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Nikolett Czipa, Dávid Andrási, Béla Kovács

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Determination of essential and toxic elements in Hungarian honeys

Nikolett Czipa*, Dávid Andrási, Béla Kovács

Institute of Food Science

Faculty of Agricultural and Food Sciences and Environmental Management

University of Debrecen, H-4032 Böszörményi Street 138. Debrecen, Hungary

Abstract

The aim of this present study was determination of essential and toxic element concentrations in 34 mono- and multi-floral honey samples from four geographical regions of Hungary, and examination of the connection between the floral origin and the element content. Ten elements (Al, Ca, Cu, Fe, K, Mg, Mn, P, S and Zn) were identified by ICP-OES and six (As, Cd, Cr, Mo, Pb, Se) were analysed by ICP-MS. Potassium, calcium, and phosphorus were the most abundant elements with mean concentrations of 372, 47.9 and 44.3 mg kg^{-1}, respectively. The essential element content was very low in the analysed samples and generally below literature values. The concentrations of toxic elements were sufficiently low as to pose no risk to human health. The concentrations of aluminium, arsenic, cadmium and lead were low, with mean concentrations of 1028, 15.6, 0.746 and 45.5 µg kg^{-1}, respectively. Three honey groups (acacia, rape and sunflower) were distinguished by linear discriminant analysis from their element content.

Keywords: honey, essential element, toxic element, ICP-OES, ICP-MS

*Corresponding author.
Tel: +36 30 3662383
Fax: +36 52 417572
E-mail address: nikolettczipa@gmail.com

1. Introduction

Honey is a natural substance produced by *Apis mellifera* from flower nectar or honeydew. Honeys have an important role in human nourishment and medicine due to their excellent dietary, antibacterial, antifungal and antiviral effects. The antibacterial activity is related to the sugar and moisture content, pH, hydrogen-peroxide content and phytochemical factors of honey (Etaraf-Oskouei & Najafi, 2013). Honey helps protect against gastrointestinal infection (Alnaqdy et al., 2005). Diarrhea control may be assisted with diluted honey (5% v/v) because it promotes uptake of potassium and water uptake (Bansal et al., 2005). Honey contains a natural mixture of glucose and fructose and, thus, is a good source of carbohydrate for athletes (Bansal et al., 2005).

The major use of honey in healing is the treatment of wounds, suppression of inflammation, clearance of infections and minimization of scarring (Bogdanov et al., 2008; Basualdo et al., 2007). Al-Waili (2001) demonstrated that topical application of crude honey could be effective in treating seborrheic dermatitis and hair loss.

The quantity of honey produced in Europe during 2010 has been estimated to be in excess of 203,571 tonnes (FAO). Hungary is one of the biggest honey producers in the EU with an annual production of 16,500 tonnes. Honey is a very complex food containing many essential nutrients, in particular amino acids, vitamins, organic acids, enzymes and minerals. The composition and properties of honeys depend on the botanical origin of the nectar or honeydew, the harvesting treatment and storage. Honey has a low mineral content (0.1-0.2% in nectar honeys and about 1.0% in honeydew honeys), which varies depending on the soil conditions and extraction techniques (Hernandez et al., 2005). Dark honey types contain higher levels of minerals (Lachman et al., 2007). Honey is useful for collecting information about the environment within the bees' "collecting area", which is about 7 km^2 (Almeida-

Silva et al., 2011; Crane, 1984). The major elements in honey are potassium, calcium, magnesium and sodium, whilst the minor elements are cadmium, chromium, cobalt, copper, iron, lead, manganese and nickel (Andrade et al., 2014). Accumulation of metals may derive from external sources such as industrial pollution, incorrect treatment and agrochemicals (Bratu & Beorgescu, 2005).

Chromium, manganese, selenium, sulphur, boron, potassium, calcium and fluorine are particularly important in human nutrition. Codex Alimentarius Hungaricus contains Recommended Dietary Allowance (RDA) values for adults, and maximum intakes for toxic elements (e.g. arsenic, lead, cadmium) were determined from WHO Technical Reports (TRS 959 (As), TRS 960 (Pb), TRS 960 (Cd)).

The aims of this study were to: (i) identify and quantify the toxic and essential elements in 34 Hungarian mono-floral and multi-floral honeys; and (ii) determine the floral origin (acacia, rape or sunflower) based on the element content of samples.

2. Materials and methods

2. 1. Samples

Thirty-four mono- and multi-floral honey samples were examined in this study (Table 1). The botanical origins of the mono-floral honeys were: acacia (*Robinia pseudoacacia*), rape (*Brassica sp.*), sunflower (*Helianthus annuus*), linden (*Tilia sp.*), hawthorn (*Crataegus sp.*), silk grass (*Aslepias sp.*), facelia (*Phacelia tanacetifolia*) and vetch (*Vicia sp.*). The samples were collected from beekeepers in eight counties of Hungary (Békés, Csongrád, Fejér, Hajdú-Bihar, Jász-Nagykun-Szolnok, Somogy, Tolna, Veszprém) (Table 1.). All honey samples (500

g) were collected and stored in new, sterile glass jars at room temperature in the dark until analysis.

