



Levels of trace elements and rare earth elements in honey from Jordan

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Abstract

Honey is a common sweetener in the Jordanian diet with an annual consumption of approximately one thousand tons, two-thirds of which are imported. It is believed that the elemental profile of honey is an indicator of safety and botanical and geographic origin. In the literature, there are a lack of studies concerning the levels of major and trace elements in honey in Jordan. A total of 46 elements, including 15 rare earth elements (REEs), were analyzed by inductively coupled plasma-mass spectrometry (ICP-MS) in 18 monofloral and multifloral imported honey samples and 12 multifloral local samples. Regarding monofloral samples, Black Forest samples had the highest total metal content, while acacia samples had the lowest total metal content. Local multifloral honey had the largest Sr and total REE levels, while it had the lowest Mn levels. Very low levels of toxic elements were found in all samples, indicating the safety of honey in Jordan for human consumption. The results of this study showed that a large number of samples (> 100) and the application of advanced statistical models are required to discriminate between multifloral imported and local honey.

Keywords Honey · ICP-MS · Safety assessment · Trace elements · Rare earth elements

Introduction

Honey is a valuable natural product that is marketed with a wide range of nutritional and medicinal properties. Honey is produced by bees (*Apis mellifera*) from the nectar of flowers or the secretions of plants other than flowers (Alvarez-Suarez et al. 2010).

Honey can be classified based on its origin and the way it has been harvested and processed. Following honey origin, it is mainly categorized into monofloral and multifloral. Monofloral honey arises predominantly from a single botanical origin and is named after the plant, such as citrus, manuka,

and acacia honey. Multifloral honey has several botanical sources where none is predominant (Anklam 1998).

The main composition of honey is carbohydrates or sugars, which represent 95% of honey dry weight. It also contains bioactive compounds, such as organic acids, proteins, amino acids, minerals, polyphenols, vitamins, and aroma compounds (Ball 2007).

Honey in Jordan is either locally produced or imported from neighboring countries (Saudi Arabia and Egypt) or from European countries (Germany, Spain, and Turkey). In 2019, Jordan produced approximately 320 tons of honey and imported approximately 600 tons (Jordan's 2019). Certified local honey is usually expensive and preferable to major consumers.

Jordan is a small country with a total area of approximately 90,000 km². Its topography is highly variable, from more than 400 m below sea level in the Dead Sea to 1,854 m above sea level at the southernmost boundary. Jordan is divided into four biogeographic zones: Mediterranean, Irano-Turania, Saharo-Arabian, and Sudania (subtropical penetration) (Al-Eisawi 1985). This variation in the physical environment is highly rich in biological diversity, especially plant biodiversity (Al-Eisawi 1982, 1996, 1998). Consequently, richness in biodiversity in general is one of the important characteristics of

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Jordan, especially plant biodiversity. Jordan's plant diversity is remarkably high considering its size and aridity. The total number of recorded vascular plant species in Jordan is now over 2,622 species belonging to 113 families and approximately 810 genera (Al-Eisawi 1982). Flowering plants are the dominant group of species, which makes Jordan an excellent place for beekeeping activities. Generally, most Jordanian beekeepers migrate their colonies to the Jordan valley between October and April to avoid cold effects during the winter season and to benefit from early flowering of cultivated and wild plants in the Jordan valley, harvesting their multifloral honey by the end of April. Afterwards, the beekeepers transport their hives to the uplands for further honey collection from wild flowering plants growing there (Zaitoun et al. 2000).

Honey contains a variety of macro- and microelements in the range of 0.02–1.03%. Minerals and trace elements in honey seem to be good indicators of environmental pollution or the botanical and geographic origin of the honey (Solayman et al. 2016). Potassium (K) is the most abundant major metal in honey. Other major metals include sodium (Na), calcium (Ca), magnesium (Mg), and phosphorous (P) (Hernández et al. 2005). The rest of the elements are considered trace elements. Some of these elements are essential due to their involvement in metabolic processes and have several physiological and biochemical functions for correct cellular metabolism. These elements include cobalt (Co), copper (Cu), chromium (Cr), iron (Fe), manganese (Mn), molybdenum (Mo), nickel (Ni), selenium (Se), and zinc (Zn) (Squadrone et al. 2020a). The rest of the trace elements are considered nonessential. However, some trace elements exert toxicity even at low concentrations and are considered potentially toxic, such as mercury (Hg), cadmium (Cd), arsenic (As), and lead (Pb). Rare earth elements (REEs), an emerging group of nonessential elements, were detected in honey and have been used as geological tracers (Squadrone et al. 2020a).

Multielemental analysis has been recommended as a suitable tool for evaluating the mineral composition of honey. Inductively coupled plasma-atomic emission spectroscopy (ICP-AES) and inductively coupled plasma-mass spectrometry (ICP-MS) are the most popular techniques due to their compatibility with most elements in the periodic table, large sensitivity, and large linear dynamic range (Devillers et al. 2002; Kiliç Altun et al. 2017; Squadrone et al. 2020a; Wetwitayaklung et al. 2018).

Investigations regarding the occurrence of metals in honey for monitoring toxic metals and assessing the botanical and geographic origin of honey have been performed in several popular honey-producing countries. European countries include Italy (Conti et al. 2018; Squadrone et al. 2020b), Spain (De Alda-Garcilope et al. 2012), Croatia (Bilandžić et al. 2017), Hungary

(Cziza et al. 2015), France (Devillers et al. 2002), Poland (Grembecka and Szefer 2013), Greece (Drivelos et al. 2021), and Serbia (Spiric et al. 2019). Non-European countries include Argentina (Cantarelli et al. 2008), Brazil (de Oliveira et al. 2017), China (Ru et al. 2013), Ethiopia (Adugna et al. 2020), Canary Islands (Hernández et al. 2005), Egypt (Rashed and Soltan 2004), Turkey (Kiliç Altun et al. 2017), Iran (Aghamirlou et al. 2015), Thailand (Wetwitayaklung et al. 2018), and Uruguay (Berriel et al. 2019).

