Analysis of trace-elements and toxic heavy metals in honeys from Tlemcen Province, north-western Algeria

Dalila BEREKSI-REGUIG[1]
Hocine ALLALI[1] (✉)
Salim BOUCHENTOUF[1,2]
Agnieszka ADAMCZUK[3]
Grażyna KOWALSKA[4]
Radosław KOWALSKI[4]

Summary

Eighteen honeys collected from various botanical and geographical sources in Tlemcen Province (north-western Algeria) were studied to determine the presence of the following fifteen trace elements and heavy metals: K, Na, Ca, Mg, Mn, Cu, Fe, Zn, V, Cr, Co, As, Cd, Pb and W. Element determination was performed by inductively coupled plasma-mass spectrometry (ICP-MS), after digestion, by concentrated nitric acid using microwave mineralizer. The most abundant minerals were K, Ca, Mg and Na ranging within 153-989 mg/kg, 33.1-377 mg/kg, 69.1-162 mg/kg and 13.3-146 mg/kg, respectively. Fe was the most abundant heavy metal followed by Mn, Zn and Cu while Cr, Co, V, W, As, Cd and Pb were the lowest trace elements detected at level < 1 mg/kg in the honey samples surveyed. The variation in the mineral content in the honey samples studied is probably due to geochemical and geographical differences. Multivariate methods were used in order to classify honey samples according to their mineral content. The present study revealed that honeys collected from the various beekeepers apiaries of Tlemcen Province were not contaminated with toxic trace elements and therefore poses no threat to consumers.

Key words

honey, trace element, heavy metal, microwave digestion, ICP-MS

[1] Laboratory of Natural and Bioactive Substances (LASNABIO), Department of Chemistry, Faculty of Sciences, Abou Bekr Belkaïd University, P.O. Box 119, Tlemcen 13000, Algeria

[2] Doctor Tahar Moulay University of Saida, Algeria, BP 138 cité EN-NASR, Saïda 20000, Algeria

[3] Institute of Agrophysics, Polish Academy of Sciences, Doświadczalna 4, 20-290 Lublin, Poland

[4] Department of Analysis and Evaluation of Food Quality, University of Life Sciences in Lublin, 8 Skromna Street, 20-704 Lublin, Poland

✉ Corresponding author: h_allali72@yahoo.fr

Received: March 24, 2020 | Accepted: June 3, 2020

Introduction

Honey is a substance widely suggested to different investigations by many researchers during the last decades in terms of its nutritive and therapeutic aspect (Bogdanov, 2010; Khan et al., 2007, 2017; Mandal and Mandal, 2011). Several studies from Poland (Grembecka and Szefer, 2013), Italy (Conti et al., 2007), Turkey (Altun et al., 2017), Spain (de Alda-Garcilope et al., 2012), France (Devillers et al., 2002), Slovenia (Golob et al., 2005), Croatia (Bilandžić et al., 2012; Lasić et al., 2018), Argentina (Cantarelli et al., 2008), Brazil (dos Santos et al., 2008) and China (Ru et al., 2013) focused on honey composition in minerals and metals. Heavy metals may cause many damages to human health through various ways by accumulating in vital body organ such as liver, heart, kidney and brain (Rehman et al., 2018). Because of their persistence in the environment, heavy metals accumulate in human body mainly via food chain (Ali et al., 2019) causing harms to quality of human life (Jaishankar et al., 2014). Even if heavy metals are present naturally on earth crust, it has been well established that anthropogenic activities are the main sources introducing metals into the environment (Tchounwou et al., 2012). Volcanic eruption and weathering have also been reported as natural phenomena contributing significantly to metal pollution (Bradl, 2005). Many authors have indicated that the presence of mineral and heavy metals in honey depends closely on the geographical and botanical origin (Oroian et al., 2016; Pisani et al., 2008; Rashed and Soltan, 2004). Several researches reported that honey may be considered as a biological indicator of the environmental contaminants including heavy metal, low-level radioactivity and pesticide residues (Bogdanov et al., 2007; Formicki et al., 2013; Fredes and Montenegro, 2006; Nozal Nalda et al., 2005; Ponikvar et al., 2005). The objective of this study was to determine the level of 15 trace elements and heavy metals in 18 honey samples from Tlemcen Province in Algeria in order to evaluate the honey quality, which may be useful for assessing the presence of environmental contaminants. To the best of our knowledge and according to main research databases, we present here below the first study of honey bees samples from Tlemcen Province located in the western region of Algeria.

Materials and methods

Honey Samples

A total of eighteen *Apis mellifera intermissa* honey samples, namely S(1 - 18), were collected from twelve geographical locations (Sidi Djillali, El Aricha, Aïn Fezza, Beni Snous, Oued Chouly, Beni Mester, Beni Ghazli, Oued es Safsâf, Sebaa Chioukh, Hennaya, Remchi and Honaïne) in Tlemcen Povince in north west Algeria (Fig. 1 and Table 1). Samples were harvested from April 2017 to August 2018 directly from beekeepers. It is to highlight that the botanical origin of the honey samples studied was determined on the basis of a survey carried out among beekeepers. This study allowed to classify the honeys into monofloral and multifloral according to where the hives were established (Table 1). In the case of the monofloral honeys, their productions were possible by establishing hives where flowers of a particular plant species are dominant. Besides, multifloral honeys have several botanical sources where none is predominant. However, these results must be confirmed by pollen analyzes, which is widely used to verify the alleged geographic and floral origin of honey samples. Collected honeys were stored in dark conditions at + 4°C until analysis.