2.2. Reagents and solutions

All chemicals were of analytical grade or better. Nitric acid (69% v/v) and hydrogen-peroxide (30% v/v) were from VWR International Ltd. (Radnor, USA). Ultrapure water produced by a Milli-Q water purification system (Millipore SAS, Molsheim, France) was used to prepare of solutions and dilutions. The element standard solutions were prepared from mono-elemental standard solution (1000 mg/l; Scharlab S.L., Barcelona, Spain).

2.3. Determination of element content

The digestion of samples was carried out according to the method of Kovács et al. (1996), which had been validated using animal and plant materials in the Institute of Food Science accredited laboratory (ISO/IEC 17025:2005). Three grams of honey were added to 10 ml nitric acid (69% v/v) and the samples allowed to stand overnight. Samples were pre-digested at 60°C for 30 min. After cooling, the 3 ml hydrogen-peroxide (30% v/v) was added and the samples heated at 120°C for 90 min. After digestion, ultrapure water was added to make a final volume of 50 ml. Samples were homogenized and filtered using qualitative filter papers (Sartorius Stedim Biotech S.A., Gottingen, Germany). Depending on the anticipated concentration, determination of elements was carried out using ICP-OES (Inductively Coupled Plasma Optical Emission Spectrometry) (Thermo Scientific iCAP 6300, Cambridge, UK) or ICP-MS (Inductively Coupled Plasma Mass Spectrometry) (Thermo Scientific XSeries 2, Bremen, Germany). In view of the interference of ^{31}P and the ^{32}S in ICP-MS, these elements and those at higher concentrations (e.g. aluminium, calcium, copper, iron, potassium, magnesium, manganese and zinc) were determined by ICP-OES. Six elements (arsenic, cadmium, chromium, molybdenum, lead and selenium) were analysed by ICP-MS with a hexapole collision/reaction cell (CCT). Yttrium, rhodium and indium were used as internal standards (100 μg L^{-1}). The conditions for analysis and wavelengths used are reported in Table 2.

Detection limits (LOD) were determined for reagent blank samples (n=10) using the software for ICP-OES (iTEVA) at a confidence level of 99%. Detection limits of ICP-OES were: 0.004 mg kg^{-1} (Al), 0.048 mg kg^{-1} (Ca), 0.002 mg kg^{-1} (Cu), 0.005 mg kg^{-1} (Fe), 0.524 mg kg^{-1} (K), 0.104 mg kg^{-1} (Mg), 0.001 mg kg^{-1} (Mn), 0.489 mg kg^{-1} (P), 0.108 mg kg^{-1} (S) and 0.005 mg kg^{-1} (Zn), respectively.

In the case of ICP-MD, the LOD was determined using the following equation: LoD = 3 * $SD_{reagent\ blank}$ (n=10) / Sensitivity. Detection limits for ICP-MS were: 0.038 μg kg^{-1} (Cr), 0.019 μg kg^{-1} (As), 0.021 μg kg^{-1} (Se), 0.019 μg kg^{-1} (Mo), 0.003 μg kg^{-1} (Cd) and 0.048 μg kg^{-1} (Pb), respectively.

In the course of measuring three reference plant samples, the percentage recovery (%) was determined to be between 86.5 and 104 % for Al-Zn and between 90.1 and 109 for Cr-Pb.

2.4. Statistical analysis

All analytical analyses were carried out in triplicate. Data are described using general terms (mean, standard deviation, minimum and maximum values) and LDA (Linear Discriminant Analysis). SPSS for Windows (version 13; SPSS Inc. Chicago, Illinois, USA) was used for all calculations.

3. Results and discussion

3.1. Essential elements

The elemental contents of Hungarian honey samples (34) are shown in Table 3. The RDAs for essential elements were determined using Codex Alimentarius Hungaricus. According to this regulation (1-190/496) the contribution is important if 100 g contains at least 15% of the RDA.

Potassium was the most abundant element present in all analysed minerals. The concentration of this element ranged from 62.4±1.2 to 1158±51 mg kg $^{-1}$ in the vetch (V) and the forest (FO) honey. The average content was 372 mg kg^{-1}. Acacia honey samples showed the lowest values (181±22 mg kg^{-1}) followed by rape (332±144 mg kg^{-1}) and sunflower (439±65 mg kg^{-1}) honeys. The potassium content showed a wide variation across the range 154-755 mg kg^{-1} for flower honeys. The reason for this difference was the different mixing of mono-floral nectar or honeys. The concentration of this element in our acacia samples agrees with the measurements of acacia honeys from Serbia (Jevtić et al., 2012). The potassium content in our forest honey was lower when compared with other studies (Vanhanen et al., 2011 and Habib et al., 2013). According to the Codex Alimentarius Hungaricus the RDA for potassium is 2000 mg day^{-1} for adults.