Multivariate data analytical methods usually accompany the elemental profile for predicting mainly the botanical origin and, to a lesser extent, the geographic origin of honey. Principal component analysis (PCA), discriminant analysis (DA), and cluster analysis are the preferred methods for performing predictive analyses. PCA and DA are the most popular due to their ease of application and interpretation (Lazarević et al. 2012; Conti et al. 2018; Berriel et al. 2019). However, a successful prediction necessitates that honey samples be obtained from producers or purchased from reliable stores. Additionally, the origin of samples must be ascertained prior to the application of data analytical methods. This process is accomplished through pollen analysis, which is known as a slow process. Also, a compromise between the number of samples and number of variables must be made (Maione et al. 2019).

The objective of this study was to determine the levels of 46 elements in imported honey (monofloral and multifloral) and local honey (multifloral) from Jordan. The elements were categorized into major elements (Na, Mg, P, K, and Ca), essential trace elements (Fe, Zn, Mn, Cu, Ni, Cr, Se, Mo, and Co), nonessential trace elements (Li, Be, Al, V, Ga, Sr, Ag, Sn, Cs, Ba, Bi, and U), potentially toxic trace elements (As, Cd, In, Tl, and Pb), and rare earth elements (Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu). Levels of elements were evaluated for nutritional and safety assessment and the possibility of characterizing the botanical and geographic origin of the honey.

Materials and methods

Reagents

Chemicals and standards used were analytical grade. HNO₃ (69% w/w, extra pure) and H₂O₂ (35% w/w, extra pure) were purchased from Sigma (St. Louis, MO, USA). Ultrapure deionized water (Milli-Q water) was employed to prepare standard solutions and sample solutions. A stock solution of combined elements (10.0 mg/L each) was purchased from Merck (Darmstadt, Germany). All plastic containers, pipette tips, polypropylene flasks, Pyrex glass digestion tubes, and

reagents that came into contact with samples or standards were checked for contamination.

Sampling and sample preparation

Local honey samples ($n=12$) were obtained directly from commercial honey producers, and imported honey samples ($n=18$) were purchased from local markets. The thirty sample brands are available to Jordanian customers from either producers or public stores. However, other local honey brands are produced but not available to public consumers. These brands were not included in this study. All samples were labeled with their floral type. Samples were stored in brown bottles at room temperature. All 12 local samples were multifloral from harvest year 2019. Details of imported samples, including imported country and floral type, are presented in Table 1S.

A 1.00 g honey sample in triplicate was mixed with 6.0 mL HNO_3 and 2.0 mL H_2O_2 as a catalyst and digested using ETHOS 1-advanced Microwave Digestion Lab Station (Milestone S.r.l., Italy). The thermal program included a gradual rise in temperature for 12 min to reach 200 °C, a constant temperature at 200 °C for 20 min, and finally a gradual decline for 12 min to reach room temperature. After digestion was completed, as indicated by the appearance of a colorless, completely clear, and homogeneous solution, the clear mixture was left to cool down, and the contents of the tubes were transferred to digestion tubes, evaporated to near dryness at 110 °C, and then diluted to 20 mL with 1% HNO_3 solution.

The obtained solutions were measured for the targeted elements using ICP-MS (Agilent 7500a Series ICP-MS). The ICP-MS configuration and operating parameters, including selected isotopes, are presented in Table 2S.

Calibration and quality control

External calibration was employed by sequential dilution of the multielemental calibration standard to prepare six calibration standards ranging from 0.10 to 20 $\mu\text{g/L}$ for REEs, 0.10 to 100 $\mu\text{g/L}$ for trace and potentially toxic elements, and from 10 to 1000 $\mu\text{g/L}$ for major elements. Absolute intensities, counts per second (Cps), versus concentration were employed in the statistical analysis. The linear least squares method was applied for the calculation of statistical parameters for calibration curves. Elemental concentrations were calculated from the respective calibration curves.

Calibration curves were linear over the range of the three calibration ranges with correlation coefficients better than 0.999.

The limit of detection (LOD), defined as $3S_b/a$, and the limit of quantitation (LOQ), defined as $10S_b/a$, where S_b is the standard deviation of the intercept and “a” is the slope of the calibration curve, were calculated for each element. The method limit of detection (MLOD) and method limit of

quantitation (MLOQ) were calculated by multiplying the LOD and LOQ by the dilution factor. The precision (% RSD), defined as $(S_a/a) \times 100$, where S_a is the standard deviation of the slope of the calibration curve, was also calculated.

For major elements, the method limits of detection (MLODs) varied 1.46 $\mu\text{g/L}$ and 6.52 $\mu\text{g/L}$ for Mg and P, respectively. For trace elements, MLODs varied between 0.008 and 4.21 $\mu\text{g/L}$ for Ga and Mn, respectively. For REEs, MLODs varied between 0.07 and 0.54 $\mu\text{g/L}$ for Sm and Eu, respectively. The precision (% RSD) was less than 5% for the majority of targeted elements except for K (8.50%), Li (7.36%), P (7.01%), Mg (5.71%), V (5.38%), and As (5%). Detailed calibration results are presented in Table 3S.

Quality control studies were performed on a composite sample prepared from the digest of the 30 honey samples. The composite sample was certified for targeted elements by the standard addition method (Tahboub et al. 2021) and was employed for further quality control measurements.

Accuracy and recovery results were above 94%, and precision results (% RSD) were less than 6% for all determined elements. The accuracy, precision, and recovery results on certified samples and postspiked certified samples are presented in Table 4S.