Inductively coupled plasma mass spectrometry (ICP-MS)

The inductively coupled plasma mass spectrometer ICP-MS Varian 820-MS (Varian, Australia) was used for the determination of Cr, Co, V, W, As, Cd and Pb of the honey samples. The ICP-MS parameters were: plasma gas flow 18 L/min, auxiliary gas flow 1.70 L/min, nebulizer gas flow 1 L/min, and frequency power 1.37 kW.

Atomic absorption spectroscopy (AAS)

The concentration of K, Na, Ca, Mg, Cu, Mn, Fe and Zn ions was determined by atomic absorption spectroscopy (AAS) method using a spectrometer SpectrAA 280 FS with autosampler SPS 3 (Varian, Australia), which was equipped with a deuterium lamp, hollow cathode lamp for each element and an air-acetylene burner. The Instrumental parameters related to each element are summarized in Table 2.

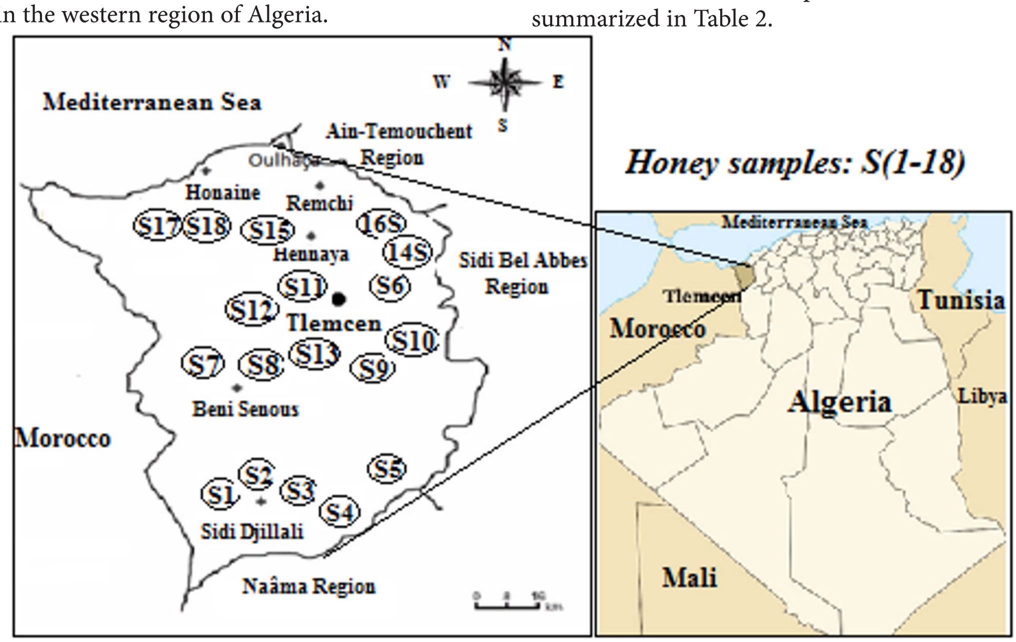

Figure 1. Geographical distribution of the sampling points in Tlemcen Province

Table 1. Honey samples and their botanical, geographical origin and production month/year

N°	Sample code	Arabic name	English name	Scientific name	Location	Month and year of harvest
1	S1	الخزامى البري	Lavender	*Lavandula vera* D.C.	Sidi Djillali	July 2018
2	S2	إكليل الجبل	Rosemary	*Rosmarinus officinalis* L.	Sidi Djillali	May 2018
3	S3	متعدد الزهور	Multifloral	Multifloral	Sidi Djillali	June 2018
4	S4	متعدد الزهور	Multifloral	Multifloral	Sidi Djillali	July 2017
5	S5	متعدد الزهور	Multifloral	Multifloral	El Aricha	August 2017
6	S6	خردل	Mild white mustard	*Sinapis alba* L.	Aïn Fezza	July 2017
7	S7	زعتر	Thyme	*Thymus vulgaris* L.	Beni Snous	June 2018
8	S8	شوك الجمل	Milk thistle	*Silybum marianum* (L.) Gaertn.	Beni Snous	July 2018
9	S9	متعدد الزهور	Multifloral	Multifloral	Oued Chouly	October 2017
10	S10	خروب	Carob tree	*Ceratonia siliqua* L.	Oued Chouly	September 2017
11	S11	زعتر	Thyme	*Thymus vulgaris* L.	Beni Mester	June 2017
12	S12	خروب	Carob tree	*Ceratonia siliqua* L.	Beni Ghazli	June 2017
13	S13	متعدد الزهور	Multifloral	Multifloral	Oued es Safsâf	August 2018
14	S14	متعدد الزهور	Multifloral	Multifloral	Sebaa Chioukh	June 2017
15	S15	متعدد الزهور	Multifloral	Multifloral	Hennaya	July 2017
16	S16	برتقال	Orange tree	*Citrus sinensis* L.	Remchi	April 2017
17	S17	متعدد الزهور	Multifloral	Multifloral	Honaïne	June 2018
18	S18	شوك الجمل	Milk thistle	*Silybum marianum* (L.) Gaertn.	Honaïne	July 2018