Calcium was the second most abundant element in our samples with an average content of 47.9 mg kg^{-1}. The lowest calcium concentration (3.65±0.10 mg kg^{-1}) was found in an acacia honey (A8 sample); and a sunflower honey (S5 sample) showed the highest value (181±13 mg kg^{-1}). The concentration of this element was particularly high in sunflower honeys. The mean contents of calcium reported by Jevtić et al. (2012) and Alves et al. (2013) were 40.3

and 20.1 mg kg^{-1}, respectively. The calcium content in our samples was similar to that reported by Habib et al. (2013). The RDA for calcium is 800 mg day^{-1} for adults.

Phosphorus concentration ranged from 19.7±0.2 mg kg^{-1} (SG sample) to 93.5±2.3 mg kg^{-1} (FO sample) with an average value of 44.3 mg kg^{-1}. The highest concentration of this element in our samples was lower than the values found by Habib et al. (2013), Vanhanen et al. (2011) and Batista et al. (2012). The RDA for phosphorus is 700 mg day^{-1} for adults.

The range and the mean value of the magnesium concentration of samples were 1.91-35.1 (FA and F8 samples) and 16.3 mg kg^{-1}. No magnesium was found in sample F9. The LOD was 0.104 mg kg^{-1}. The measured values were lower than 10 mg kg^{-1} in seven samples (A5, A6, A7, F4, F7, F9, FA samples). The observed concentrations were lower than those found in New Zealand (Vanhanen et al., 2011) and Brazilian honeys (Batista et al., 2012), but higher than in Serbian honeys (Jevtić et al., 2012). The RDA for magnesium is 375 mg day^{-1} for adults.

The average content of sulphur was 20.6 mg kg^{-1}. The lowest and highest concentrations were measured in A8 sample (6.93±0.08) and in FO sample (76.1 mg kg^{-1}). In New Zealand honey samples similar results were determined (Vanhanen et al., 2011).

The iron content ranged from 0.108 (V sample) to 2.86 mg kg^{-1} (R2 samples) with an average content of 0.760 mg kg^{-1}. Iron was not detected in four samples (A5, A7, S3, F9). The LOD was 0.005 mg kg^{-1}. The iron concentrations in our samples were lower than those reported in honey samples from United Arab Emirates (Habib et al., 2013), Turkey (Yücel & Sultanoğlu, 2013) and Brazil (Batista et al., 2012). In other studies (Jevtić et al., 2012 and Vanhanenet al., 2011), the concentrations obtained for iron were similar to our results. The RDA for iron is 14 mg day^{-1} for adults. The PMTDI (Provisional Maximum Tolerable Daily Intake) is 0.8 mg kg^{-1} bw (body weight) (FAO, 1983).

Copper was not detected in two samples (F9, FA). The LOD was 0.002 mg kg^{-1}. The lowest and highest contents measured were 0.018±0.001 mg kg^{-1} (A6 sample) and 0.783±0.073 mg kg^{-1} (FO sample) respectively. The mean content in all honeys samples was 0.189 mg kg^{-1}. These results were similar to the Brazilian and New Zealand honeys analysed by Batista et al. (2012) and Vanhanen et al. (2011). Copper content was much higher in Turkish honeys (Yücel & Sultanoğlu, 2013). This is an essential element for humans (RDA=1.0 mg day^{-1} for adults), however, it may cause Wilson's disease when in excess. The PMTDI is 0.5 mg kg^{-1} bw (FAO, 1982). None of the Hungarian samples contained toxic quantities of copper.

Manganese content ranged from 0.026 (V sample) to 4.23 mg kg^{-1} (FO samples) with an average value of 1.03 mg kg^{-1}. The manganese concentrations in our samples were similar to most literature values (Yücel & Sultanoğlu, 2013 and Vanhanen et al., 2011), but some higher values have been reported, e.g. in Brazilian honeys (Batista et al., 2012). The RDA for manganese is 2 mg day^{-1} for adults.

The lowest and highest zinc content was measured in A5 and R2 samples (0.185±0.028 and 7.20±0.08 mg kg^{-1}). The mean content was 2.32 mg kg^{-1}, which is similar to that reported by Jevtić et al. (2012), Habib et al. (2013) and Batista et al. (2012). The RDA for zinc is 10 mg day^{-1} for adults. The PMTDI is 0.3-1.0 mg kg^{-1} bw. Zinc is an essential element for man but the high levels of dietary zinc can cause anaemia and decreased copper and iron absorption. (WHO, 1982).

