International Food Research Journal 29(3): 646 - 658 (June 2022)
Journal homepage: *http://www.ifrj.upm.edu.my*

Mineral and nutritional assessments of soybean, buckwheat, spelt, and maize grains grown conventionally and organically

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Article history

Received:
20 January 2020
Received in revised form:
13 April 2021
Accepted:
21 November 2021

Keywords

mineral,
grain,
organic growing system,
conventional growing system,
principal component analysis

Abstract

The present work determined complete mineral profile of four different types of grains (spelt, soybean, maize, and buckwheat) grown under two growing systems – conventional and organic. The contents of 20 macro-, micro- and trace elements were analysed in the examined grains by inductively coupled plasma-optical emission spectrometry (ICP-OES). In most samples, nine elements were present in concentrations higher than 10 mg/kg. The remaining elements were present at lower concentrations or in traces or not detected in certain samples. Aluminium and arsenic, as two toxic elements, were detected only in organic buckwheat grains. Based on the obtained results for the mineral contents, a nutritive assessment of the quality of grains of spelt, soybean, maize, and buckwheat were made. Results of nutritional assessment showed that spelt, soybean, and buckwheat grains could potentially be good sources of several minerals for human diet. On the other hand, the presence of some toxic elements, such as cadmium and strontium, should be monitored. Results of principal component analysis (PCA) and hierarchical cluster analysis (HCA) shown that the mineral composition, to a much greater extent, depended on the botanical origin of grains as compared to the production system.

Introduction

Macro- and microelements (the old name is minerals), along with trace elements, are the essential components of everyday human nutrition. The importance of balanced elements input through food can be associated with their role in enzyme activation for metabolic functions. The lack of essential minerals in nutrition can lead to the development of some acute or chronic diseases (Branca and Ferrari, 2002). Metal ions and complexes have an important role in vital functions of organisms. Micronutrients such as iron (Fe), zinc (Zn), copper (Cu), and manganese (Mn) are important parts of several biochemical processes in all living organisms, and because of which, they are considered as essential (Hicsonmez *et al*., 2012). Grains and cereal grains are rich sources of different minerals, and perfectly suitable for human or animal feeding. Bakoglu *et al*. (2009) found that the grains of several plant species contain Cu, Mn, Cr, Ni, Zn, Fe, Mg, and Al in the range of 18.40 - 45.54%. In addition, some of these elements (Al, Sr, Cd, As, Pb, *etc.*) are toxic to both plants and humans. Their presence is undesirable due to safety issues, thus very important to determine.

The chemical composition of different cereals/pseudocereals can vary depending on geographical origins and plant cultivation systems, but generally, they are good sources of proteins, starches, oils, sugars, vitamins, macro-, and micro-elements. Grains of soybean [*Glycine max* (L.) Merr.] are rich in proteins (up to 40%), oils (up to 20%), carbohydrates (17 - 19%), and minerals (6 - 7%) (Kumar *et al*., 2017), which explains its great applications as food, feed, and industrial raw material (Gibson and Mullen, 2001). Three main

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DOI: https://doi.org/10.47836/ifrj.29.3.16

macroelements in soybean grains are Ca, K, and Mg, while Fe, Cu, and Zn are important microelements. Spelt (*Triticum aestivum* ssp. *spelta*) possesses significantly higher content of different elements as compared to wheat (*T. aestivum* ssp. *vulgare*), especially P, Fe, K, Zn, Mg, and Cu (Golijan *et al*., 2017). Buckwheat (*Polygonum fagopyrum* L.), as one of the most important pseudocereals, contains high amounts of K, Mg, P, Ca, Fe, Cu, and Zn located predominantly in its grainlings. Buckwheat grains are also good sources of Se, Ba, B, I, Co, and Pt (Bonafaccia *et al*., 2003). Maize (*Zea mays* L.) is an important food source for humans and animals, especially in developing countries. Predominant mineral in maize grains is P, but low content of Ca and trace elements also exist (FAO, 1992).

The production of crops in organic and conventional production systems leads to significant differences in the mineral composition of the crop grains (Bourn and Prescott, 2002). Due to good composition and quality, which are in accordance with the modern nutritional requirements, spelt and buckwheat take the leading role in organic production system. Organic agriculture is one of the fastest growing fields, currently spread on 50.9 million hectares of land area with a profit of more than USD 80 billion (Lernoud and Willer, 2017). The aim of the present work was therefore to study and determine whether the mineral compositions of soybean, buckwheat, spelt, and maize grains grown in Serbia under conventional and organic production systems are significantly different. Principal component analysis (PCA) and hierarchical cluster analysis (HCA) were used for this assessment. Furthermore, theoretical assessment of nutrition value, expressed through mineral composition of selected grains, was also determined to establish their potential as a source of elements for human diet.

Materials and methods

Grain samples

Depending on availability, ten different samples of grains grown under conventional and organic conditions were collected and analysed during one (2016) or two seasons (2015 - 2016). Samples were coded as organic and conventional maize grains (OM and CM, respectively); organic and conventional spelt grains (OS and CS, respectively); organic and conventional soybean grains (OSo and CSo, respectively), and organic and conventional buckwheat grain (OB and CB, respectively).

Grain samples were grown and collected at three different locations: (1) Maize Research Institute at Zemun Polje (Belgrade) – (i) organic maize (Rumenka hybrid) collected in 2015 (OM15) and 2016 (OM16), (ii) conventional maize (Rumenka hybrid) collected in 2016 (CM16), and (iii) organic spelt (Nirvana hybrid) collected in 2015 (OS15) and 2016 (OS16); (2) Institute of Field and Vegetable Crops at Novi Sad – (i) organic (OSo16) and conventional (CSo16) soybean (Kaća hybrid) collected in 2016; and (3) Nova Varoš – (i) conventional spelt collected in 2016 (CS16) and organic (OB16) and conventional (CB16) buckwheat collected in 2016.