Statistical analysis

JMP PRO 14 software (*jmp*-statistical discovery from SAS, NC, USA) was employed for the calculation and evaluation of statistical parameters. Q-Q plots were employed to assess the normality of the data; if the variable could be assumed to be normally distributed, a *t*-test for unequal variances, which was insensitive to heterogeneous variables, was employed to discriminate between local and imported honey samples. Then, the nonparametric Wilcoxon test was used. Q-Q plots indicated that the individual levels of the elements Mn, Ni, Ba, La, Ce, Pr, Nd, Sm, and Gd, and ΣREEs were not normally distributed. Detailed Q-Q plots are presented in Table 5S.

Results and discussion

The analytical results for the basic 31 elements and 15 rare earth elements (REEs) in targeted honey samples ($n=30$) are displayed in Tables 1 and 2.

Levels for each targeted element (mg/kg or $\mu\text{g/kg}$) include the mean \pm SD and range (minimum-maximum). Additionally, the elements were categorized as local or imported, and the level of each element in its category is reported in Tables 1 and 2 as the mean \pm SD. The basic elements in Table 1 were classified into major elements ($n=5$) (Na, Mg, P, K, and Ca), essential trace elements ($n=9$) (Cr, Mn, Fe, Co, Ni, Cu, Zn, Se, and Mo), nonessential elements ($n=12$) (Li, Be, Al, V, Ga,

Table 1 Statistical results for major and trace elements in honey samples.

Element	Total ^{a, b} (mean ± SD)	Range ^{a, b} (min-max)	Local ^{a, c} (mean ± SD)	Imported ^{a, d} (mean ± SD)	<i>p</i> -value ^e
Major elements (mg/kg)					
Na	66.9 ± 49.3	19.2–258	88.6 ± 65.7	54.8 ± 35.6	NS
Mg	43.4 ± 27.4	9.1–107.8	37.6 ± 23.5	46.3 ± 29.2	NS
P	74.3 ± 48.4	12.3–187.3	59.5 ± 21.0	81.7 ± 56.5	NS
K	718 ± 746	27.7–2404	624 ± 710	840 ± 771	NS
Ca	100.0 ± 44.9	26.9–205	88.4 ± 50.3	106 ± 41.9	NS
∑Total	1052 ± 844	132–2872	898 ± 673	1129 ± 738	NS
Essential trace elements (mg/kg)					
Cr	0.180 ± 0.053	0.06–0.306	0.175 ± 0.073	0.182 ± 0.043	NS
Mn	1.96 ± 3.48	0.13–13.40	0.435 ± 0.160	2.77 ± 4.11	NS
Fe	19.4 ± 11.0	7.03–49.3	21.3 ± 13.8	18.5 ± 9.66	NS
Co	0.011 ± 0.010	0.003–0.049	0.008 ± 0.005	0.012 ± 0.012	---
Ni	0.123 ± 0.181	0.030–0.688	0.050 ± 0.015	0.160 ± 0.214	NS
Cu	0.606 ± 0.478	0.145–2.34	0.615 ± 0.650	0.601 ± 0.382	NS
Zn	1.67 ± 0.785	0.48–3.72	1.92 ± 0.681	1.51 ± 0.821	NS
Se	0.041 ± 0.019	0.005–0.105	0.041 ± 0.025	0.041 ± 0.016	---
Mo	0.030 ± 0.013	0.020–0.096	0.029 ± 0.003	0.031 ± 0.016	---
Nonessential trace elements (µg/kg)					
Li	18.3 ± 18.0	0.29–72.9	15.6 ± 9.30	19.6 ± 21.2	---
Be	1.30 ± 0.43	0.42–2.31	1.30 ± 0.43	1.30 ± 0.45	---
Al	4670 ± 3510	2660–21900	4442 ± 1529	4985 ± 1950	0.008
V	11.6 ± 6.80	5.14–37.7	15.7 ± 9.4	9.5 ± 3.9	---
Ga	13.6 ± 18.8	ND–78.6	11.0 ± 7.3	14.9 ± 22.6	---
Sr	580 ± 440	75.0–1620	781 ± 380	480 ± 437	0.007
Ag	35.2 ± 69.9	5.70–386	27.7 ± 23.2	39.1 ± 84.7	---
Sn	56.9 ± 29.9	33.6–186	70.7 ± 46.7	50.1 ± 13.3	---
Cs	4.80 ± 8.60	0.17–37.9	2.00 ± 3.61	6.2 ± 10.1	---
Ba	251 ± 233	59–1060	204 ± 87	278 ± 278	NS
Bi	0.77 ± 0.48	0.28–2.56	0.78 ± 0.39	0.77 ± 0.53	---
U	1.23 ± 0.65	0.49–3.23	1.31 ± 0.51	1.23 ± 0.65	---
Potentially toxic elements (µg/kg)					
As	3.10 ± 1.50	ND–6.90	3.10 ± 1.60	3.10 ± 1.51	NS
Cd	3.02 ± 1.18	0.38–6.47	2.90 ± 0.60	3.10 ± 1.41	NS
In	0.68 ± 0.21	0.40–1.52	0.72 ± 0.20	0.68 ± 0.23	NS
Tl	1.33 ± 2.56	0.16–13.4	10.7 ± 2.82	1.73 ± 3.07	NS
Pb	13.6 ± 6.60	3.61–31.5	11.1 ± 2.5	15.0 ± 7.52	0.02

^a Four replicates^b *n*=30^c *n*=12^d *n*=18^e Between local and imported samples*p* > 0.05 (NS no significant difference)

Sr, Ag, Sn, Cs, Ba, Bi, and U), and potentially toxic elements (*n*=5) (As, Cd, In, Tl, and Pb). The rare earth elements (REEs), *n*=15 (Y, La, Ce, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, and Lu), are presented in

Table 2. The classifications of selected basic elements and selected REEs based on floral type are presented in Table 3. Selected elements were those having significant concentrations.

Table 2 Statistical results for rare earth elements ($\mu\text{g}/\text{kg}$) in honey samples.