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The molybdenum content ranged from 2.15±0.05 (A5 sample) to 66.2±6.5 μg kg^{-1} (R4 sample) with an average value of 27.8 μg kg^{-1}. The concentration of this element was higher in New Zealand honey samples (Vanhanen et al., 2011). The RDA is 50 μg day^{-1} for adults.

The mean content of chromium and selenium was similar, with values of 13.3 and 13.2 μg kg^{-1}, respectively. The lowest content was measured in the S4 sample (4.80±0.45 μg kg^{-1}) for chromium and in the A5 sample (2.66±0.33 μg kg^{-1}) for selenium. The linden honey showed the highest chromium concentration (36.7±3.8 μg kg^{-1}) and the A1 sample showed the highest selenium content (36.4±0.8 μg kg^{-1}). Results for chromium in New Zealand honeys (Vanhanen et al., 2011) were higher than ours. The RDA for chromium is 40 μg day^{-1} for adults.

Considering the element content of other studies (Table 4), the Hungarian honeys showed low element content, except calcium. Vanhanen et al. (2011) measured similar results in New Zealand honeys for sulphur, copper, iron and manganese. Zinc concentration was very similar in our and other honeys from Brazil, Serbia and United Arab Emirates (Batista et al., 2012; Jevtić et al., 2012 and Habib et al., 2013).

The calculated daily mineral intake for 20 grams of honeys is very low. As a percentage of RDA, the contributions from our highest measured concentrations were as follows: 1.16% for potassium, 0.45% for calcium, 0.27% for phosphorus, 0.19% for magnesium, 0.41% for iron, 1.57% for copper, 4.23% for manganese, 1.44% for zinc, 1.84% for chromium, 2.65% for molybdenum and 1.32% for selenium.

3.2. Toxic elements

Aluminium, arsenic, cadmium and lead are present in the environment, but the anthropogenic sources are very important, e.g. industrial or urban discharges, mining and agricultural output (Zhang & Wong, 2007). Honey is a very good environmental indicator because it reflects the toxic element content of water, soil and air. One aim of this study was to determine the toxic trace element content in honey samples from different regions of Hungary. The measured toxic element content in our samples is shown in Table 5.

Arsenic concentration ranged from 3.19±0.29 to 30.4±0.3 µg kg^{-1}, the mean content in all honey samples was 15.9±7.84 µg kg^{-1}. The highest and the lowest values were measured in Hajdú-Bihar County (A5, R4 samples). The concentration of this trace element was higher in the Northeast Croatian honeys (Bilandžić et al., 2011). The PTWI (Provisional Tolerable Weekly Intake) for inorganic arsenic has been withdrawn (WHO, 2011b). According to the thirty-third meeting of JECFA the PTWI for arsenic was 15 µg kg^{-1} bw (WHO, 1988). Most inorganic arsenic is found in cereals, rice, meats and milk products; the arsenic content of examined honey samples was negligible. In Hungary, the main source of arsenic is drinking water, and is an established problem in Alföld. The most significant problem is in Békés County but the measured arsenic content of honey samples from this county did not verify this problem.

Cadmium levels ranged from 0.032 to 3.31 µg kg^{-1}, the lowest and the highest content of this trace element were found in Jász-Nagykun-Szolnok County (V, F8). In ten samples no cadmium was detected (A4, A5, R5 samples from Hajdú-Bihar; A7, R6, F7, FA samples from Békés; F2 sample from Fejér; F9 sample from Veszprém and H sample from Somogy County). The LOD was 0.003 µg kg^{-1}. The levels of cadmium reported by Andrade et al. (2014) and Vanhanen et al. (2011) were higher than in our samples. The PTMI for cadmium is 25 µg kg^{-1} bw (WHO, 2013). The main sources of cadmium are kidney, mushrooms, oil seeds, cereals and fish, but cadmium can also get into the food from low quality ceramic tableware and plastics.

The highest lead concentration was measured in Hajdú-Bihar (R5 sample) and Békés Counties (F7 sample), with average values of 122 and 133 µg kg^{-1}. The lowest lead concentration (11.2±8.1 µg kg^{-1}) was measured in a rape honey from Békés County (R6 sample). The mean content in all honey samples was 45.4 µg kg^{-1}. In comparison to the literature data, the measured lead concentration was lower (Yücel & Sultanoğlu, 2013; Andrade et al., 2014; Bilandžić et al., 2011). The PTWI for lead was 25 µg kg^{-1} bw (WHO, 1999), but there is no new PTWI, because it is "not possible to establish a new PTWI that would be considered health protective" (WHO, 2011a).

The measured aluminium concentration ranged between 0.103-4.39 mg kg^{-1}. This element was not detected in the A6 sample. The lowest values (0.103 mg kg^{-1}) were measured in two acacia honey samples (A5 and A7) from Hajdú-Bihar and Békés Counties. The highest aluminium content was found in vetch honey from Jász-Nagykun-Szolnok County. The aluminium results measured in this work were lower, when compared with the results obtained by other authors, such as Yücel & Sultanoğlu (2013) and Vanhanen et al. (2011). The PTWI value of aluminium is 1 mg kg^{-1} bw (WHO, 2006).