Table 2. Instrumental operation conditions used for elements analysis by AAS method

Elements	Flame type	Fuel flow (L/min)	Lamp current (mA)	Wave length (nm)	Slit width (nm)	Air flow (L//min)
Na	Air-acetylene	2.00	-	589.0	0.2	10.00
Mg	Air-acetylene	2.00	40	202.6	1.0	10.00
K	Air-acetylene	2.00	-	766.5	0.2	10.00
Ca	Air-acetylene	2.00	10	422.7	0.5	10.00
Mn	Air-acetylene	2.00	5	279.5	0.2	10.00
Fe	Air-acetylene	2.00	5	248.3	0.2	10.00
Cu	Air-acetylene	2.00	4	324.8	0.5	10.00
Zn	Air-acetylene	2.00	5	213.9	1.0	10.00

In order to avoid sample ionization during potassium analysis Schinkel buffer solution (mixture contents 10 g/L cesium chloride and 100 g/L lanthanum chloride) was used.

Reagents and solutions

Certified single-element standard solutions (1000 mg/L) used to prepare calibration curve were of highest purity grade (99.999%) and were purchased from Ultra Scientific (North Kingstown, RI, USA). The calibration standards for ICP-MS analysis were prepared by diluting certified standard solutions in 1% HNO_3 and for AAS analysis by diluting in high purity de-ionized Milli-Q water (Millipore, Bedford, MA, USA) purified by reverse osmosis followed by ion-exchange cartridges.

Preparation of Honey Sample

Digestion

Approximately 0.5 g of honey samples were placed in a Teflon vessel; 10 mL of 65% HNO_3 (suprapur grade®, Merck, Germany) was added into the vessel and the sealed vessel was put into microwave mineralizer MARS Express (CEM, USA). The microwave mineralization was performed stepwise at 400 W and 363 K, at 800 W and 393 K, at 1600 W and 483 K. The cooled digestion solution was then diluted to 50 mL using high purity de-ionized water. All sample solutions were clear before determination of the mineral content in honey sample.

Statistical method

Principal component analysis (PCA) and hierarchical cluster analysis (HCA) were performed using the XLStat 2014.5.03 software for Microsoft Excel (Addinsoft, Bordeaux, France) to rationalize and interpret the analytical data. The analyses of the eighteen honey samples involving the determination of the fifteen mineral elements resulted in a (18 × 15) data matrix using the 18 honey samples (objects) as rows and the elements (variables) as columns. The data were auto-scaled because the element concentrations are of different orders of magnitude. So, the data were evaluated employing the PCA technique. Fifteen dimensions are necessary for the total explanation of the data variability. In the case of the evaluation using HCA, these data were also auto-scaled using the single linkage method, and the Euclidean distances were used to calculate sample interpoint distances and similarities.

Results

Trace elements and heavy metals found in eighteen analyzed honeys, expressed in mg/kg fresh weight, collected from Tlemcen Province (northwest region of Algeria) are shown in Tables 3 as mean ± standard deviation (SD). The range of variation of each element was very wide in the samples from the different locations (Fig. 1 and Table 1). K The most abundant mineral element was K with a mean concentration of 540.5000 ± 240.8559 mg/kg. The highest concentration of trace-elements and heavy metals detected in honey samples was the potassium (989.0000 mg/kg) representing 61.30% of the total mineral content of the multifloral honey from Sebaa Chioukh region (S14). This essential element was followed by calcium, magnesium, sodium and iron with 23.37% (S14), 14.68% (S8), 10.49% (S6) and 5.38% (S1) of the total mineral content, respectively. The lowest levels were found for the following elements: Cd, W, Co, V, Pb and Cr at mean concentrations of 0.0012 ± 0.0018 mg/kg, 0.0017 ± 0.0021 mg/kg, 0.0072 ± 0.0056 mg/kg, 0.0102 ± 0.0176 mg/kg, 0.0201 ± 0.0288 mg/kg and 0.0486 ± 0.0162 mg/kg, respectively (Table 3). Mineral contents significantly differed in honey samples that belonged to the different studied locations. The mean levels of the essential elements (K, Ca, Mg, Na and Fe) in honeys samples from east Tlemcen Province (S6, S10 and S14) were higher than concentrations of these mineral elements in the honey samples from other locations (Table 1). Based on our results the lowest mean values for mineral elements belonged to honey samples S12, S7, S5 and S16 from Beni Ghazli, Beni Snous, El Aricha and Remchi locations, respectively. Due to the existence of different factors, such as botanical origins, soil characteristics, climates and pollution, which could affect the content of the trace elements and heavy metals in honey, the multivariate analysis and, in particular, PCA and HCA were applied to the elemental composition data in order to study the chemistry variables capable of characterizing the honey produced in each location (Fig. 2 and 3).

