

Global assessment of Algerian honeys quality by palynological, physicochemical analyses, trace elements and potentially toxic elements screening

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Abstract: The quality of twenty Algerian honeys was assessed based on their palynological and physicochemical properties, and their trace and toxic elements composition. A qualitative pollen analysis was conducted to estimate the botanical origin. The physicochemical analyses included moisture content, pH, electrical conductivity, 5-hydroxymethylfurfural (HMF), colour, and the content of 3 sugars (fructose, glucose, and sucrose). The analysis of mineral and heavy metals included Zn, Mn, Fe, Cu, Cr, Ni, Pb, Cd, and As. The pollen spectrum showed a great diversity with 60 taxa identified. The palynological analyses revealed the presence of 15 honeys with pollen dominance (unifloral): *Citrus* sp., *Eucalyptus* sp., *Ziziphus lotus* (L.) Lam., *Sinapis arvensis* L., *Dorycnium* sp., *Bupleurum* sp., *Echium* sp., *Lotus* sp., and 5 honeys without pollen dominance (polyfloral). The physicochemical results showed that the samples conform to international quality standards, with few exceptions related to HMF, mainly due to beekeeping practices. The colour was from water white to dark amber. Pb and Cd concentrations were found to be below the maximum residue limits set by the European Directive with which the toxic elements were compared. These results would contribute to the assessment of Algerian honey and provide a database for the regulation of honey trade and consumer protection.

Key words: honey, Algeria, mellisopalynology, quality, toxic elements

Celokupna ocena kakovosti alžirskih medov s palinološkimi in fizikalno-kemijskimi analizami ter pregledom vsebnosti elementov v sledeh in potencialno strupenih elementov

Izvleček: Sestava in kakovost dvajsetih vzorcev alžirskega medu sta bili ocenjeni na osnovi palinoloških in kemijsko-fizikalnih analiz ter vsebnosti elementov sledeh in potencialno strupenih elementov. Kvalitativna analiza peloda v medu je bila narejena z namenom ugotoviti njegovo botanično poreklo. Fizikalno kemijske analize so obsegale določanje vsebnosti vode, pH, električno prevodnost, vsebnost HMF (hidroksimetilfurfural), barve in vsebnost treh sladkorjev (fruktoze, glukoze in saharoze). Analiza elementov in težkih kovin je obsegala analizo vsebnosti Zn, Mn, Fe, Cu, Cr, Ni, Pb, Cd in As. Pelodni spekter je pokazal veliko raznolikost s šestdesetimi ugotovljenimi taksoni. Pri petnajstih vzorcih medu je bila ugotovljena dominanca posameznih rastlinskih vrst oziroma rodov (unifloralni med) kot so: *Citrus* sp., *Eucalyptus* sp., *Ziziphus lotus* (L.) Lam., *Sinapis arvensis* L., *Dorycnium* sp., *Bupleurum* sp., *Echium* sp. in *Lotus* sp., pri petih vzorcih dominacije posameznih vrst ni bilo (polifloralni med). Fizikalno-kemijske analize so pokazale, da vzorci medu ustrezajo mednarodnim standardom kakovosti, z nekaj izjemami, ki se nanašajo na HMF, v glavnem zaradi razlik v čebelarjenju. Barva medu je bila od prozorne do jantrne. Vsebnosti Pb in Cd so bile znatno pod maksimalnimi vrednostmi, ki jih predpisuje Evropska direktiva o vsebnosti strupenih elementov v medu. Rezultati te raziskave prispevajo k oceni alžirskega medu in dajejo osnovne podatke za trgovanje z medom in zaščito potrošnikov.

Ključne besede: med, Alžirija, melisopalnologija, kakovost, strupeni elementi

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1 INTRODUCTION

Honey is a natural sweet substance mainly composed of carbohydrates at around 80–85 % (w/w). Its moisture content is about 15–20 % (w/w), and to a minor extent of about 1 % other components are represented such as organic compounds, inorganic ions, enzymes, vitamins, hormones, flavonoids, proteins, and amino acids. Several nutritional and therapeutic properties like antioxidant, antibacterial and anti-inflammatory activities are recognized for honey due to this composition (Aljohar et al., 2018).

In general, honey quality and its composition are related to several factors such as geographical and botanical origins with their characteristics, and the climatic and seasonal conditions. The Codex Alimentarius (Codex Alimentarius, 2001) as well as the European Community (Council Directive, 2002) have both adopted standards to assess honey quality. These standards established a maximum moisture content of 20 %, and hydroxymethylfurfural (HMF/ a result of fructose degradation) at 40 mg kg⁻¹. These two parameters are considered among the most widely used quality parameters to determine honey stability and freshness.

Certain metals are recognized to be essentials for the different metabolism needs. The following trace elements: iron (Fe), zinc (Zn), copper (Cu), manganese (Mn), chromium (Cr) and nickel (Ni) have several physiological and biochemical activities making them indispensable for good cellular metabolism (APVMA, 2015). However, the presence of these elements in high amounts can have an opposite effect and be harmful instead of.

Nonessential elements, such as lead (Pb), cadmium (Cd) and arsenic (As) are of no use or interest for biological functioning. Moreover, they can be very toxic even at low rates. The contamination of honey by these elements may be due to mining and industrial pollution, or due to the use of Cd- or As-based fertilizers that can be spread into the soil and water and consequently can contaminate plants harvested by bees.