The tolerable daily intake (TDI) for a 60 kg weight adult is 8.57 mg for aluminium and 50 µg for cadmium. Aluminium and cadmium were 1.02% and 0.13% of the TDI in 20 grams vetch (V) and 20 grams flower (F8) honeys, which contained the highest aluminium and cadmium concentrations. The PTWI values for arsenic and lead have been withdrawn, however, considering the previously used PTWI value, the measured concentrations were below 1.0% of TDI for arsenic. The highest calculated value was 5.32% of TDI in 20 grams flower honey (F8) from Békés County.

Compared to other studies (Table 6) the concentration of aluminium was found to be lower in our samples. The cadmium content was higher in samples from different countries, except the honeys from Brazil. Lead content was much higher in the Turkish and Croatian honeys (Yücel & Sultanoğlu, 2013; Bilandžic et al., 2011).

3.3 Linear discriminant analysis (LDA)

The LDA was used for the categorization of our samples. The grouping variables were the types of honey (acacia, rape and sunflower) and the independent variables were the elements (calcium, copper, iron, potassium, magnesium, manganese, phosphorus, sulphur and zinc). Two discriminant functions were determined. Figure 1 shows the scatter plot of honey samples defined by the discriminant functions. The first discriminant function involved the following elements: phosphorus, calcium, copper, manganese, and the second contained the other elements. Based on the eigenvalue the first discriminant function was more important to the classification of the cases of independent variables than the second one. The first discriminant function explained 92% of the variance of independent variables; this value was 83% for the second one. Both discriminant functions were significant. The first group (acacia honeys) had high values in the first dimension (1.399), but showed low values in the second dimension (-2.182). The second group (rape honeys) showed high values in both dimensions (2.415 and 2.518). The third group (sunflower honeys) had very low values in the first dimension (-5.136), but showed relative high values in the second one (0.470). The sunflower honeys formed a clearly defined group at the left side of the plot. However the acacia and rape honeys did not show big difference in the first dimension; these two honey types showed a significant difference in the second one. Validation was carried out using a cross-validation method. The program, based on the independent variables, correctly classified 89.5% of the cross-validated grouped cases.

The LDA was successfully applied to the separation of these three honey types. Knowing the element content the LDA can be a useful method for the determination of the botanical origin of mono-floral honeys.

4. Conclusion

In this study 16 elements were measured in 34 honey samples from eight counties of Hungary in four geographical areas (North Alföld, South Alföld, Middle Transdanubium, South Transdanubium), which corresponded to two big regions (Alföld and Transdanubium). The concentration of essential elements decreased in the following order: K>Ca>P>S>Mg>Zn>Mn>Fe>Cu. The aluminium, copper, iron, potassium, magnesium, manganese, phosphorus, sulphur and selenium showed higher concentrations in samples from Transdanubium. Concentrations of the other elements varied across the four geographical areas. The highest concentration of arsenic and chromium was measured in the honey of the South Transdanubium, the highest lead content in Middle Transdanubium and the highest cadmium and molybdenum content in North Alföld. The toxic element concentration in our samples decreased in the following order: Al>Pb>As>Cd. The concentration of these elements was very low in our samples; thus the consumption of these honeys does not present a problem for human health. The essential element content of honey was very low and, therefore, the average daily consumption of honey (about 20 grams) contains only 1% of the RDA values of essential elements.