Table 3. Mean concentration, standard deviation and range (mg/kg) for 15 trace-elements and toxic heavy metals analyzed by ICP-MS and AAS method. Samples collected in Tlemcen Province S(1 - 18) during 2017 to 2018

N°	Elements	Min.	Max.	Mean±SD
1	$Cr^{b,c}$	0.0276	0.0874	0.0486±0.0162
2	$Co^{b,c}$	0.0004	0.0208	0.0072±0.0056
3	$V^{b,c}$	0.0014	0.0797	0.0102±0.0176
4	$W^{b,c}$	0.0000	0.0065	0.0017±0.0021
5	As^{b}	0.0000	0.0216	0.0060±0.0055
6	$Cd^{b,c}$	0.0000	0.0081	0.0012±0.0018
7	$Pb^{b,c}$	0.0000	0.1327	0.0201±0.0288
8	K^{a}	153.0000	989.0000	540.5000±240.8559
9	Na^{a}	13.3000	146.0000	61.4667±40.3394
10	Ca^{a}	33.1000	377.0000	95.2278±80.9629
11	Mg^{a}	69.1000	162.0000	119.0778±29.2762
12	$Cu^{b,c}$	0.0000	9.6200	4.0175±2.6801
13	$Mn^{b,c}$	1.3600	13.9000	10.2539±3.2554
14	$Fe^{b,c}$	11.7000	59.6000	26.9333±12.8490
15	$Zn^{b,c}$	0.2230	13.9000	4.4796±3.0702

[a] Major elements; [b] Trace-elements; [b,c] Trace-elements/Heavy metals; SD: standard deviation

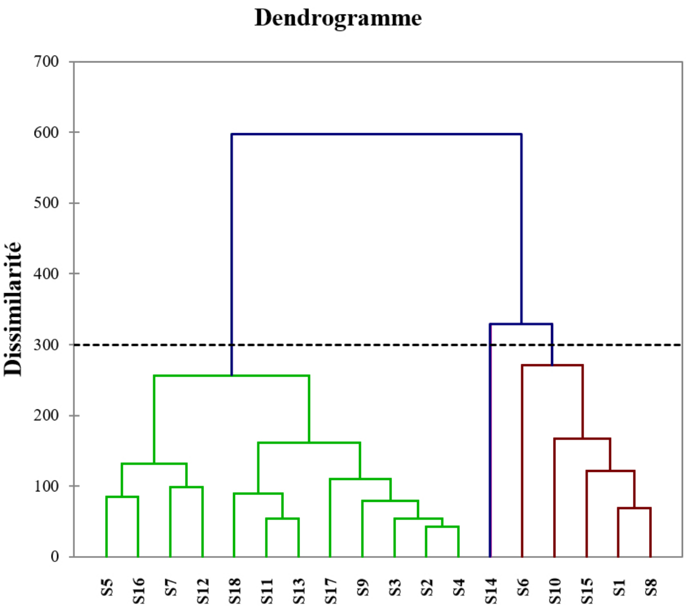

Figure 2. Dendrogram for honey samples S(1 - 18) showing single linkage with Euclidean distances

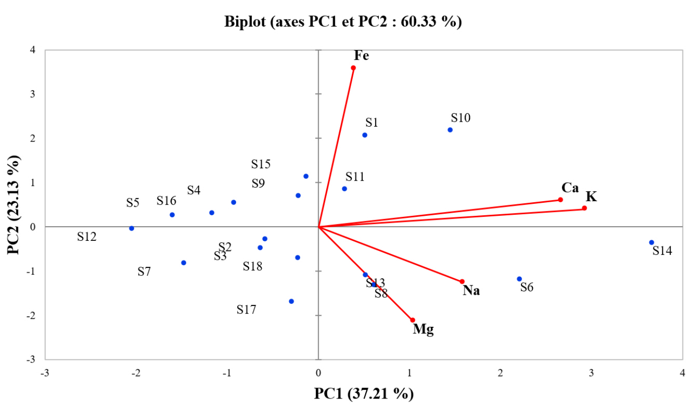

Figure 3. Principal component analysis. Distribution of honey samples S(1 - 18) and dominant elements (K, Ca, Mg, Na and Fe) on scores plot

Discussion

Mineral composition

Our study shown that mineral content in honeys is low and accounts for 0.16% to 0.04% of the total composition, which is in agreement with the composition of nectar honeys (Hernández et al., 2005; Solayman et al., 2016).