Analytical methods

The determination of 20 macro- (K, P, S, Mg, Ca, Zn, Fe, Na, Mn), micro-, and trace (Al, As, B, Cd, Co, Cu, Li, Ni, Sb, Se, Sr) elements was conducted by inductively coupled plasma-optical emission spectrometry (ICP-OES) following the procedures described by Kostić *et al*. (2015).

Statistical methods

Results were expressed as mean ± SD of triplicate measurements (n = 3) obtained using Statistica 8.0 software program (StatSoft Co., Tulsa, OK, USA). Significant differences between samples were determined by t-test ($p < 0.05$). The correlation analysis between macro-, micro-, and toxic elements, *i.e.* between two cultivation systems for different soybean, buckwheat, spelt, and maize grains was performed by Pearson's correlation coefficient (r) using Statistica software version 8.0. Correlations at $p < 0.05$ were considered significant. Principal component analysis (PCA) and hierarchical cluster analysis (HCA) were carried out by PLS ToolBox, v.6.2.1, for MATLAB 7.12.0 (R2011a). All data were autoscaled prior to any multivariate analysis. PCA was carried out using a singular value decomposition algorithm and a 0.95 confidence level for Q, and T2 Hotelling limits for outliers. Results of HCA were presented as a dendrogram in which the steps in the hierarchical clustering solution and values of the distances between clusters (Euclidean distance) were represented.

Results and discussion

Macroelement determination

The utilisation of ICP-OES allowed for the simultaneous determination of a large number of macro- and microelements, as well as the trace elements present in the samples. Comparing the contents of macro- (K, P, S, Mg, Ca, Fe, Zn, Na, and Mg), micro-, and trace elements (Al, As, B, Cd, Co, Cu, Li, Ni, Sb, Se, and Sr) in the examined organically and conventionally grown grains, significant differences for certain minerals were observed.

The contents of macroelements (mg/kg) detected in the grain samples are shown in Table 1. Phosphorus (P) ranked the highest in all four types of grains, and is in accordance with Soetan *et al*. (2010) who remarked that the high concentrations of P are characteristic for grains. In plants, P serves as a structural component of carbohydrates and proteins, and in metabolism, it is particularly important for the composition of adenosine triphosphate (ATP). P in the tested grain samples ranged from 1,830.47 (OM15) to 6,046.98 (OSo16) mg/kg. For maize grains, the highest content of P was detected in OM16 sample (3,221.14 mg/kg). For spelt grains, the highest content of P was detected in OS15 sample (3,541.92 mg/kg), and the lowest in CS16 sample (2,780.59 mg/kg). Examining the mineral composition of various wheat genotypes, Hussain *et al*. (2010) found that P content in spelt grain was 4.280 mg/kg. The soybean grains contained the highest contents of P (CSo16: 4,473.50 mg/kg; OSo16: 6,046.98 mg/kg) as compared to other tested grains. Similar results were reported by Rezende Costa *et al*. (2015) between 4,620.21 and 6,151.77 mg/kg. In buckwheat grains, grown organically and conventionally, the amount of P was similar to that in spelt grains (OB16: 2,692.17 mg/kg; CB16: 2,905.15 mg/kg). According Zhang and Xu (2017), the contents of P in buckwheat grains ranged from 3,258 to 3,953 mg/kg.

In plants, magnesium (Mg) is important as a component of chlorophyll, and as a coenzyme in a large number of enzymes; its deficiency leads to chlorosis and decay of leaves. In human body, it is important for the proper development of bones, teeth, and as a cofactor of a large number of enzymes. Mg deficiency in human nutrition (after only five days) leads to disorders of the cardiovascular system (Soetan *et al*., 2010). Among the tested grain samples, the lowest amount of Mg was found in OM15 grain (420.22 mg/kg), while the maize grains collected in 2016 contained two times higher values (950.92 – 1,017.09 mg/kg), as well as the spelt grains collected in 2015 and 2016. Other authors also found a similar amount of Mg in maize and spelt grains: 1,153.9 mg/kg of Mg in maize grains (Özcan, 2006), and 1,280 mg/kg of Mg in spelt grains (Hussain *et al*., 2010). Buckwheat grains contained a slightly higher amount of Mg than in maize and spelt grains (OB16: 1,125.89 mg/kg; CB16: 1,164.34 mg/kg), which is consistent with the results of Mann *et al*. (2012) who reported 1,230 - 1,817 mg/kg of Mg in buckwheat grains. The highest concentration of Mg was detected in soybean grains (CSo16: 1,214.44 mg/kg; OSo16: 1,240.76 mg/kg). Rezende Costa *et al*. (2015) found slightly higher concentrations of Mg in soybean grains (1,900.22 - 2,625.78 mg/kg).

Calcium (Ca) is a mineral with numerous essential functions in the human body. It is very important as a building element of teeth and bones, and its role in the regulation of muscle and nerve functions, activation of enzymes, and blood coagulation is particularly important. In plants, Ca is found in an ionised form of Ca^{2+}. The leaves have high content of Ca and low content of P, whereas in the grains, this is reversed. The family Leguminosae is usually very rich in Ca, especially when compared to grasses that have lower content (Soetan *et al*., 2010). In the present work, the lowest content of Ca was detected in OM15 (0.23 mg/kg), while slightly higher values were found in CM16 (10.44 mg/kg) and OM16 (11.39 mg/kg). In Turkey, Özcan (2006) found the Ca content in maize grains to be 68.4 mg/kg as compared to 1,867.21 mg/kg in soybeans. The low Ca content in maize grains was also reported by other authors (FAO, 1992; Soetan *et al*., 2010). In spelt grains, the contents of Ca ranged from 87.24 to 166.08 mg/kg, while higher contents were detected in buckwheat grains (OB16: 297 mg/kg; CB16: 348.16 mg/kg). In spelt grains, 327 mg/kg of calcium was found (Hussain *et al*., 2010). Mann *et al*. (2012) proved that both types of buckwheat (*Fagopyrum esculentum* and *F. tataricum*) were rich sources of minerals such as Mg, P, K, Ca, and Na. The Ca content in *F. esculentum* was 748.3 mg/kg, while in *F. tataricum*, it was lower (505.48 mg/kg). In the present work, the highest content of Ca was found in organic soybean grains (1,531.09 mg/kg), which is also the characteristic of plants from the leguminous family (Soetan *et al*., 2010).