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Figure captions

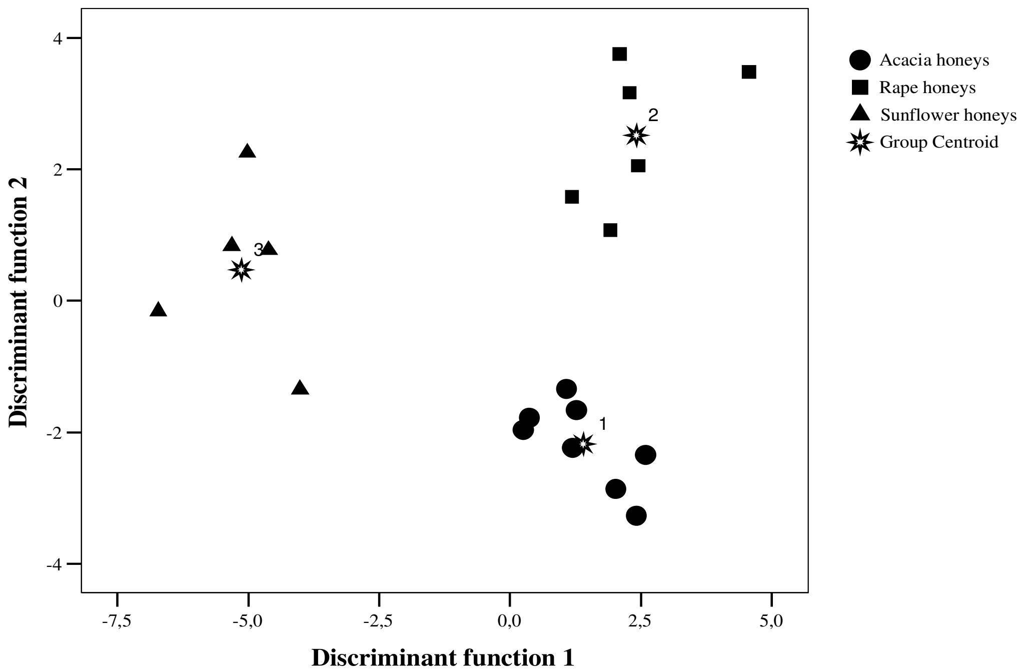

Figure 1. Linear discriminant analysis of mono-floral honey samples

Tables

Table 1
Studied honey samples

Sample No.	Botanical origin	County	Geographical areas
A1	Acacia	Somogy	South Transdanubium
A2	Acacia	Jász-Nagykun-Szolnok	North Alföld
A3	Acacia	Fejér	Middle Transdanubium
A4	Acacia	Hajdú-Bihar	North Alföld
A5	Acacia	Hajdú-Bihar	North Alföld
A6	Acacia	Csongrád	South Alföld
A7	Acacia	Békés	South Alföld
A8	Acacia	Jász-Nagykun-Szolnok	North Alföld
R1	Rape	Somogy	South Transdanubium
R2	Rape	Fejér	Middle Transdanubium
R3	Rape	Jász-Nagykun-Szolnok	North Alföld
R4	Rape	Hajdú-Bihar	North Alföld
R5	Rape	Hajdú-Bihar	North Alföld
R6	Rape	Békés	South Alföld
S1	Sunflower	Somogy	South Transdanubium
S2	Sunflower	Csongrád	South Alföld
S3	Sunflower	Fejér	Middle Transdanubium
S4	Sunflower	Hajdú-Bihar	North Alföld
S5	Sunflower	Békés	South Alföld
F1	Multifloral	Somogy	South Transdanubium
F2	Multifloral	Fejér	Middle Transdanubium
F3	Multifloral	Hajdú-Bihar	North Alföld
F4	Multifloral	Hajdú-Bihar	North Alföld
F5	Multifloral	Tolna	South Transdanubium
F6	Multifloral	Csongrád	South Alföld
F7	Multifloral	Békés	South Alföld
F8	Multifloral	Jász-Nagykun-Szolnok	North Alföld
F9	Multifloral	Veszprém	Middle Transdanubium
L	Linden	Somogy	South Transdanubium
H	Hawthorn	Somogy	South Transdanubium
SG	Silk grass	Tolna	South Transdanubium
FA	Facelia	Békés	South Alföld
FO	Forest	Veszprém	Middle Transdanubium
V	Vetch	Jász-Nagykun-Szolnok	North Alföld

Table 2
Operating parameters of ICP-OES and ICP-MS

Parameters (ICP-OES)		*Parameters (ICP-MS)*	
Operating power	1350 W	Rf power	1400 W
Plasma gas flow rate	12 l min^{-1}	Plasma gas flow rate	14.0 l min^{-1}
Auxiliary gas flow rate	1.0 l/min^{-1}	Auxiliary gas flow rate	1.0 l min^{-1}
Nebulizer gas flow rate	0.75 l/min^{-1}	Nebulizer gas flow rate	0.9 l min^{-1}
Rinsing time	35 sec	Spray chamber temperature	
Rinsing pump speed	75 rpm		2 °C
Stabilization time	3.0 sec	Sample uptake rate	0.5 ml min^{-1}
Transfer pump speed	50 rpm	CCT gas	7% H_2 in He
Integration time		Dwell time	1000 ms
- Low WL range	10 sec	Sweeps	11
- High WL range	10 sec	Main runs	3
Elements	*Wavelengths (nm)*	*Elements*	*Measured isotope (amu)*
Aluminium	361.153	Arsenic	75
Calcium	413.764	Cadmium	111
Copper	224.700	Chromium	52
Iron	259.940	Molybdenum	95
Potassium	766.490	Lead	208
Magnesium	285.213	Selenium	80
Manganese	191.510		
Phosphorus	213.617	Yttrium	89
Sulphur	181.975	Rhodium	103
Zinc	202.548	Indium	115