Element	Total ^{a, b} (mean \pm SD)	Range ^{a, b} (min-max)	Local ^{a, c} (mean \pm SD)	Imported ^{a, d} (mean \pm SD)	<i>p</i> -value ^e
Y	2.42 \pm 0.92	1.32–4.43	3.20 \pm 0.98	2.15 \pm 0.71	----
La	19.4 \pm 21.9	3.9–78	35.0 \pm 27.0	12.5 \pm 15.5	0.006
Ce	66 \pm 82	15.1–314	111 \pm 100	48 \pm 69	0.011
Pr	5.5 \pm 7.7	1.10–28.3	9.4 \pm 9.1	3.9 \pm 6.7	0.025
Nd	20.8 \pm 31.4	3.40–116	36.0 \pm 37.0	14.3 \pm 27	0.01
Sm	3.60 \pm 4.20	0.90–15.9	5.8 \pm 4.8	2.70 \pm 3.60	0.02
Eu	0.47 \pm 0.38	0.22–1.28	0.48 \pm 0.33	0.47 \pm 0.33	----
Gd	1.40 \pm 1.80	ND–6.3	2.33 \pm 2.02	1.29 \pm 0.30	NS
Tb	0.23 \pm 0.12	0.10–0.58	0.30 \pm 0.14	0.28 \pm 0.26	----
Dy	1.01 \pm 0.38	0.41–1.70	1.44 \pm 0.52	1.34 \pm 1.48	----
Ho	0.37 \pm 0.21	0.11–1.07	0.37 \pm 0.15	0.39 \pm 0.22	----
Er	0.41 \pm 0.24	ND–0.93	0.54 \pm 0.18	0.40 \pm 0.28	----
Tm	0.18 \pm 0.18	ND–0.80	0.17 \pm 0.14	0.16 \pm 0.20	----
Yb	0.55 \pm 0.28	0.16–1.36	0.58 \pm 0.26	0.54 \pm 0.29	----
Lu	0.28 \pm 0.18	0.02–0.86	0.23 \pm 0.16	0.29 \pm 0.18	----
ΣREE	145 \pm 157	41–489	203 \pm 165	98 \pm 113	0.02

^a Four replicates

^b *n*=30

^c *n*=12

^d *n*=18

^e Between local and imported samples

p > 0.05 (NS no significant difference). ND not detected

Major elements

The major elements Na, Mg, P, K, and Ca represented more than 90% of the total elemental content in most foods, including honey, and their levels are related to their presence in soil, fertilizer, and irrigation water. A wide range, max/min > 10, was observed for all major elements. Their decreasing order in honey was $\text{K} \gg \text{Ca} > \text{P} > \text{Na} > \text{Mg}$ (Table 1). The same order held for imported honey samples; however, in local samples, Na was more abundant than P. Additionally, total major elements (Σ_{major}) were significantly more abundant in imported honey samples (Table 1). The dependence of major elemental content on botanical origin was demonstrated between the monofloral Black Forest ($\Sigma_{\text{major}} = 2131 \text{ mg}/\text{kg}$) and monofloral acacia ($\Sigma_{\text{major}} = 420 \text{ mg}/\text{kg}$) (Table 3). The levels of major elements in local and imported multifloral honey samples were similar (Table 3).

Essential trace elements

Levels of essential trace elements in honey samples varied between 19.4 mg/kg for Fe and 0.011 mg/kg for Co. A wide range, max/min > 10, was observed for all essential trace elements. Their decreasing order in honey was $\text{Fe} \gg \text{Mn} > \text{Zn} > \text{Cu} > \text{Cr} > \text{Ni} > \text{Se} > \text{Mo} > \text{Co}$ (Table 1).

Iron, Fe, is an element required for essential functions in cells related to oxygen transport, oxidase activities, and energy metabolism (Squadrone et al. 2020c). Levels of Fe varied between 7.03 mg/kg in an imported multifloral sample and 49.3 mg/kg in a local multifloral sample.

Manganese, Mn, is a metalloenzyme involved in various metabolic processes (Squadrone et al. 2020c). Mn levels in honey varied between 0.13 mg/kg in an imported monofloral sample (acacia) and 13.3 mg/kg in an imported Black Forest monofloral sample.

Zinc, Zn, is utilized in various physiological functions and is ubiquitous in the body; three general functional classes (catalytic, structural, and regulatory) define the role of Zn in biological processes. Zn is important for regulation of gene expression, stabilizing DNA, and has several specific functions in zinc enzymes (Jurowski et al. 2014). Zn levels varied between 0.48 mg/kg in an imported acacia monofloral sample and 3.72 mg/kg in an imported multifloral sample.

Copper, Cu, is classified as an essential element due to the wide range of enzymes that use this element as a cofactor. However, the symptoms of Cu deficiency are diverse. On the other hand, an excess of Cu may cause genetic disorders such as Wilson disease (Jurowski et al. 2019a). Cu levels varied between 0.145 mg/kg in a local multifloral sample and 2.34 mg/kg in another local multifloral sample.

Table 3 Statistical results for elements in honey samples based on flora type: Black Forest, acacia, multiflora (imported), and multiflora (local).