Table 1. Contents of macro-, micro-, and toxic elements (mg/kg ± SD) in conventionally and organically grown grains of maize, spelt, soybean, and buckwheat.

	Macro-element								
Sample	**K**	**P**	**S**	**Mg**	**Ca**	**Zn**	**Fe**	**Na**	**Mn**
OS16*	94.30 ± 1.30^{a}	2,910.18 ± 36.93^{a}	1,030.10 ± 12.24^{a}	828.63 ± 8.59^{a}	87.24 ± 1.98^{a}	15.26 ± 0.30^{a}	8.79 ± 0.10^{a}	24.84 ± 0.29^{a}	20.39 ± 0.85^{a}
CS16	109.67 ± 1.66^{b}	2,780.59 ± 84.35^{af}	1,029.06 ± 16.72^{a}	835.70 ± 13.73^{a}	88.82 ± 1.30^{a}	15.67 ± 0.17^{a}	10.14 ± 0.20^{b}	24.30 ± 0.72^{a}	29.64 ± 0.33^{b}
CM16	113.28 ± 1.19^{c}	2,598.72 ± 31.29^{b}	802.15 ± 10.40^{b}	950.92 ± 14.00^{b}	10.44 ± 0.12^{b}	12.17 ± 0.29^{b}	7.26 ± 0.12^{c}	9.26 ± 0.11^{b}	2.41 ± 0.11^{c}
OM16	119.08 ± 2.49^{d}	3,221.14 ± 34.31^{c}	925.68 ± 11.65^{c}	1,017.09 ± 12.25^{c}	11.39 ± 0.14^{c}	17.81 ± 0.30^{c}	7.12 ± 0.12^{c}	9.18 ± 0.16^{b}	2.92 ± 0.12^{d}
OSo16	309.56 ± 4.56^{e}	6,046.98 ± 65.02^{d}	2,814.02 ± 106.98^{d}	1,240.76 ± 12.65^{d}	1,531.09 ± 1.13^{d}	19.92 ± 0.76^{d}	27.25 ± 0.30^{d}	147.53 ± 4.59^{c}	21.13 ± 0.43^{a}
CSo16	285.74 ± 7.40^{f}	4,473.50 ± 51.59^{e}	2,249.30 ± 29.84^{e}	1,214.44 ± 17.10^{d}	1,138.42 ± 14.99^{e}	25.05 ± 0.71^{e}	18.60 ± 0.67^{e}	117.92 ± 1.58^{d}	16.95 ± 0.80^{e}
OB16	106.81 ± 2.42^{b}	2,692.17 ± 36.18^{f}	993.59 ± 23.21^{a}	1,125.89 ± 12.34^{e}	297.00 ± 4.00^{f}	14.18 ± 0.59^{f}	122.55 ± 2.90^{f}	51.11 ± 0.68^{e}	116.63 ± 1.71^{f}
CB16	123.79 ± 4.36^{d}	2,905.15 ± 104.78^{a}	1,051.05 ± 12.60^{a}	1,164.34 ± 20.01^{f}	348.16 ± 6.27^{g}	15.20 ± 0.50^{af}	94.33 ± 3.00^{g}	55.26 ± 0.62^{f}	110.70 ± 2.56^{g}

	Micro- and toxic element									
Sample	**Al**	**As**	**B**	**Cd**	**Co**	**Cu**	**Li**	**Ni**	**Sb**	**Se**	**Sr**
OS16*	n.d.	n.d.	n.d.	0.050 ± 0.002^{a}	n.d.	2.79 ± 0.13^{a}	8.93 ± 0.17^{a}	0.32 ± 0.01^{a}	0.12 ± 0.05^{a}	0.140 ± 0.004^{a}	8.21 ± 0.10^{ab}
CS16	n.d.	n.d.	n.d.	0.060 ± 0.002^{b}	n.d.	3.13 ± 0.08^{b}	8.96 ± 0.27^{a}	0.44 ± 0.01^{b}	0.120 ± 0.005^{ab}	0.120 ± 0.004^{b}	8.42 ± 0.13^{af}
CM16	n.d.	n.d.	n.d.	0.030 ± 0.001^{c}	n.d.	1.53 ± 0.04^{c}	2.14 ± 0.03^{b}	0.29 ± 0.01^{c}	0.12 ± 0.01^{a}	0.100 ± 0.002^{c}	6.51 ± 0.12^{c}
OM16	n.d.	n.d.	n.d.	0.030 ± 0.001^{c}	n.d.	1.32 ± 0.03^{d}	2.47 ± 0.04^{c}	0.20 ± 0.01^{d}	0.110 ± 0.003^{b}	0.17 ± 0.01^{d}	8.15 ± 0.10^{b}
OSo16	n.d.	n.d.	23.75 ± 0.28^{a}	0.050 ± 0.002^{a}	0.130 ± 0.005^{a}	10.07 ± 0.17^{e}	68.03 ± 0.85^{d}	2.17 ± 0.04^{e}	n.d.	0.25 ± 0.01^{e}	17.93 ± 0.24^{d}
CSo16	n.d.	n.d.	24.03 ± 0.98^{a}	0.040 ± 0.001^{d}	0.060 ± 0.001^{b}	10.48 ± 0.16^{f}	54.22 ± 2.08^{e}	2.17 ± 0.07^{e}	0.030 ± 0.001^{c}	0.170 ± 0.002^{d}	13.49 ± 0.37^{e}
OB16	86.76 ± 1.65	0.010 ± 0.005	2.24 ± 0.07^{b}	0.060 ± 0.001^{b}	0.3 ± 0.01^{c}	5.58 ± 0.01^{g}	22.19 ± 0.76^{f}	10.60 ± 0.15^{f}	0.13 ± 0.01^{a}	0.070 ± 0.003^{f}	8.63 ± 0.11^{f}
CB16	n.d.	n.d.	2.66 ± 0.88^{c}	0.060 ± 0.002^{b}	0.21 ± 0.01^{d}	5.70 ± 0.27^{g}	24.23 ± 0.27^{g}	11.39 ± 0.12^{g}	0.18 ± 0.01^{d}	0.050 ± 0.002^{g}	9.28 ± 0.27^{g}