The most abundant macro minerals found in these analyzed honey types were K, Ca, Mg and Na, which ranged from 153 to 989 mg/kg, 33.1 - 377 mg/kg, 69.1 - 162 mg/kg and 13.3 - 146 mg/kg, respectively. The results for K and Ca in the analyzed types were coherent with Moroccan, Tunisian, Portuguese, Spanish and Italian honey, in which the total mineral content consisted mainly of potassium ranging from 42.56% to 75.41% (Chakir et al., 2011; Boussaid et al., 2018; Imtara et al., 2018; Silva et al., 2017; Machado De-Melo et al., 2017; Di Rosa et al., 2019). Indeed, potassium was the most important mineral among the fifteen evaluated elements with prevalence in the sample S14 (989 mg/kg), while S12 had the lowest value (153 mg/kg). The calcium was the second most abundant mineral in the studied honeys with the highest concentration in S14 being present at levels of 377 mg/kg and the lowest in S7 (33.1 mg/kg) (Table 3). These results were in agreement with Spanish, Italian, Latvian and Palestinian honeys as described in literature (Fernandez-Torres et al., 2005; Pisani et al., 2008; Vincevica-Gaile et al., 2011; Abdulkhaliq and Swaileh, 2017). Our results for magnesium (69.1 - 162 mg/kg) and sodium (13.3 - 146 mg/kg) were high as compared to the results reported by Moujanni et al. (2017) in Moroccan *Euphorbia resinifera* honeys (Mg = 19.85-45.84 mg/kg; Na = 18.81 - 118.74 mg/kg) and those reported by Lasić et al. (2018) in chestnut honey from Croatia (Mg = 7.326 - 68.320 mg/kg; Na = 18.363 - 48.550 mg/kg). Among the trace elements quantified in the eighteen honey types, Fe content was the highest in *Lavandula vera* D.C. honeys (S1; 59.6 mg/kg) and the lowest in *Silybum marianum* (L.) Gaertn. (S8; 11.7 mg/kg) (Table 3). The concentration of iron was higher compared to other Moroccan honey types (1.46 - 13.95 mg/kg) (Chakir et al., 2011; Moujanni et al., 2017) but similar Fe content was found in United Arab Emirates honey (1.15 - 110.79 mg/kg) (Habib et al., 2014). Besides, these values were lower than those found in honeys from Turkey (< 0.001 - 7.255 mg/kg) (Altun et al., 2017) and Canari Island (0.400 - 5.251 mg/kg) (Hernández et al., 2005), which may be due to the different composition of soils and consequently vegetation diversity (Oroian et al., 2016).

The quantification of the mineral elements such as potassium, calcium, magnesium, sodium and iron in honey is useful to evaluate the nutritional quality of honey samples and to determine a botanical origin differentiation. However, the mineral content is not yet a quality parameter of the European Union Directives (Codex Alimentarius, 2001).

The manganese concentration of the analyzed honey types ranged from 1.36 mg/kg to 13.9 mg/kg with maximum concentration of 13.9 mg/kg in mild white mustard (*Sinapis alba* L.; S6) and minimum in multifloral honey from the Hennaya region (S15; 1.36 mg/kg). As compared with all trace minerals and heavy metals, the mean content of Zn and Cu were 4.4796 ± 3.0702 mg/kg and 4.0175 ± 2.6801 mg/kg, respectively. In addition, the following elements: Cr, Mn, Co, Cu, V, As, Pb and Cd were present in the eighteen analyzed honeys, except for W that is totally absent in almost half of them. These trace elements and heavy metals were detected at level < 1 mg/kg. The resulting data, summarized in Table 3, were consistent with the ranges indicated for honeys from other studies (Di Rosa et al., 2019; Lasić et al., 2018; Moujanni et al., 2017; Oroian et al., 2015; Aghamirlou et al., 2015; Boussaid et al., 2018; Akbari et al., 2012). It was reported by Silva et al. (2009) that the mineral content of honey samples is a potential indicator of its geographical origin. Furthermore, it can be considered as a biomarker of possible pollution by toxic metals (Machado De-Melo et al., 2017; Pohl, 2009; Alves et al., 2013; Devillers et al., 2002). It should be noted that the present study revealed no evidence of contamination with toxic metals of the honey samples collected from the various locations in Tlemcen Province, which could be considered as an indication of honey quality.

Classification of Honey Samples Based on the mineral content

The data mentioned in Table 3 were analyzed using statistical techniques including Principal Components Analysis (PCA) and hierarchical cluster analysis (HCA) to discriminate the honey samples based on their mineral contents.

A matrix (18 × 15) with rows representing the different analyzed honey samples and columns corresponding to the content of the fifteen investigated trace elements and heavy metals was constructed for the PCA (Fig. 3). Since the concentrations of the elements were of different orders of magnitude, the data were first automatically scaled. Fifteen dimensions were needed for the total explanation of the data variability. Table 4 shows that the first four PCs jointly explain 60.33% of the total variance of the data. The first component eigenvalue was 2.417 and accounted for 36.90 % of the total variance, the second component eigenvalue was 1.572 (cumulatively explaining 44.51%), the third component eigenvalue was 1.521 (cumulatively explaining 48.17%) and the fourth component eigenvalue was 1.156.