Table 3
Essential element contents in our honey samples

Sample	**Statistics**	**Cr (µg kg^{-1})**	**Mo (µg kg^{-1})**	**Se (µg kg^{-1})**	**Ca (mg kg^{-1})**	**Cu (mg kg^{-1})**	**Fe (mg kg^{-1})**
Acacia (n=8)	Mean±SD	14.5	33.8	15.7	23.6	0.130	0.429
	Range	6.44-33.9	2.15-65.1	2.66-36.4	3.65-43.2	0.018-0.317	<0.005-0.997
Rape (n=6)	Mean±SD	11.7	31.8	12.5	51.2	0.164	1.35
	Range	7.44-15.1	6.73-66.2	3.68-27.3	23.7-79.8	0.033-0.370	0.096-2.86
Sunflower (n=5)	Mean±SD	10.1	29.2	14.3	111	0.270	0.648
	Range	4.80-14.7	4.86-52.0	7.55-19.1	58.2-181	0.098-0.507	<0.005-1.48
Flower (n=9)	Mean±SD	13.7	18.6	10.5	46.0	0.160	0.778
	Range	9.11-21.2	3.30-58.5	3.10-21.2	13.1-118	<0.002-0.333	<0.005-2.69
Linden (n=1)	Mean±SD	36.7±3.8	31.8±1.2	21.9±1.7	45.7±1.4	0.320±0.073	0.612±0.289
Hawthorn (n=1)	Mean±SD	17.6±1.4	30.1±2.3	13.3±0.8	21.4±2.0	0.072±0.014	0.227±0.030
Silk grass (n=1)	Mean±SD	8.80±1.29	15.6±1.6	8.19±1.04	37.5±2.1	0.268±0.034	0.534±0.377
Facelia (n=1)	Mean±SD	5.95±0.45	25.9±1.7	14.6±1.2	4.24±0.51	<0.002	0.225±0.007
Forest (n=1)	Mean±SD	9.64±0.86	38.3±2.7	15.1±1.1	51.3±2.0	0.783±0.073	2.36±0.19
Vetch (n=1)	Mean±SD	13.5±1.2	27.9±0.3	8.65±0.68	4.41±0.56	0.182±0.005	0.108±0.006
All samples (n=34)	Mean±SD	13.3±6.9	27.8±17.8	13.2±7.38	47.9±40.0	0.189±0.168	0.760±0.758
	Range	4.80-36.7	2.15-66.2	2.66-36.4	3.65-181	<0.002-0.783	<0.005-2.86
Sample	**Statistics**	**K (mg kg^{-1})**	**Mg (mg kg^{-1})**	**Mn (mg kg^{-1})**	**P (mg kg^{-1})**	**S (mg kg^{-1})**	**Zn (mg kg^{-1})**
Acacia (n=8)	Mean	181	12.8	0.837	26.9	8.90	1.58
	Range	130-197	3.41-23.2	0.029-1.69	22.0-35.1	6.93-12.0	0.185-3.92
Rape (n=6)	Mean	332	17.7	0.614	42.2	23.2	3.66
	Range	178-587	10.1-34.1	0.136-1.68	31.4-68.2	16.4-43.0	0.527-7.20
Sunflower (n=5)	Mean	439	22.4	1.10	66.1	23.7	3.35
	Range	376-539	11.3-32.3	0.179-2.28	48.2-89.2	17.3-33.5	0.760-6.66
Flower (n=9)	Mean	371	14.7	1.16	53.0	23.6	2.02
	Range	154-755	<0.104-35.1	0.101-1.72	23.9-89.1	9.02-51.5	0.573-3.69
Linden (n=1)	Mean±SD	955±4	28.6±1.5	1.36±0.06	26.0±0.2	23.5±1.9	2.15±0.06
Hawthorn (n=1)	Mean±SD	1112±123	14.5±1.9	1.35±0.05	32.9±0.7	30.7±1.6	0.959±0.098
Silk grass(n=1)	Mean±SD	237±4	11.5±0.1	1.63±0.12	19.7±0.2	11.5±1.0	3.87±0.18
Facelia (n=1)	Mean±SD	143±2	1.91±0.06	0.031±0.001	31.6±2.1	8.58±0.01	0.535±0.054
Forest (n=1)	Mean±SD	1158±51	31.7±0.1	4.23±0.05	93.5±2.3	76.1±2.1	1.18±0.00
Vetch (n=1)	Mean±SD	62.4±1.2	12.9±12.9	0.026±0.001	27.8±0.1	9.07±0.10	0.632±0.082
All samples (n=34)	Mean±SD	372±270	16.3±9.67	1.03±0.89	44.3±22.9	20.6±14.5	2.32±1.84
	Range	62.4-1158	<0.104-35.1	0.026-4.23	19.7-93.5	6.93-76.1	0.185-7.2

SD = standard deviation

Table 4
Comparison between essential element contents in Hungarian honey and honey from other countries