O: organic; C: conventional; S: spelt; M: maize; So: soybean; B: buckwheat; and n.d.: not detected. Means followed by the same lowercase superscripts in the same column are not significantly different ($p < 0.05$).

Zinc (Zn) and iron (Fe) represent essential nutrients, both for plants and humans (Hambridge, 2000; Soetan *et al*., 2010). They are cofactors in many enzymes, while Fe is important for the functioning of haemoglobin and myoglobin. About 800,000 children around the world die due to the deficiency of Zn (Ortiz-Monasterio *et al*., 2007). *Triticum* species such as *T. dicoccoides*, *T. monococcum*, *T. boeticum*, and *Aegilops tauschii* are best sources of Zn and Fe for human consumption (Ortiz-Monasterio *et al*., 2007). In the present work, the lowest content of Zn (only 9.06 mg/kg) was detected in OM15 as compared to other grains, while OSo16 and CSo16 had the highest (19.92 and 25.05 mg/kg, respectively). Similar values of Zn in maize grains (15 - 47 mg/kg) were found by Ortiz-Monasterio *et al*. (2007). According to Oury *et al*. (2006), the Zn content in *T. aestivum* ranged from 15 - 35 mg/kg, and the Fe content from 20 - 60 mg/kg, as compared to 39.2 mg/kg in spelt grains (Hussain *et al*., 2010). Rezende Costa *et al*. (2015) found that the Zn content in the grains of commercial soybean varieties ranged from 38.72 - 60.20 mg/kg. In the present work, in spelt grains, the content of Zn ranged from 15.26 - 19.11 mg/kg, and similar content was detected in buckwheat grains (OB16: 14.18 mg/kg; CB16: 15.20 mg/kg). For Fe, it was found the lowest (4.37 - 7.26 mg/kg) in maize grains, while in spelt grains it was higher (8.79 - 12.04 mg/kg). When compared to the results of other authors: 20 - 60 mg/kg (Oury *et al*., 2006), 38 mg/kg (Hussain *et al*., 2010), the spelt grains in the present work had a lower content. Significantly higher concentrations of Fe (from 70.82 - 99.22 mg/kg) were found in soybean samples (Rezende Costa *et al*., 2015). In the present work, the highest concentration of Fe was found in buckwheat grains (CB16: 94.33 mg/kg; OB16: 122.55 mg/kg). Mann *et al*. (2012) found great differences in the Fe contents: 2.208 mg/kg in *F. esculentum* and 15.92 mg/kg in *F. tataricum*. The observed differences may indicate the importance of geographical origins and the growing methods of the plant.

Sodium (Na) plays a very important role in numerous biological processes in the cell (Soetan *et al*., 2010) where it is found as the main cation in extracellular fluid. The Na contents in the grain samples examined in the present work ranged from 7.11 mg/kg (OM15) to 147.53 mg/kg (OSo16).

Manganese (Mn) is an important cofactor in a number of enzymes (Soetan *et al*., 2010), and has a particularly important role in redox processes in plants, especially during photosynthesis. In the grain samples examined in the present work, the lowest content of Mn was found in maize grains (1.90 - 2.92 mg/kg), while spelt grains yielded higher amount (Table 1). A similar amount of Mn was also detected in soybean grains (CSo16: 16.95 mg/kg; OSo16: 21.13 mg/kg). Buckwheat grains yielded the highest Mn contents (CB16: 110.70 mg/kg; OB16: 116.63 mg/kg), exceeding that reported by Ikeda (2002) who reported that 100 g of buckwheat flour could fulfil the following daily needs: 40 - 53% of Mn, 21 - 28% of Zn, 30 - 39% of Cu, 4% of Ca, 75 - 100% of Mg, 22% of K, and about 59% of P.

Micro- and trace element determination

The results of microelements (below 10 mg/kg in all or the most of examined samples) and trace elements in different grain samples are shown in Table 1.

Lithium (Li) is an important microelement for human neurological system, and its main sources in diet are grains and vegetables (Schrauzer, 2002). The lowest content of Li was found in maize grains, ranging from 1.39 - 2.47 mg/kg, while the highest in soybean grains (CSo16: 54.22 mg/kg; OSo16: 68.03 mg/kg).

Copper (Cu) deficiency leads to disorders in Fe metabolism in the liver (Soetan *et al*., 2010). In the tested grain samples, Cu contents ranged from 1.08 mg/kg (OM15) to 10.48 mg/kg (CSo16). The highest concentrations were found in soybean grains, and similar to the concentrations found by Rezende Costa *et al*. (2015) which were 9.48 - 15.92 mg/kg.

Nickel (Ni) is an essential trace element for animals and humans as an important enzyme cofactor, prolactin, and cell membrane function controller (Soetan *et al*., 2010). Given that the need for Ni is very low, everyday nutrition generally satisfies its requirement. At higher concentrations, Ni can lead to toxic effect.