The HCA resulted in a dendrogram in which three clusters (A - C) were noticed. The first cluster A included S5, S16, S7, S12, S18, S11, S13, S17, S9, S3, S2 and S4; the second one (cluster B) contained S14 and then cluster C was comprised of S6, S10, S15, S1 and S8. The optimal linkage distance level was 300, represented by the dashed line in Fig. 2. It represents a relative measure of similarity between analyzed honeys. Considering the dominant variables (potassium, calcium, magnesium, sodium and iron concentrations), Fig. 3 shows clearly that the honey samples: S1, S6, S8, S10, S15 and S14 (clusters B and C) have higher concentrations of these elements than the other sampling sites.

It can be observed that the cluster A contains orange tree, thyme, rosemary and many multifloral honey types with low mineral contents, which showed similarities to each other. The cluster B contains one multifloral honey type. Furthermore, the cluster C comprises lavender, mild white mustard, milk thistle and carob tree honey types with high levels of minerals, which correlated to each other. Based on PCA and HCA, different honey samples were found to vary not only in their geographic locations, but also in their mono or multifloral characteristics (Table 1). In addition, the mineral element composition contained enough information to develop a classification method of the honey samples studied.

Table 4. Variance estimates (eigenvalues) and cumulative percentage of total variance (%), obtained by PCA considering 18 honey samples and elements characters

Principal components	Honey samples	
	Eigenvalues	Cumulative %
PC1	2.417	36.90
PC2	1.572	44.51
PC3	1.521	48.17
PC4	1.156	60.33

Conclusion

The trace-elements and toxic heavy metals content were found in the eighteen honeys analyzed from the Tlemcen Province of north west region of Algeria; their levels were comparable with other honeys coming from the neighboring countries during the last decade. From the minerals analyzed the most abundant was K, followed by Ca, Mg, Na, Fe, Zn and Cu, while Cr, Mn, W, Co, V, As, Pb and Cd were in low concentrations.

The results obtained in this study showed that the honey samples studied were rich in mineral elements. It was concluded that among all honey samples, multifloral (Sebaa Chioukh, S14) and mild white mustard (Aïn Fezza, S6) honeys contained the highest levels of mineral content. By applying multivariate techniques (PCA and HCA), the study confirmed that dominant minerals (K, Ca, Mg, Na and Fe) possessed a high discriminating power and were a useful tool to characterize different types of honey.

Furthermore, the present study showed that the honey samples investigated were not contaminated by heavy metals, which indicates that apiaries are located far from the various anthropogenic activities that cause environmental pollution. Also, the data suggested that honey samples were of good quality, because they contain higher levels of the essential and trace mineral elements, which are beneficial to human health.

Acknowledgements

Authors are very much thankful to Abou Bekr Belkaïd University and to the Algerian Ministry of Higher Education and Scientific Research for the support under the PRFU project (approval No. B00L01UN130120180009).

Declarations

Author contribution statement

A. H. conceived and designed the experiments; B-R. D., B. S., A. A., K. G. and K. R. performed the experiments; analyzed and interpreted the data; A. H., wrote and edited the article. All authors read and approved the final manuscript.

Competing interest statement

The authors declare no conflict of interest.

References

Abdulkhaliq A., Swaileh K.M. (2017). Physico-chemical properties of multi-floral honey from the West Bank, Palestine. Int J Food Prop 20(2): 447-454. doi: 10.1080/10942912.2016.1166128

Aghamirlou H.M., Khadem M., Rahmani A., Sadeghian M., Mahvi A.H., Akbarzadeh A., Nazmara S. (2015). Heavy metals determination in honey samples using inductively coupled plasma-optical emission spectrometry. J Environ Health Sci Eng 13: 39. doi: 10.1186/s40201-015-0189-8

Akbari B., Gharanfoli F., Khayyat M.H., Khashyarmanesh Z., Rezaee R., Karimi G. (2012). Determination of heavy metals in different honey brands from Iranian markets. Food Addit Contam Part B Surveill 5(2): 105-111. doi: 10.1080/19393210.2012.664173

Ali H., Khan E., Ilahi I. (2019). Environmental chemistry and ecotoxicology of hazardous Heavy metals: environmental persistence, toxicity, and bioaccumulation. J Chem 2019: 1-14. doi: 10.1155/2019/6730305

Altun S.K., Dinç H., Paksoy N., Temamoğullari F.K., Savrunlu M. (2017). Analyses of mineral content and heavy metal of honey samples from south and east region of Turkey by using ICP-MS. Int J Anal Chem. Article ID 6391454, 6 pages. doi: 10.1155/2017/6391454

Alves A., Ramos A., Gonçalves M.M., Bernardo M., Mendes B. (2013). Antioxidant activity, quality parameters and mineral content of Portuguese monofloral honeys. J Food Compost Anal 30(2): 130-138. doi: 10.1016/j.jfca.2013.02.009

Bilandžić N., Đokić M., Sedak M., Varenina I., Kolanović B.S., Končurat A., Rudan N. (2012). Content of five trace elements in different honey types from Koprivnica-Kri Evci County. Slov Vet Res 49: 167-175.

Bogdanov S. (2010). Nutritional and functional properties of honey. Vopr Pitan 79(6): 4-13.