Element	Black Forest (mean ± SD)	Acacia (mean ± SD)	Multiflora (imported) (mean ± SD)	Multiflora (local) (mean ± SD)
Major elements (mg/kg)				
Na	50 ± 23	20 ± 1.5	66 ± 39	89 ± 70
Mg	67 ± 28	14.5 ± 1.6	45 ± 26	39 ± 24
P	136 ± 54	42 ± 3.5	68 ± 49	60 ± 22
K	1750 ± 677	222 ± 38	615 ± 569	668 ± 738
Ca	128 ± 27	54 ± 16	110 ± 41	91 ± 52
∑Major	2131 ± 825	420 ± 72	904 ± 574	947 ± 743
Essential elements (mg/kg)				
Cr	0.18 ± 0.064	0.16 ± 0.047	0.19 ± 0.033	0.18 ± 0.076
Mn	7.64 ± 4.52	0.201 ± 0.072	1.25 ± 2.42	0.432 ± 0.169
Fe	16.1 ± 9.80	12.6 ± 6.04	20.9 ± 10.0	22.8 ± 13.6
Co	29 ± 15	4.0 ± 1.70	7.5 ± 3.1	8.7 ± 4.7
Ni	0.48 ± 0.21	0.053 ± 0.013	0.052 ± 0.020	0.048 ± 0.014
Cu	1.07 ± 0.33	0.196 ± 0.034	0.51 ± 0.25	0.66 ± 0.67
Zn	2.04 ± 0.65	0.85 ± 0.414	1.51 ± 0.85	1.93 ± 0.73
Se	0.035 ± 0.012	0.031 ± 0.012	0.046 ± 0.018	0.040 ± 0.027
Mo	0.045 ± 0.028	0.023 ± 0.003	0.026 ± 0.003	0.029 ± 0.003
Selected nonessential elements (mg/kg)				
Al	5.11 ± 1.13	2.71 ± 0.19	5.62 ± 5.84	4.54 ± 1.59
Sr	0.53 ± 0.43	0.080 ± 0.004	0.560 ± 0.450	0.840 ± 0.350
Ba	0.58 ± 0.42	0.080 ± 0.004	0.191 ± 0.083	0.20 ± 0.092
Sn	0.082 ± 0.015	0.082 ± 0.001	0.10 ± 0.02	0.12 ± 0.078
Ag	0.009 ± 0.002	0.009 ± 0.002	0.059 ± 0.11	0.028 ± 0.024
Potentially toxic elements (µg/kg)				
As	3.5 ± 1.5	1.5 ± 1.6	3.3 ± 1.37	3.1 ± 1.7
Cd	4.5 ± 1.5	2.3 ± 1.7	2.7 ± 0.90	2.9 ± 0.60
Pb	16 ± 11	20 ± 5.8	13 ± 6.0	11 ± 2.5
Selected rare earth elements (µg/kg)				
Y	2.40 ± 0.73	1.8 ± 0.33	2.11 ± 0.79	3.2 ± 1.0
Nb	15.6 ± 28	3.5 ± 3.5	2.7 ± 4.3	2.62 ± 3.85
La	8.2 ± 3.5	10.0 ± 6.1	16 ± 21	35.1 ± 27.4
Ce	29.6 ± 18.5	31.2 ± 6.9	63 ± 92	111 ± 100
Pr	2.01 ± 0.84	2.20 ± 1.37	5.5 ± 9.1	9.4 ± 9.2
Nd	6.7 ± 3.6	8.1 ± 5.3	20 ± 37	36 ± 37
Sm	1.6 ± 0.5	1.70 ± 0.50	3.6 ± 4.9	5.8 ± 4.8
Th	13.2 ± 10.9	8.0 ± 5.9	13 ± 14	23 ± 17
∑REE	51.2 ± 35.7	55.5 ± 12.7	117 ± 121	203 ± 135

Chromium, Cr, is a very problematic element due to the number of its oxidation states, especially Cr(III) and Cr(VI). Cr(III) is the most abundant form in the environment and plays a role in glucose metabolism. Cr(III) deficiency may be associated with cardiovascular diseases and numerous system disorders (Jurowski et al. 2019b). Levels of Cr varied between 0.06 mg/kg in a local multiflora sample and 0.31 mg/kg in another local multiflora sample.

Other essential elements, Ni, Se, Mo, and Co, were detected at lower levels and had less impact on nutrition or safety assessments. No significant differences were observed between local and imported samples for all targeted essential trace elements except for Mn, where its mean in imported samples was 5 times larger than local samples.

Nonessential trace elements

Among the 12 targeted nonessential elements, Al, Sr, Ba, and Sn had significant levels and were found in decreasing order: Al >> Sr > Ba > Sn (Table 1).

Aluminum, Al, a common metal in the earth's crust, was detectable in all samples, ranging from 2.67 mg/kg in an imported acacia monoflora sample to 21.9 mg/kg in an imported multiflora sample. Mean levels of Al in local and imported samples were similar.

Strontium, Sr, was measured at significant levels in all samples, ranging from 0.075 mg/kg in an imported acacia monoflora sample to 1.62 mg/kg in an imported multiflora sample. Mean Sr levels between local and imported samples were significantly different. Sr could be employed as an elemental marker to distinguish between local and imported honey.

For barium, Ba, levels varied between 0.059 mg/kg in an imported multiflora sample and 1.06 mg/kg in a Black Forest monoflora sample. There was no significant difference in mean Ba levels between local and imported samples.

Tin and Sn were found at low levels in honey, with the lowest concentration of 0.033 mg/kg in an imported Black Forest monoflora honey sample and the highest concentration of 0.186 mg/kg in a local multiflora sample. Sn levels in local samples were slightly higher than those in imported samples.

Potentially toxic trace elements

Thallium, Tl, and indium, In, had lower levels and were not considered for toxicity and assessment. Arsenic, As, is a highly toxic element. Its levels were not detected (ND) in 50% of the samples and were up to 6.9 µg/kg in an imported multifloral sample. Cadmium, Cd, and lead, Pb, are common pollutants that can enter the food chain after contamination of flowering plants. Cd levels were low in most samples and reached 6.7 µg/kg in an imported Black Forest monofloral sample. Pb levels were low in most samples and reached 31.5 µg/kg in the same imported Black Forest monofloral sample. Levels of Cd and Pb as well as As were much lower than regulated levels in food (Paz 2017).