Selenium (Se) plays a very important role in protecting organisms from oxidative stress, and maintaining the normal functioning of glutathione peroxidase and thioredoxin reductase enzymes. Toxicity of Se is manifested by hair loss and/or dermatitis (Soetan *et al*., 2010). Among the tested grains, the lowest contents of Se (0.05 mg/kg) were found in OM15 and CB16, while the highest (0.25 mg/kg) in OSo16.

Cobalt (Co) and boron (B) were found in soybean and buckwheat grains in the range of 0.06

(CSo16) to 0.21 mg/kg (CB16) for Co, and 24 mg/kg (soybean) and 2.24 - 2.66 mg/kg (buckwheat) for B.

Aluminium (Al) is one of the most toxic metals for plants, whose level usually does not exceed 200 mg/kg of dry weight (Mossor-Pietraszewska, 2001), with grains usually having the lowest content of Al. Unlike plants known as Al-accumulators, which can grow in the presence of high concentrations of Al in the soil, maize is a plant highly sensitive to Al. High concentrations of Al have very unfavourable effects on human health, which is why in the food industry it is very important to monitor its content in cereals and to limit it (Shen *et al*., 2006). In accordance with the claim that Al is a potential cause of a large number of neurodegenerative diseases, it was necessary to establish a provisional tolerant weekly intake (PTWI) of 70 mg/kg body weight for adult, *i.e.* 10 mg/kg daily (Shaw and Tomljenović, 2013). In the tested grain samples, the presence of Al was only detected in OB16 (86.76 mg/kg). It should be emphasised that buckwheat is known as a plant that has a high resistance to Al in the soil, and is classified as an Al-accumulator plant (Shen *et al*., 2006). Earlier authors also confirmed the presence of Al in buckwheat grain shell (21.3 - 41.2 mg/kg). Interestingly, that, in the same grain sample (OB16), arsenic (As) was present in trace amount (0.01 mg/kg), which can also be characterised as a toxic and undesirable element.

For antimony (Sb), its content is not often found in cereals. In the present work, Sb was not detected in organic soybean grain samples (OSo16), while in other grain samples, it was present at 0.03 mg/kg (CSo16) to 0.18 mg/kg (CB16). The main source of Sb in food is migration from PET-packaging where it is used as the initiator of polymerisation as antimony trioxide, Sb_2O_3 (EFSA, 2004). Cadmium (Cd) and strontium (Sr) are two potentially toxic elements for humans, and their presence in some food samples is not desirable. In all examined grains, Cd was found at very low concentrations (0.03 - 0.06 mg/kg), while Sr was present as a microelement (6.51 - 17.93 mg/kg).

Nutritional assessment

The bioavailability of certain elements in grains is limited due to the presence of phytic acid (PA), an important anti-nutrient (Reddy *et al*., 1982; Kumar *et al*., 2010). This organic acid forms insoluble complexes with several metal ions (especially multivalent ions) or proteins, thus rendering them inaccessible at pH 6 in human duodenum (Reddy *et al*., 1982). Furthermore, the lack of phytase enzyme in non-ruminant animals (including human) disable the degradation of these complexes. Among nutritionally important elements, Zn, Cu, Co, Mn, Fe, Ca, and Mg have the greatest capability to bind with PA (Reddy *et al*., 1982). In cereals, PA contents are mostly between 1 - 2%, but can also reach up to 3 - 6% (Febles *et al*., 2002). Grain legumes contain from 0.40 - 2.06% of PA (Urbano *et al*., 2000). PA content is strongly dependent on plant variety, growing soil composition, geographical origin, and climatic conditions (Urbano *et al*., 2000). It was shown by Ruibal-Mendieta *et al*. (2005) that spelt grains contained significantly lower PA amount (about 40%) as compared to regular wheat grains. Also, Mason *et al*. (1993) showed that the presence of phytates does not influence Ca bioavailability. In addition to its anti-nutritional properties, lately, there has been an increasing amount of information on the beneficial properties of PA and phytates, especially in the prevention of cancerous diseases (Kumar *et al*., 2010).

Due to these, the assessment of nutritional values of examined grains was based on theoretical calculation regardless of the PA content, similar to the literature estimation (Gülfen and Özdemir, 2016; Koubová *et al*., 2018). The obtained results are presented in Tables 2 and 3.

Since more than 70% of P in grains is in PA form (García-Estepa *et al*., 1999), it is predominantly unavailable for human diet. Based on adequate daily intake (ADI) value for K and recommended daily intake (RDI) value for S (Institute of Medicine, 2005) (Table 2), input of these two elements through grain consumption is negligible (K) or low (S) despite their high concentration in grain samples. Comparing the RDI value for Mg (Table 2) (Institute of Medicine, 1997) with the results obtained for the examined grain samples, it was noted that the estimated daily intake of 100 g of grains would meet daily Mg requirement between 10% (maize) and 29.54% (soybean). By only including soybean grains in the diet could make a significant contribution to the daily intake of Ca (11.38 - 15.31%), while with cereals this contribution would be low or negligible (Table 2). Since RDI value for Zn is 11 mg/day (Institute of Medicine, 2001), the consumption of 100 g of examined grains would satisfy between 8.23% (maize) and 22.77% (soybean) of its daily needs (Table 2). For Fe, maize grains can be characterised as a poor source (100 g of maize grains would satisfy 5.47 - 9.07% of daily needs; Institute of Medicine, 2001) (RDI value for iron is 8 mg/day). For spelt (10.99 - 15.05%) and soybean (about 25%), the contribution would be slightly higher, and was consistent with the higher Fe content. Consuming buckwheat grains would meet the daily needs of an adult (117.91 - 153.18%; expressed through the consumption of 100 g of grains). Given the high daily needs for Na (1,500 mg/day), its presence in the tested grains would have a negligible contribution to everyday diet (Table 2). For Mn, maize grains proved to be a poor source, while spelt and soybean (73.69 - 128.89%; Table 2) could be good sources. The intake of only about 20 g of buckwheat grains would completely satisfy the daily needs of an adult man for Mn.