Bogdanov S., Haldimann M., Luginbühl W., Gallmann P. (2007). Minerals in honey: environmental, geographical and botanical aspects. J Apic Res 46(4): 269-275. doi: 10.1080/00218839.2007.11101407

Boussaid A., Chouaibi M., Rezig L., Hellal R., Donsı` F., Ferrari G., Hamdi S. (2018). Physicochemical and bioactive properties of six honey samples from various floral origins from Tunisia. Arab J Chem 11(2): 265-274. doi: 10.1016/j.arabjc.2014.08.011

Bradl H.B. (2005). Heavy Metals in the Environment: Origin, Interaction and Remediation, first ed. Elsevier Academic Press, Amsterdam.

Cantarelli M.A., Pellerano R.G., Marchevsky E.J., Camiña J.M. (2008). Quality of honey from Argentina: Study of chemical composition and trace elements. J Argent Chem Soc 96: 33-41. doi: 10.12691/jfnr-2-9-1

Chakir A., Romane A., Barbagianni N., Bartoli D., Ferrazzi P. (2011). Major and Trace Elements in Different Types of Moroccan Honeys. Aust J Basic Appl Sci 5(4): 223-231.

Codex Alimentarius Commission (2001). Codex Alimentarius Commission Standards. Codex Stan 12-1981, 1-8.

Conti M., Stripeikis J., Campanella L., Cucina D., Tudino M. (2007). Characterization of Italian honeys (Marche Region) on the basis of their mineral content and some typical quality parameters. Chem Cent J 1:14. doi: 10.1186/1752-153X-1-14

de Alda-Garcilope C., Gallego-Picó A., Bravo-Yagüe J.C., Garcinuño-Martínez R.M., Fernández-Hernando P. (2012). Characterization of Spanish honeys with protected designation of origin "Miel de Granada" according to their mineral content. Food Chem 135: 1785-1788. doi: 10.1016/j.foodchem.2012.06.057

Devillers J., Doré J.C., Marenco M., Poirier-Duchêne F., Galand N., Viel C. (2002). Chemometrical analysis of 18 metallic and nonmetallic elements found in honeys sold in France. J Agric Food Chem 50: 5998-6007. doi: 10.1021/jf020497r

Di Rosa A.R., Leone F., Cheli F., Chiofalo V. (2019). Novel approach for the characterisation of Sicilian honeys based on the correlation of physico-chemical parameters and artificial senses. Ital J Anim Sci 18(1): 389-397. doi: 10.1080/1828051X.2018.1530962

dos Santos J.S., dos Santos N.S., dos Santos M.L.P., dos Santos S.N., de Jesus Lacerda J.J. (2008). Honey classification from semi-arid, Atlantic and transitional forest zones in Bahia, Brazil. J Braz Chem Soc 19: 502-508. doi: 10.1590/S0103-50532008000300018

Fernandez-Torres R., Perez-Bernal J.L., Bello-Lopez M.A., Callejon-Mochon M., Jomenez-Sanchez J.C., Guiraum-Perez A. (2005). Mineral content and botanical origin of Spanish honeys. Talanta 65(3): 686-691.

Formicki G., Greń A., Stawarz R., Zyśk B., Gał A. (2013). Metal content in honey, propolis, wax, and bee pollen and implications for metal pollution monitoring. Pol J Environ Stud 22: 99-106.

Fredes C., Montenegro G. (2006). Heavy metals and other trace elements contents in Chilean honey. Cien Inv Agr 33: 50-58.

Golob T., Doberšek U., Kump P., Nečemer M. (2005). Determination of trace and minor elements in Slovenian honey by total reflection X-ray fluorescence spectroscopy. Food Chem 91: 593-600. doi: 10.1016/j.foodchem.2004.04.043

Grembecka M., Szefer P. (2013). Evaluation of honeys and bee products quality based on their mineral composition using multivariate techniques. Environ Monit Assess 185(5): 4033-4047. doi: 10.1007/s10661-012-2847-y

Habib H.M., Al Meqbali F.T., Kamal H., Souka U.D., Ibrahim W.H. (2014). Physicochemical and biochemical properties of honeys from arid regions. Food Chem 153: 35-43. doi: 10.1016/j.foodchem.2013.12.048

Hernández O.M., Fraga J.M.G., Jimenez A.I., Jimenez F., Arias J.J. (2005). Characterization of honey from the Canary Islands: determination of the mineral content by atomic absorption spectrophotometry. Food Chem 93(3): 449-458. doi: 10.1016/j.foodchem.2004.10.036

Imtara H., Elamine Y., Lyoussi B. (2018). Physicochemical characterization and antioxidant activity of Palestinian honey samples. Food Sci Nutr 6(8): 2056-2065. doi: 10.1002/fsn3.754

Jaishankar M., Tseten T., Anbalagan N., Mathew B.B., Beeregowda K.N. (2014). Toxicity, mechanism and health effects of some heavy metals. Interdiscip Toxicol 7: 60-72. doi: 10.2478/intox-2014-0009

Khan F.R., Abadin Z.U.l., Rauf N. (2007). Honey: nutritional and medicinal value. Int J Clin Pract 61(10): 1705-1707. doi: 10.1111/j.1742-1241.2007.01417.x

Khan R.U., Naz S., Abudabos A.M. (2017). Towards a better understanding of the therapeutic applications and corresponding mechanisms of action of honey. Environ Sci Pollut Res 24: 27755-27766. doi: 10.1007/s11356-017-0567-0

Lasić D., Bubalo D., Bošnir J., Šabarić J., Konjačić M., Dražić M., Racz A. (2018). Influence of the botanical and geographical origin on the mineral composition of honey. Agri Conspec Sci 83(4): 335-343.