Rare earth elements

REEs are concentrated in different geological environments, and their presence in honey is related to geographic origin and geochemical soil composition. Significant levels were detected for La, Ce, Pr, Nd, and Sm in decreasing order: Ce > Nd > La > Pr > Sm (Table 2). Total REE levels varied between 41 µg/kg in an imported acacia monofloral sample and 583 µg/kg in a local multifloral sample with a mean of 145 µg/kg and standard deviation (SD) of 157 µg/kg. Local samples had significantly larger means than imported samples. Significantly larger deviations, mainly in imported samples, were due to the difference in geographic origins between imported countries.

Safety assessment

Honey is a popular sweetener in Jordanian diets. An adult usually consumes 10–50 g daily. Thus, honey may contribute significantly to the total daily intake of elements. Table 4 presents the levels of estimated daily intake (EDI), recommended daily intake (RDI) (Paz 2017), and percentage contribution of each metal. The percentage contributions of Na, Mg, P, K, Ca, Mn, and Zn were negligible relative to their respective EDI values (<1.0%). Little percentage contributions were observed for the rest of the essential elements. Levels of potentially toxic elements (As, Cd, and Pb) were negligible to their RDI values, indicating the safety of honey in Jordanian markets, either local or imported, for human consumption.

Assessment of botanical and geographic origins

Botanical and geographic origins are usually considered in the assessment and authentication of honey (Gallmann 2007; Salvador et al. 2019; Squadrone et al. 2020b; Squadrone et al. 2020b; Zhou et al. 2018). A single factor ANOVA test is usually employed for discrimination between means of variables (elements) in two populations (local and imported). A *p* > 0.05 indicates no significant difference between variables. However, ANOVA is sensitive to the normality and heterogeneity of the data. In our study, Q-Q plots were employed to assess the normality of the levels of each element in the thirty samples. Q-Q plots revealed that the levels of essential trace elements Ni and Mn; nonessential elements Ba; REEs La, Ce, Pr, Nd, Sm, and Gd; and ΣREEs were not normally distributed (Table 5S). Additionally, the variances of elements in local

Table 4 Estimated daily intake (EDI), recommended daily intake (RDI), and percentage contribution for selected elements.

Element	Mean (mg/kg)	EDI ^a (mg/day)	RDI (mg/day)	% Contribution
Na	66.9	0.67–3.35	2300	0.03–0.15
Mg	43.4	0.434–2.17	420	0.10–0.15
P	74.3	0.743–10.9	700	0.11–0.55
Al	4.67	0.047–0.235	20	0.24–1.18
K	718	7.18–35.9	4700	0.15–0.76
Ca	100	1.0–5.0	1000	0.10–0.50
Cr	0.18	0.0018–0.009	0.120	1.5–7.5
Fe	19.4	0.19–0.95	18	1.05–5.30
Mn	1.96	0.02–0.10	3.0	0.67–3.0
Zn	1.7	0.017–0.085	15	0.11–0.55
Co	11 µg/kg	0.11–0.55	6.0 µg	1.83–9.16
Ni	0.13	0.0013–0.0065	0.15	0.87–4.33
Cu	0.61	0.0061–0.035	2.0	0.30–1.50
As	3.1 µg/kg	0.031–0.155	150 µg	0.02–0.10
Cd	3.02 µg/kg	0.032–0.16	75 µg	0.042–0.213
Pb	13.6 µg/kg	0.136–0.68	250 µg	0.054–0.272

^aBased on an adult consuming 10–50 g honey

and imported samples were different (Tables 1 and 2). Thus, ANOVA could not be applied and was replaced by alternative *t*-tests (unequal variances) for normally distributed elements and the nonparametric Wilcoxon test for nonnormally distributed elements. Both tests are insensitive to heterogeneous variables. As in ANOVA, $p > 0.05$ indicates no significant difference (NS). The *p*-values in Table 1 indicate no significant differences between local and imported honey samples for all basic elements except the nonessential elements Al and Sr and the potentially toxic element Pb. Environmental factors may have contributed to the differences in the levels of Pb. The *p*-values in Table 2 indicate significant differences between local and imported honey samples for all targeted REEs except Nd.

A closer look at imported samples consisted of Black Forest monofloral samples ($n=4$), Acacia monofloral samples ($n=4$), and multifloral samples ($n=10$). All local samples were multifloral ($n=12$). Thus, botanical origin was considered in the assessment and authentication. The levels of selected elements (mean \pm SD) are presented in Table 3.

The precision of results of monofloral samples (Table 3) was much smaller than those in Tables 1 and 2. Σ major elements in Black Forest samples were much larger than those in acacia samples. While it was easier to discriminate between monofloral and multifloral honey, more factors are needed to discriminate between multifloral local and multifloral imported samples. Aluminum, Al, strontium, Sr, and Σ REEs were significantly different between multifloral local and imported honey samples. These elements may be considered chemical markers for tentative discrimination between local and imported multifloral samples.

Pearson's correlation coefficients between targeted elements in honey samples were computed and are presented in Table 5.

In this study, we combined monofloral Black Forest and acacia samples in one category under the title "monofloral." The other two categories were multifloral imported and multifloral local. Rare earth elements in the three categories were positively correlated with each other ($1.00 > r > 0.80$) and negatively correlated with other elements; thus, they were not considered in this study.

Selected correlations were based on categories with positive correlation coefficients ($r > 0.50$). Among the 60 selected correlations, the largest positive correlation coefficients were distributed as 22 for multifloral local samples, 11 for multifloral imported samples, and 27 for monofloral samples. The correlations Na/Mg, Na/Se, Mg/Se, Cr/Fe, Cr/Co, Cr/Ni, Fe/Ni, and Al/Sr were distinctive for multifloral local samples. The correlations K/Fe, K/Cu, Σ major/Cu, Fe/Cu, Co/Mo, and Se/Sn were distinctive for multifloral imported samples.