Table 2. Estimated theoretical daily intake (% of total intake) of selected macro-elements through the consumption of 100 g of tested grains.

Macro-element	RDI/ADI*/TDI** [mg/day]	Maize [%intake/100 g grain]	Spelt [%intake/100 g grain]	Soybean [%intake/100 g grain]	Buckwheat [%intake/100 g grain]
K*	4,700[1]	0.24 - 0.25	0.20 - 0.27	0.61 - 0.66	0.23 - 0.26
S	2,800[1]	2.86 - 3.31	3.675 - 3.679	8.03 - 10.05	3.55 - 3.75
Mg	420[2]	22.64 - 24.22	19.73 - 19.9	28.92 - 29.54	26.81 - 27.72
Ca	1,000[2]	0.10 - 0.11	0.87 - 0.89	11.38 - 15.31	2.97 - 3.48
Zn	11[3]	11.06 - 16.19	13.87 - 14.25	18.11 - 22.77	12.89 - 13.82
Fe	8[3]	8.9 - 9.07	10.99 - 12.68	23.25 - 34.07	117.91 - 153.18
Na*	1,500[1]	0.061 - 0.062	0.162 - 0.166	0.79 - 0.98	0.34 - 0.37
Mn**	2.3[3]	10.48 - 12.69	88.65 - 128.89	73.69 - 91.88	483.31 - 507.07

[1]Institute of Medicine (2005); [2]Institute of Medicine (1997); [3]Institute of Medicine (2001); RDI: recommended daily intake; *ADI: adequate daily intake; and **TDI: tolerable daily intake.

Table 3. Estimated theoretical daily intake (% of total intake) of selected micro- and trace elements through the consumption of 100 g of tested grains.

Micro-element	RDI/PTDI* [mg/day]	Maize [%intake/100 g grain]	Spelt [%intake/100 g grain]	Soybean [%intake/100 g grain]	Buckwheat [%intake/100 g grain]
Al*	10[1]	/	/	/	86.76
B*	20[2]	/	/	11.88 - 12.02	1.12 - 1.33
Cu	0.9[2]	14.67 - 17.03	31.01 - 34.80	111.92 - 116.5	61.97 - 63.28
Cd*	0.00036[3]	799.72 - 833.33	1,388.89 - 1,695.28	1,125.56 - 1,388.89	1666.67
Li	1[4]	21.4 - 24.70	89.30 - 89.6	549.16 - 680.33	221.90 - 242.30
Ni*	0.005[2]	399.84 - 579.38	640.00 - 871.26	4340	21,194.82 - 22,789.44
Sb*	0.04[5]	27.5 - 29.88	30.00	7.50	32.71 - 45.12
Se	0.055[6]	18.18 - 30.71	21.82 - 25.45	31.73 - 45.87	8.62 - 12.44
Sr*	0.13[7]	500.77 - 626.66	631.3 - 647.69	1,037.98 - 1,379.37	664.15 - 713.65

[1]Shaw and Tomljenović (2013); [2]Institute of Medicine (2001); [3]EFSA (2009); [4]Schrauzer (2002); [5]EFSA (2004); [6]Institute of Medicine (2000); [7]WHO (2010); *PTDI: provisional tolerable daily intake.

The RDI value for Li is 1 mg/day (Schrauzer, 2002). Based on the obtained results for tested grain samples, the recommended daily intake would be fulfilled through the consumption of 100 g of grains in all cases (Table 3), except for maize (13.93 - 24.70%). For Cu, 100 g of soybean grains would completely meet the daily needs of an adult man (111.92 - 116.5%). Considering the very low tolerable daily intake (TDI) of Ni (Table 3), it is expected that in all tested grain samples, the TDI for Ni would be significantly excessive (from 244% in OS15, to 4340% per 100 g of soybean grains). Considering the recommended daily intake of Se (Table 3) (Institute of Medicine, 2000), it can be seen that spelt (21.82 - 32.73%) and soybean (31.73 - 45.87%) grains could make a significant contribution to daily human diet. Spelt (30 - 35%) and buckwheat (32.7 - 45.1%) grains can significantly contribute to intake of Sb (Table 3). Although in all examined grains Cd was found at very low concentrations (0.03 to 0.06 mg/kg), while Sr was present as a microelement (6.51 to 17.93 mg/kg), consuming less than 35 g of grain samples as food would satisfy the TDI of these two elements. The provisionally tolerable daily intake (PTDI) values (0.00036 mg/kg for Cd, and 0.13 mg/kg for Sr), are proposed at a very low level. It has been reported (Persson *et al*., 1998; Kumar *et al*., 2010) that during *in vitro* experiments, at pH range from 3 to 7, Cd^{2+} ion, besides Zn^{2+} and Cu^{2+} ions, has shown the highest affinity towards PA. In this case, PA can be characterised as a “useful” component since it will make this toxic element “unavailable” for human diet.

PCA, HCA, and correlation analysis

Based on the elements (Table 1) found in grain samples, principal component analysis (PCA) and hierarchical cluster analysis (HCA) were performed to obtain a detailed insight into the data structure, and identify similarities and specificities of object groupings. PCA resulted in a two-component model that explained 89.29% of total variance within the data. The results obtained through analysing the first two principal components based on the contents of elements (Table 1; Ba, Ca, Fe, K, Mg, Na, B, Al, Mn, Ni, Co, Cd, Sb, Cu, Li, Sr, Zn, Pb, Se, and S) in grain samples are shown on score plots and loading plots (Figure 1).

