (w/v) ascorbic acid. A 10 mL of solution which we used for determination of arsenic was transferred in 50 mL volumetric flask and then 1 mL of 5% KI and 1 mL ascorbic acid was added. Finally, solution was made up to mark with 2M hydrochloric acid (HCl). All reagents were of analytical grade. All sample containers and glassware were acid soaked in 10% v/v nitric acid for at least 24 h and then rinsed several times with deionized water before use. Arsenic is quantitatively determined at a wavelength of 193.7 nm.

For determining a calibration curve, certified standards AccuTraceTM Reference Standard concentration 1000 g/L were used. High purity deionized water was used for dilution of standards and samples. Each sample was measured in duplicate. Arsenic content values measured in herbal tea samples are expressed in mg/kg.

The names of medicinal plants and plant parts used in the study are presented in Table 1.

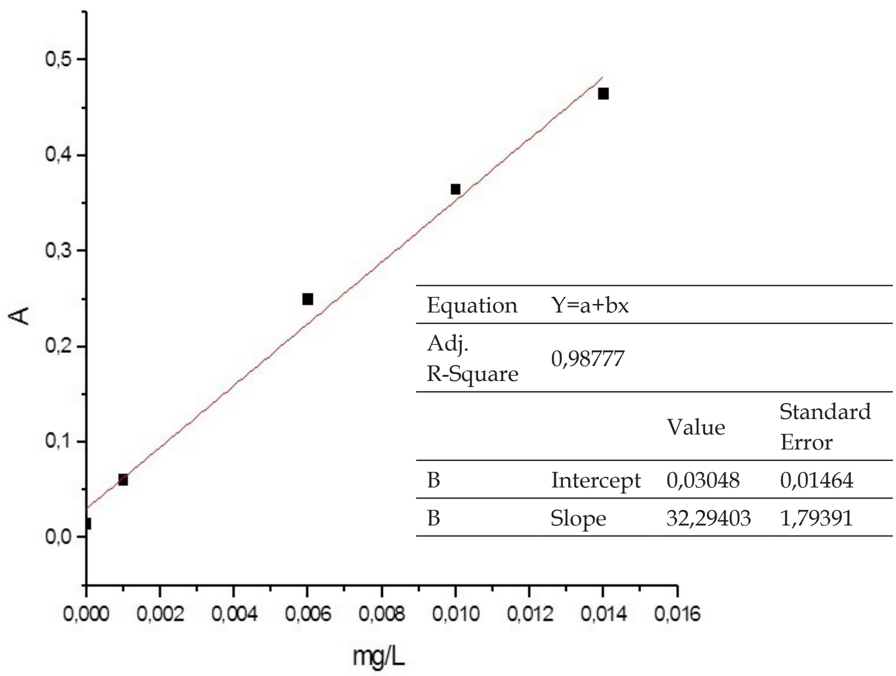

Fig. 1 Calibration curve used to determine arsenic content in tested samples

Results

Figure 1 shows a calibration curve used to determine arsenic content in tested samples.

Table 2. Arsenic content in herbal teas (mg/kg)

Commercial name	First measurement	Second measurement	Mean	Standard deviation
Green[R]	0.012	0.014	0.013	0.001
Black[R]	0.017	0.020	0.014	0.003
Chamomile[R]	0.011	0.007	0.009	0.003
Mint[R]	0.014	0.012	0.013	0.001
Thyme[R]	0.021	0.022	0.021	0.001
Anisi[R]	0.02	0.025	0.022	0.003
Sambucus nigra[R]	0.17	0.12	0.145	0.035
Uva ursi[R]	0.007	0.011	0.009	0.003
Althea[R]	0.09	0.07	0.08	0.014
Urtica dioica[R]	0.03	0.04	0.035	0.007
Hypericum perforatum[R]	0.02	0.04	0.03	0.014
Rose hip [R]	0.03	0.05	0.04	0.014
Milfoil[R]	0.026	0.027	0.026	0.001

Table 3. Arsenic content in fruit teas (mg/kg)

Commercial name	First measurement	Second measurement	Mean	Standard deviation
Fruit mix	0.02	0.035	0.027	0.010
Aronia	0.017	0.021	0.019	0.002
Cranberry	0.012	0.016	0.014	0.003

Correlation coefficient of the calibration curve is 0.988, with a linear equation $y = 0.031 + 32.294x$.

Arsenic concentration in the herbal tea samples ranged from 0.009 to 0.145 mg/kg (Table 2). The highest arsenic content was measured in the sample of Sambucus nigra tea, actually 0.145 mg/kg, leading to a conclusion that herbs were collected in the fields where arsenic from the atmosphere could contaminate Sambucus nigra used for tea preparation. The lowest arsenic concentration of 0.007 mg/kg was found in Chamomile tea and Uva ursi. Uva ursi is collected as a wild plant at elevation above 1200 m, where pollution and contamination are certainly less than in urban and industrial areas indicating a clear connection between arsenic content and herb origin (Table 2).

Arsenic content in the fruit tea samples is presented in Table 3 and ranged from 0.014 mg/kg (Cranberry) to 0.027 mg/kg (Fruit mix).