Table 5 Pearson's correlation coefficients between selected elements in honey

Elements	Monofloral	Multifloral (imported)	Multifloral (local)
Na/Mg	0.49	0.30	0.70
Na/K	0.49	-0.33	0.53
Na/Ca	0.70	0.22	0.85
Na/ Σ major	0.51	-0.23	0.61
Na/Se	0.67	0.18	0.91
Na/Mo	-0.15	0.41	0.43
Mg/K	0.97	0.46	0.50
Mg/Ca	0.89	0.48	0.72
Mg/ Σ major	0.97	0.56	0.58
Mg/Se	0.03	0.19	0.71
Mg/P	0.93	0.72	0.37
Mg/Fe	-0.21	0.55	-0.68
Mg/Co	0.72	0.57	-0.60
Mg/Ni	0.79	0.86	-0.63
Mg/Cu	0.51	0.54	-0.42
P/K	0.95	0.89	0.60
P/Ca	0.85	0.42	0.35
P/ Σ major	0.96	0.91	0.57
P/Fe	-0.36	0.72	-0.25
P/Ni	0.84	0.78	-0.30
P/Cu	0.44	0.80	-0.46
K/Ca	0.94	0.42	0.87
K/ Σ major	1.00	0.99	0.99
K/Fe	-0.15	0.55	-0.40
K/Ni	0.75	0.61	-0.26
K/Cu	0.52	0.64	-0.45
Ca/ Σ major	0.95	0.51	0.91
Ca/Se	0.30	0.30	0.80
Σ major/Fe	-0.15	0.59	-0.56
Σ major/Cu	0.53	0.65	-0.42
Cr/Fe	0.33	0.45	0.79
Cr/Co	0.01	0.27	0.83
Cr/Ni	0.00	0.17	0.73
Mn/Fe	0.21	0.43	0.61
Mn/Co	0.96	0.53	0.64
Mn/Ni	0.77	-0.01	0.51
Mn/Cu	0.86	0.53	0.17
Mn/Al	0.99	0.97	0.64
Mn/Sr	0.59	0.06	0.54
Mn/Sn	0.29	0.03	0.74
Mn/Ba	0.61	0.32	0.63
Fe/Co	0.00	0.72	0.96
Fe/Cu	0.44	0.88	0.29
Fe/Ni	-0.22	0.49	0.93
Fe/Sn	0.44	0.35	0.77
Co/Ni	0.90	0.60	0.86
Co/Cu	0.80	0.79	0.33
Co/Mo	0.05	0.56	-0.12
Ni/Sn	0.05	0.10	0.66
Ni/Mo	0.36	0.52	-0.33
Ni/Cu	0.72	0.52	0.24
Cu/Al	0.89	0.44	0.71
Mo/Ba	-0.03	0.51	0.11
Al/Sr	0.61	0.20	0.77
Al/Sn	0.22	0.06	0.55
Al/Ba	0.67	0.54	0.45
Se/Sn	0.31	0.69	0.07
Se/Ba	0.55	0.51	0.77
Sr/Ba	0.84	0.48	0.74
Sr/Sn	0.65	0.71	0.77

Table 6 Statistical results for major and trace elements in honey samples.

Element	This study		Solayman et al. (2016) ^a	
	Mean ± SD	Range (min-max)	Mean ± SD	Range (min-max)
Major elements (mg/kg)				
Na	66.9 ± 49.3	19.2–258	96.5 ± 80.6	3.23–227
Mg	43.4 ± 27.4	9.1–107.8	74 ± 163	2.18–563
P	74.3 ± 48.4	12.3–187.3	84 ± 48	29–118
K	718 ± 746	27.7–2404	742 ± 453	40–1349
Ca	100.0 ± 44.9	26.9–205	84 ± 68	4.85–218
Essential trace elements (mg/kg)				
Cr	0.180 ± 0.053	0.06–0.306	0.15 ± 0.44	ND–0.47
Mn	1.96 ± 3.48	0.13–13.40	1.42 ± 1.47	ND–4.35
Fe	19.4 ± 11.0	7.03–49.3	30 ± 65	0.40–224
Co	0.011 ± 0.010	0.003–0.049	0.17 ± 0.29	ND–0.80
Ni	0.123 ± 0.181	0.030–0.688	1.24 ± 2.62	ND–9.00
Cu	0.606 ± 0.478	0.145–2.34	1.99 ± 4.31	0.05–17.3
Zn	1.67 ± 0.785	0.48–3.72	9.33 ± 0.681	0.23–73.6
Se	0.041 ± 0.019	0.005–0.105	0.01 ± 0.01	ND–0.03
Mo	0.030 ± 0.013	0.020–0.096	0.23 ± 0.30	0.01–0.44
Selected nonessential trace elements (µg/kg)				
Li	18.3 ± 18.0	0.29–72.9	3.8 ± 2.6 ^b	0.90–7.8 ^b
Be	1.30 ± 0.43	0.42–2.31	9.93 ± 0.05	9.83–10.0
Al	4670 ± 3510	2660–21900	5120 ± 3610	1390–11360
V	11.6 ± 6.80	5.14–37.7	30 ± 30	10–50
Ga	13.6 ± 18.8	ND–78.6	11.6 ± 0.50 ^b	10.5–14.8 ^b
Sr	580 ± 440	75.0–1620	1630 ± 1030	900–2360
Ag	35.2 ± 69.9	5.70–386	299 ± 419	3.21–596
Sn	56.9 ± 29.9	33.6–186	4.8 ± 2.1 ^c	1.5–8.5 ^c
Cs	4.80 ± 8.60	0.17–37.9	4.35 ± 2.62 ^b	2.1–6.9 ^b
Ba	251 ± 233	59–1060	198 ± 75 ^b	147–286 ^b
U	1.23 ± 0.65	0.49–3.23	4.3 ± 2.1 ^b	3.8–6.1 ^b
Potentially toxic elements (µg/kg)				
As	3.10 ± 1.50	ND–6.90	50 ± 40	ND–100
Cd	3.02 ± 1.18	0.38–6.47	90 ± 130	0.17–373
Pb	13.6 ± 6.60	3.61–31.5	424 ± 839	0.36–3232