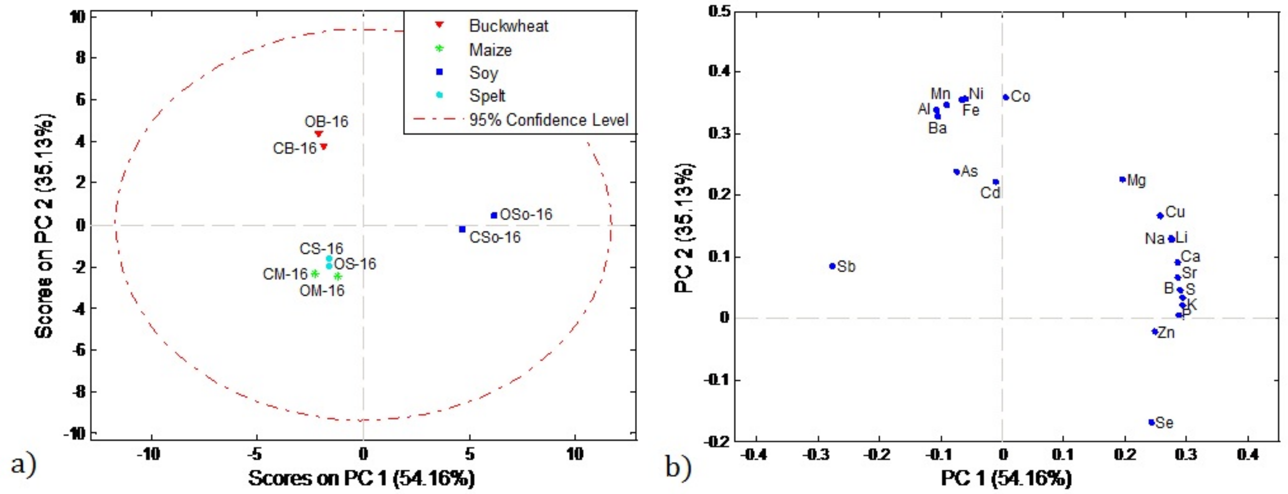

Figure 1. Principal component analysis scores plot **(a),** and loading plots **(b)**.

Three groups of objects stood out in the score plot (Figure 1a). Samples of soybean (OSo-16 and CSo-16) (Table 1) were separated from all the other samples, and created the first group. The second group consisted of samples of buckwheat (OB-16 and CB-16) (Figure 1a, Table 1), while samples of spelt and maize (CS-16, OS-16, CM-16, and OM-16) comprised the third group. Cu, Li, Ca, Sr, B, S, and K had the most positive influence along the PC1 axis on the separation of the first group of objects, which was in accordance with the fact that the concentrations of these elements were the highest in these samples (Figure 1, Table 1). Moreover, the contents of Mg, Na, Zn, P, and Se which were the highest in these samples also influenced the separation of the first group of objects. Al, Mn, Ba, Fe, Ni, Co, and partly As and Cd had the strongest positive effect along the PC2 axis on the separation of the second group of objects (*i.e.* samples OB-16 and CB-16), while Se had a negative effect on their separation along the PC2 axis. The separation of the third group of objects (samples CS-16, OS-16, CM-16, and OM-16) was mostly influenced by Sb, Se, Mg, and Cd; the concentrations of which were higher in spelt and maize samples, than in those of soybean and buckwheat, also due to the fact that other elements were present in far less concentrations as compared to the samples of the first and the second group of objects (Figure 1, Table 1). This was confirmed by the hierarchical cluster analysis (HCA). The results of HCA are shown in a dendrogram (Figure 2).

At a distance of 10, HCA resulted in the separation of samples into three clusters. Samples of soybean belong to the first cluster, buckwheat to the second, while samples of spelt and maize to the third cluster. Additionally, the dendrogram also showed that, at a distance of 2, two sub-clusters were separated within the third cluster. The first sub-cluster consisted of samples of spelt, and the second consisted of samples of maize collected in 2016 (Figure 2). It was also observed that botanical origin of examined seeds had very clear and accurate influence on separating samples since buckwheat (Poligonaceae) and soybean (Fabaceae) always stood out and were separated from maize and spelt samples (Poaceae). On the other hand, production system impact could not be characterised as regular and consistent. Based on PCA and HCA analyses, it could not be concluded that there was a difference in the type of cereal production. In fact, the samples were separated based on the type of grains, but not based on the type of productions. Within each type of sample, there was no clear difference between samples produced under organic and/or conventional productions (Figures 1 and 2).

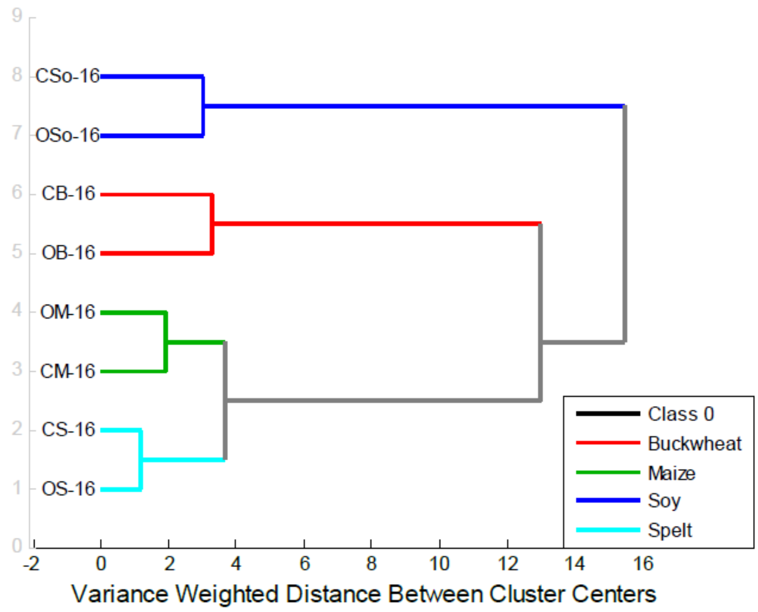

Figure 2. Dendrogram of hierarchical cluster analysis.