Discussion

Fertile soils supply plants with all chemical elements necessary for growth. Thereby, soil can transmit toxic elements to plants [6]. Additional sources of contamination of plants with heavy metals are atmospheric precipitation, protective chemicals, fertilizers, industrial plants, thermal power plants, the proximity of mines and frequent roads; arsenic is considered to have carcinogenic properties and its very low concentrations are highly toxic to living organisms [6]. Toxic elements (mercury, arsenic, lead and cadmium), which are present in some plants, can severely damage haemopoietic, immune, nervous and reproductive systems. For this reason, a content of heavy metals is one of the criteria for the use of plant material in the production of traditional medicines and herbal infusions [7]. This instigates need for constant and organized safety control of plants that are used as raw materials in pharmaceutical industry.

The arsenic content in plants is normally lower than in soil and it does not influence plant growth and development. Some authors appoint stimulant effects of low concentrations of arsenic to root growth. However, a high concentration of arsenic has a negative effect. A normal concentration of arsenic in developed leaves of various plants is 1-1.7 ppm/dry matter and the value of 5-20 ppm of arsenic is considered toxic [8]. Karak and Bhagat [9] state the fact that arsenic concentration in fresh leaves and young tea shoots (China origin) is 0.024– 0.066 mg/kg and 0.021–0.073 mg/kg, respectively.

Arsenic content in tea samples presented here is below the value of 1 mg/kg, stipulated by the national legislation [10]. Ražić and Kuntić [5] have examined arsenic content in samples of mint, chamomile and rose hip. Arsenic content in mint samples (original package) of 0.086 mg/kg and of 0.155 mg/kg (bulk) is significantly higher than those in our study 0.013 ± 0.001 mg/kg. Also, the values of arsenic content in rose hip samples of 0.101 mg/kg (original package) and of 0.088 mg/kg (bulk) are higher than ours.

Szentmihályi et al. [11] have found arsenic content of 3.63 ± 60 mg/kg in nettle (herba), which is considerably different from our test results. Also, arsenic content of 1.66 mg/kg in selected tea products from the Ghanaian market is considerably higher in comparison to our results [12].

On the contrary, Gruszecka-Kosowska et al. [13] have found no arsenic and low concentrations of other metals in oolong teas from China but it has been recommended to discard the first tea infusion to reduce the metal concentrations before consumption. Similar results have been obtained by Fatemeh at al. [14] for Iranian black tea.

Conclusion

Sixteen samples of different types of herbal and fruit teas, commonly used among the population of the Republic of Srpska, were analyzed for arsenic, a toxic element, in order to gain insight into safety of medicinal plants used to prepare popular tea beverages. Arsenic content in all analyzed tea samples is below the value of 1 mg/kg, stipulated by the national legislation ranging from 0.007 mg/kg to 0.17 mg/kg.

Teas are traditionally very often used not only as a beverage but also as an adjunct to therapy and often as self-medication. The results presented here have confirmed safety of analyzed herbal and fruit teas which were produced and distributed in the Republic of Srpska.

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Određivanje sadržaja arsena u uzorcima čajeva dostupnih na tržištu Republike Srpske primjenom atomske apsorpcione spektrofotometrije

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Uvod. Arsen u prirodi i živim organizmima postoji u različitim oblicima. Toksični elementi, uključujući i arsen, koji su prisutni u nekim biljkama mogu ozbiljno oštetiti hematopoetski, imuni, nervni i reproduktivni sistem čovjeka. Iz tog razloga, sadržaj teških metala je jedan od kriterijuma za procjenu bezbjednosti upotrebe biljnog materijala u proizvodnji tradicionalnih lijekova i biljnih čajeva. To zahtijeva stalnu i organizovanu kontrolu biljaka koje se koriste kao sirovine u farmaceutskoj industriji. Cilj ove studije je utvrđivanje sadržaja arsena u odabranim čajevima koji su dostupni na tržištu Republike Srpske.

Metode. Analizirano je 13 biljnih i 3 voćna čaja. Uzorci od po 10 g su mineralizovani i sadržaj arsena je određen atomskim apsorpcionim spektrofotometrom Agilent Technologies Series 200 sa vazduh/acetilen gorionikom i korekcijom pozadine D2.

Rezultati. Srednje vrijednosti koncentracije arsena u uzorcima biljnog čaja kretale su se od 0,009 mg/kg do 0,145 mg/kg. Najniža koncentracija arsena od 0,007 mg/kg pronađena je u čaju od kamilice i uve, koja je sakupljena kao divlja biljka na nadmorskoj visini iznad 1200 m. Najveća koncentracija arsena pronađena je u uzorku čaja od zove (0,145 mg/kg). U voćnim čajevima koncentracija arsena bila je u rasponu od 0,014 mg/kg (brusnica) do 0,027mg/kg (voćna mješavina).

Zaključak. Sadržaj arsena u svim analiziranim uzorcima čajeva niži je od 1 mg/kg, tj. od maksimalno dozvoljene količine propisane nacionalnim zakonodavstvom.

Ključne riječi: arsen, biljni čajevi, voćni čajevi, atomska apsorpciona spektrofotometrija