^a Extracted from 32 publications between the years 1999 and 2014

^b Wetwitayaklung et al. (2018)

^c Squadrone et al. (2020b)

Comparison with previous studies

A large number of studies (above 200) have been reported for multielemental measurements of basic elements (major and trace) in honey from popular honey-producing countries. A summary of these results was reported in a review by Solayman et al. (2016). Table 6 presents a comparison between the results of our study and those by Solayman et al. (2016). Regarding major elements, the same trend was followed, with K being the largest and Mg being the smallest. Major elements (Na, K, Ca, Mg, and P) are abundant in the

environment and available to plants from soil, water, and fertilizers. Regarding essential elements, an agreement was observed between the levels of Fe, Cr, Mn, Cr, and Se. However, surprisingly, in the review of Solayman et al. (2016), a sample had 224 mg/kg Fe. The levels of Co, Ni, Cu, Zn, and Mo in our study were significantly lower than those reported in the review of Solayman et al. Regarding nonessential elements, Al, Sr, and Ba were the most abundant elements, and their ranges in our study and the review of Solayman et al. were similar. The three elements are available in soil at significant levels. Sn levels were significantly higher in our study, while

Table 7 Statistical results for rare earth elements ($\mu\text{g}/\text{kg}$) in honey samples.

Element	This study		Drivelos et al. (2021) ^a	
	Mean \pm SD	Range (min-max)	Median	Range (min-max)
Y	2.42 \pm 0.92	1.32–4.43	0.65	ND–5.69
La	19.4 \pm 21.9	3.9–78	0.96	ND–9.85
Ce	66 \pm 82	15.1–314	ND	ND–16.5
Pr	5.5 \pm 7.7	1.10–28.3	0.26	ND–1.65
Nd	20.8 \pm 31.4	3.40–116	0.63	ND–6.34
Sm	3.60 \pm 4.20	0.90–15.9	0.23	ND–1.60
Eu	0.47 \pm 0.38	0.22–1.28	0.13	ND–0.98
Gd	1.40 \pm 1.80	ND–6.3	0.50	ND–3.81
Tb	0.23 \pm 0.12	0.10–0.58	0.09	ND–0.33
Dy	1.01 \pm 0.38	0.41–1.70	0.15	ND–1.13
Ho	0.37 \pm 0.21	0.11–1.07	0.06	ND–0.23
Er	0.41 \pm 0.24	ND–0.93	0.12	ND–0.58
Tm	0.18 \pm 0.18	ND–0.80	0.052	ND–0.13
Yb	0.55 \pm 0.28	0.16–1.36	0.08	ND–0.26
Lu	0.28 \pm 0.18	0.02–0.86	0.04	ND–0.10

^a 57 samples from Poland, 30 samples from Greece, and 6 samples from other European countries. ND not detected

Ag levels were significantly lower than those reported in other studies. Regarding potentially toxic elements (As, Cd, and Pb), the results reported by Solayman et al. were alarming, 50, 90, and 424 $\mu\text{g}/\text{kg}$ for As, Cd, and Pb, respectively. Regulation 1005/2015 of the European Community set a maximum limit (100 $\mu\text{g}/\text{kg}$) for lead in honey. While Cd and As are still unregulated, their limits are expected to be much lower than 100 $\mu\text{g}/\text{kg}$. The World Health Organization (WHO 2017) set regulation values of 2.0, 10, and 10 $\mu\text{g}/\text{L}$ for Cd, As, and Pb in drinking water. The results of our study are much lower, indicating the safety of honey in Jordanian markets for human consumption.

Rare earth elements (REEs) have been proven to be reliable markers in foods, including honey, since they are not affected by environmental conditions (Drivelos et al. 2021; Squadrone et al. 2020c). Our study found 145 $\mu\text{g}/\text{kg}$ total ΣREEs and 95 $\mu\text{g}/\text{kg}$ in imported samples and 203 $\mu\text{g}/\text{kg}$ in local samples. Squadrone et al. (2020c) reported a range of 9.0–65 $\mu\text{g}/\text{kg}$ in various types of honey samples. Table 7 presents the levels of REEs from our study and a study reported by Drivelos et al. (2021). The levels of REEs in our study were 3 to 10 times larger than those in Drivelos et al. (2021).

Conclusions

Forty-six elements, including 15 rare earth elements, were monitored in honey in Jordan, including multifloral local

samples ($n=12$), multifloral imported samples ($n=10$), and monofloral imported samples ($n=8$). Our results indicated a wide range for each element, with an order larger than 10 between maximum/minimum concentrations. The precision (% RSD) was larger than 50% for most elements, supporting the importance of geographic origin. Potassium, K, dominated major elements with a mean of 718 mg/kg. Four essential elements, Fe, Mn, Zn, and Cu, had significant concentrations, and Al, Sr, and Ba were the only nonessential elements with significant concentrations. Levels of potentially toxic elements, As, Cd, and Pb, in all samples were much lower than their guideline limits, indicating the safety of honey in Jordan for human consumption.

Elemental profiles have a strong influence on the discrimination between different floral types and origins of honey. Selected elements may be adequate for monofloral honey due to the importance of botanical origin. However, for multifloral honey, a large number of samples combined with monitoring of more elements are required. Multivariate methods combining correlations and statistical models, such as principal component analysis (PCA), are needed to perform exploratory and predictive analyses. Additionally, elemental profiles for water and soil from various geographic regions are essential for accurate assessment and authentication.

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Author contribution The authors contributed to this research in the following manner. Y. Tahboub and A. Al-Ghzawi supervised all stages from the collection of samples to the writing of the manuscript. S. Al-Zayadneh and M. AlGhotani validated the methods and performed elemental analysis.

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Declarations

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