Therefore, in addition to PCA and HCA, statistical analysis was also performed in order to determine the possible linkage between the contents of biogenic and toxic elements, as well as between both applied production systems. The results are given in Tables 4 and 5. It was observed that Ar content was strongly positively related with Fe content (r = 0.97), while Sr content was strongly positively correlated with Ca content (r = 0.98), K content (r = 0.94), Na content (r = 0.96), P content (r = 0.98), and S content (r = 0.99). Interestingly, a strong negative correlation was observed with Se and Sb contents (r = -0.90). This toxic element was also in negative correlation with two other important biogenic elements: P (r = -0.90) and S (r = -0.90), which can possibly indicate that they were antagonistic elements.

By comparing applied production systems, correlation analysis revealed that there was strong positive agreement between results obtained for both organic and conventional systems (Table 5). It can thus be concluded that there was no clear assumption whether any of the applied systems could be recommended as a better choice for improvement of mineral composition of maize, soybean, spelt, or buckwheat grains. The obtained results for correlation analysis were in line with the results of PCA and HCA.

Conclusion

Mineral composition results (20 macro-, micro- and trace elements) of ten grain crop samples grown under different conditions (conventional and organic systems) have shown significant differences for most of the elements. Soybean grains had the highest contents of several elements (potassium, phosphorus, sulphur, calcium, boron, copper, lithium, and strontium), as well as buckwheat grains

Table 4. Pearson's correlation coefficient (r) for the relationship between macro-, micro-, and toxic elements of soybean, buckwheat, spelt, and maize grains grown under different conditions.

	Al	As	B	Ca	Cd	Co	Cu	Fe	K	Li	Mg	Mn	Na	Ni	P	S	Sb	Se	Sr	Zn
Al	/	1.00	-0.16	-0.10	0.39	0.74	0.06	0.97	-0.24	-0.03	0.19	0.67	-0.03	0.61	-0.26	-0.20	0.20	-0.40	-0.16	-0.27
As		/	-0.16	-0.10	0.39	0.74	0.06	0.97	-0.24	-0.03	0.19	0.67	-0.03	0.61	-0.26	-0.20	0.20	-0.40	-0.16	-0.27
B			/	0.97	-0.06	0.13	0.93	-0.03	0.99	0.96	0.74	-0.19	0.96	-0.07	0.92	0.97	-0.88	0.68	0.93	0.85
Ca				/	0.08	0.26	0.95	0.06	0.97	0.99	0.77	-0.06	0.99	0.05	0.95	0.98	-0.84	0.66	0.98	0.75
Cd					/	0.60	0.26	0.51	-0.12	0.18	0.05	0.74	0.19	0.62	-0.08	0.01	0.27	-0.35	0.11	-0.17
Co						/	0.43	0.89	0.07	0.36	0.63	0.91	0.35	0.93	0.06	0.11	0.16	-0.38	0.21	-0.09
Cu							/	0.23	0.89	0.97	0.81	0.17	0.97	0.27	0.80	0.89	-0.69	0.43	0.89	0.75
Fe								/	-0.11	0.15	0.38	0.80	0.14	0.77	-0.13	-0.07	0.17	-0.40	-0.00	-0.20
K									/	0.94	0.72	-0.26	0.94	-0.13	0.95	0.98	-0.90	0.74	0.94	0.83
Li										/	0.80	0.05	1.00	0.15	0.91	0.96	-0.78	0.58	0.96	0.74
Mg											/	0.34	0.80	0.51	0.65	0.67	-0.45	0.24	0.71	0.56
Mn												/	0.05	0.97	-0.28	-0.21	0.52	-0.67	-0.11	-0.29
Na													/	0.15	0.91	0.96	-0.78	0.58	0.96	0.73
Ni														/	-0.18	-0.12	0.44	-0.62	-0.02	-0.20
P															/	0.98	-0.90	0.85	0.98	0.73
S																/	-0.90	0.77	0.99	0.78
Sb																	/	-0.90	-0.85	-0.76
Se																		/	0.75	0.65
Sr																			/	0.73
Zn																				/

Table 5. Pearson's correlation coefficient (*r*) for the relationship between macro-, micro-, and toxic elements of soybean, buckwheat, spelt, and maize grains grown under different conditions.

	OS16	CS16	OM16	CM16	OSo16	CSo16	OB16	CB16
OS16	/	1.000	0.997	0.996	0.971	0.972	0.990	0.992
CS16		/	0.997	0.996	0.971	0.973	0.992	0.993
OM16			/	0.999	0.954	0.956	0.989	0.990
CM16				/	0.951	0.955	0.993	0.993
OSo16					/	0.998	0.964	0.969
CSo16						/	0.972	0.976
OB16							/	0.999
CB16								/

(manganese, zinc, iron, and nickel). Maize grains had significantly lower calcium content as compared to all other investigated samples. Comparing the obtained results with nutritional recommendations for daily intake of elements, the following theoretical assessment can be made: all examined grain samples (except maize) are good sources of manganese and lithium for human nutrition, while soybean and buckwheat grains are good sources of copper. In addition, spelt and soybean grains are rich in selenium, while buckwheat grains are rich in iron. The presence of some potentially toxic elements (especially cadmium and strontium) was determined, thus must be monitored. The present work demonstrated that there was no regularity to confirm which of the two breeding systems would provide a better mineral composition of the grains.

Acknowledgement

The present work was financially supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia (grant no.: 451-03-68/2022-14/200116 and 451-03-68/2022-14/200168).

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