Elements	**Present study (n=34)**	**Brazil (Batista et al., 2012)**	**Turkey (Yücel & Sultanoğlu, 2013)**	**Portugal (Alves et al., 2013)**	**Serbia (Jevtić et al., 2012)**	**New Zealand (Vanhanen et al., 2011)**	**United Arab Emirates (Habib et al., 2013)**
Cr (μg kg^{-1})	4.80-36.7	-	100-540	-	-	120-550	-
Mo (μg kg^{-1})	2.15-66.2	-	-	-	-	100	-
Se (μg kg^{-1})	2.66-36.4	-	-	-	-	-	-
Ca (mg kg^{-1})	3.65-181	-	-	0.43-72.3	29-54.6	7.21-94.3	7.87-184
Cu (mg kg^{-1})	<0.002-0.783	0.01-0.7	0.58-9.52	-	-	0.09-0.70	0.26-1.91
Fe (mg kg^{-1})	<0.005-2.86	0.5-14.1	7.40-92.38	<0.70-7.06	0.58-4.21	0.67-3.39	1.15-111
K (mg kg^{-1})	62.4-1158	-	-	30.9-1441	62.8-212	34.8-3640	86.0-2690
Mg (mg kg^{-1})	<0.104-35.1	12.4-360	-	3.05-82.2	1.23-11.2	7.52-86.3	2.28-93.0
Mn (mg kg^{-1})	0.026-4.23	0.08-18.8	0.13-5.61	-	0.00-0.62	0.18-4.75	<0.01-10.3
P (mg kg^{-1})	19.7-93.5	8-486	-	-	-	29.5-255	8.99-264
S (mg kg^{-1})	6.93-76.1	-	-	-	-	13.4-93.9	6.97-334
Zn (mg kg^{-1})	0.185-7.20	0.01-7.1	1.02-30.67	-	1.41-6.51	0.20-2.46	0.30-6.73

Table 5
Toxic element contents in our honey samples

Sample	**Statistics**	**Al (mg kg^{-1})**	**As (μg kg^{-1})**	**Cd (μg kg^{-1})**	**Pb (μg kg^{-1})**
Acacia (n=8)	Mean±SD	0.504	17.8	0.584	30.4
	Range	<0.004-1.42	3.19-26.5	<0.003-2.20	16.8-40.7
Rape (n=6)	Mean±SD	1.12	18.8	0.736	30.4
	Range	0.137-3.36	8.95-30.4	<0.003-2.92	16.8-40.7
Sunflower (n=5)	Mean±SD	0.810	19.2	1.15	49.3
	Range	0.128-1.54	6.15-27.8	0.90-1.62	26.7-66.1
Flower (n=9)	Mean±SD	0.827	11.7	0.688	51.8
	Range	0.264-2.28	4.66-21.2	<0.003-3.31	23.4-133
Linden (n=1)	Mean±SD	1.20±0.05	22.8±0.1	1.25±0.16	61.4±5.2
Hawthorn (n=1)	Mean±SD	1.49±0.02	10.9±0.3	<0.003	37.8±0.2
Silk grass (n=1)	Mean±SD	0.940±0.054	11.7±0.8	0.31±0.04	37.3±2.4
Facelia (n=1)	Mean±SD	2.60±0.22	7.86±0.57	<0.003	20.9±3.0
Forest (n=1)	Mean±SD	2.07±0.08	19.0±1.0	2.77±0.29	74.8±0.2
Vetch (n=1)	Mean±SD	4.39±0.14	11.0±0.7	0.032±0.001	33.9±0.9
All samples n=34	Mean±SD	1.03±0.98	15.9±7.4	0.746±0.889	45.4±27.0
	Range	<0.004-4.39	3.19-30.4	<0.003-3.31	11.2-133

SD = standard deviation

Table 6
Comparison of toxic element contents in honeys from Hungary and from other countries

Elements	Present study	Brazil (Batista et al., 2012)	Brazil (Andrade et al., 2014)	Turkey (Yücel & Sultanoğlu, 2013)	Croatia (Bilandžić et al., 2011)	New Zealand (Vanhanen et al., 2011)
Al (mg kg^{-1})	<0.004-4.39	0.23-7.40	-	2.54-11.6	-	0.21-21.3
As (μg kg^{-1})	3.19-30.4	-	-	-	4.00-105	40.0-170
Cd (μg kg^{-1})	<0.003-3.31	<0.02-1.1	<2.0-8.0	-	1.00-24.0	10.0-450
Pb (μg kg^{-1})	11.2-133	1.2-31.4	141-228	60-2,020	10.0-841	10.0-40.0

HIGHTLIGHTS

(**Determination of essential and toxic elements in Hungarian honeys)**

1. We measure the element content of Hungarian honeys.
2. We compare the element content of Hungarian honeys with international honeys.
3. We measure the toxic and essential element content in our honey samples.
4. We determine the effect of the geographical origin.
5. We separated the honey types based on their element content.