Machado De-Melo A.A., Almeida-Muradian L.B., Sancho M.T., Pascual-Maté A. (2017). Composition and properties of *Apis mellifera* honey: A review. J Apic Res. doi: 10.1080/00218839.2017.1338444

Mandal M.D., Mandal S. (2011). Honey: its medicinal property and antibacterial activity. Asian Pac J Trop Biomed 1(2): 154-160. doi: 10.1016/S2221-1691(11)60016-6

Moujanni A., Partida L., Essamadi A.K., Hernanz D., Heredia F.J., Terrab A. (2017). Physicochemical characterization of unique unifloral honey: *Euphorbia resinifera*. CyTA - J Food 1-8. doi: 10.1080/19476337.2017.1333529

Moujanni A., Terrab A., Eddoha R., Nasser B., Benbachir M., Tannaoui M., Zouaoui A., Essamadi A.K. (2017). Quantification of heavy metals and pesticides residues in labeled Moroccan *Euphorbia resinifera* honey from Tadla-Azilal. J Mater Environ Sci 8(5): 1826-1836.

Nozal Nalda M.J., Bernal Yagüe J.L., Diego Calva J.C., Martín Gómez M.T. (2005). Classifying honeys from the Soria province of Spain via multivariate analysis. Anal Bioanal Chem 382(2): 311-319. doi: 10.1007/s00216-005-3161-0

Oroian M., Amariei S., Leahu A., Gutt G. (2015). Multi-element composition of honey as a suitable tool for its authenticity analysis. Pol J Food Nutr Sci 65(2): 93-100.

Oroian M., Prisacaru A., Hretcanu E.C., Stroe S.G., Leahu A., Buculei A. (2016). Heavy metals profile in honey as a potential indicator of botanical and geographical origin. Int J Food Prop 19(8): 1825-1836. doi: 10.1080/10942912.2015.1107578

Pisani A., Protano G., Riccobono F. (2008). Minor and trace elements in different honey types produced in Siena County (Italy). Food Chem 107(4): 1553-1560. doi: 10.1016/j.foodchem.2007.09.029

Pohl P. (2009). Determination of metal content in honey by atomic absorption and emission spectrometries. TrAC-Trends Analyt Chem 28(1): 117-128. doi: 10.1016/j.trac.2008.09.015

Ponikvar M., Šnajder J., Sedej B. (2005). Honey as a bioindicator for environmental pollution with SO_2. Apidologie 36: 403-409. doi: 10.1051/apido:2005027

Rashed M.N., Soltan M.E. (2004). Major and trace elements in different types of Egyptian mono-floral and non-floral bee honeys. J Food Compos Anal 17: 725-735. doi: 10.1016/j.jfca.2003.10.004

Rehman K., Fatima F., Waheed I., Akash M.S.H. (2018). Prevalence of exposure of heavy metals and their impact on health consequences. J Cell Biochem 119: 157-184. doi: 10.1002/jcb.26234

Ru Q-M., Feng Q., He J-Z. (2013). Risk assessment of heavy metals in honey consumed in Zhejiang province, southeastern China. Food Chem Toxicol 53: 256-262. doi: 10.1016/j.fct.2012.12.015

Silva L.R., Sousa A., Taveira M. (2017). Characterization of Portuguese honey from Castelo Branco region according to their pollen spectrum, physicochemical characteristics and mineral contents. J Food Sci Technol 54(8): 2551-2561. doi: 10.1007/s13197-017-2700-y

Silva L.R., Videira R., Monteiro A.P., Valentão P., Andrade P.B. (2009). Honey from Luso region (Portugal): Physicochemical characteristics and mineral contents. Microchem J 93: 73-77.

Solayman M., Islam M.A., Paul S., Ali Y., Khalil M.I, Alam N., Gan S.H. (2016). Physicochemical properties, minerals, trace elements, and heavy metals in honey of different origins: A Comprehensive Review. Compr Rev Food Sci Food Saf 15(1): 219-233. doi: 10.1111/1541-4337.12182

Tchounwou P.B., Yedjou C.G., Patlolla A.K., Sutton D.J. (2012). Heavy metal toxicity and the environment, in: Luch A. (Ed.), Molecular, Clinical and Environmental Toxicology. Springer Basel, Basel, pp. 133-164. doi: 10.1007/978-3-7643-8340-4_6

Vincevica-Gaile Z., Klavins M., Rudovica V., Viksna A. (2011). Geographical dissemination of trace and major elements in honey. WIT Trans Ecol Envir 167: 211-220.

aCS85_39