Metal contamination of food is a problem of major concern. In order to protect public health, a limit of 1 and 0.1 mg kg⁻¹ for Pb and Cd respectively in honey has been proposed to the European Community (Bal-Prylypko et al., 2018; Bogdanov et al., 2003). On 2015, the European Community, set a limit of 0.1 mg kg⁻¹ for Pb for honey consumption by children and persons with specific dietary needs (Commission Regulation (EU), 2015), while there is no specification about the maximum acceptable limit for Arsenic (Bal-Prylypko et al., 2018).

The determination of heavy metals in honey is

therefore of great interest, mainly for quality control and nutritional purposes. It is important to note that there are no specific limits established by the Algerian regulation concerning the heavy metals in honey. Consequently, we are required to adhere to the standards of the codex alimentarius or those of the countries where our products are being received.

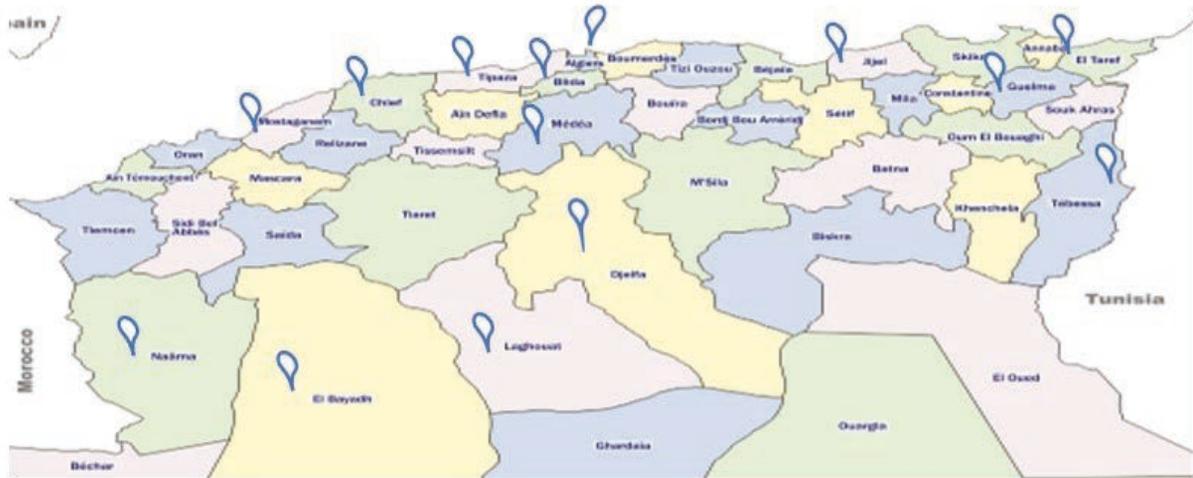
Honey in Algeria has an excellent reputation and a notable place among Algerian consumers, as it is consumed for both nutritional and therapeutic purposes. The main feature of Algerian honey is its organoleptic quality which is also related to the botanical and geographical origins. Algerian honey possesses comparable and competing advantages, such as a diverse array of flora (cultivated crops, wild plants, forests, and mountains), a range of climates (Mediterranean and Saharan climates) for production, and expansive unpolluted fields. Lots of research have been carried out to evaluate the physicochemical characteristics but a little has been conducted on minerals and toxic elements aspect of Algerian honey. It is therefore crucial to define its quality parameters to establish standards for specific Algerian honey and to protect the consumer from frauds and health risk. The present study aims to assess the quality of Algerian honey according to its botanical origin and to verify its compliance with the standards set by the Codex and the European Community.

Twenty samples of different botanical and geographical origins were analysed. We first carried out a melissopalynological analysis to determine the pollen spectrum and botanical origin, a physicochemical analysis to determine the corresponding quality: colour, moisture, acidity, pH, electrical conductivity, and HMF. Sugars (fructose, glucose, and sucrose) were also determined. Finally, certain trace elements and heavy metals (Zn, Mn, Fe, Cu, Cr, Ni, Pb, Cd, and As) were analysed to certify the safety of these honeys according to the European Directive.

2 MATERIALS AND METHODS

2.1 HONEY SAMPLES

Twenty honey samples (250 g each); produced in Algeria; were collected from beekeepers over two campaigns in 2020 and 2021. Collected samples were stored at a temperature of +4 °C in the refrigerator throughout the analysis process. The location of samples according to their geographical origin is shown in Figure 1.



Samples Nbr	Locality	Samples Nbr	Locality
E1/E32	Tessala El Mardja/ Beni Messous/ Alger	E39	Chahbounia/Médéa
E2/E41	Boufarik/ Blida	E40	Tébessa
E31	El Bayadh	E42	Naama
E33	Kolea/ Tipaza	E44/E46	Chlef
E34/E38/E51/E69	Aflou /Laghouat	E71	El Taref
E35	Guelma	E74	Mostaganem
E36	Djelfa	E78	Jijel

Figure 1: Location of honey samples (presented in map and list)

2.2 METHODS OF ANALYSIS

2.2.1 Qualitative pollen analysis

Pollen grains were identified and counted according to the harmonized methods of melissopalynology described by Louveaux et al. (1977) and Von Der Ohe et al. (2004). This method establishes all the pollen types present by determining their pollen frequencies expressed as a percentage relative to the total number of pollen grains counted (500 grains in our case). The pollens are then divided into four pollen frequency classes: predominant pollens (+ 45 %); secondary pollens (16-45 %); important minor pollens (3-15 %); and minor pollens (< 3 %). The pollen grains were observed using an optical microscope, and pollen types were identified by comparing them with the pollen reference established by (Ricciardelli d'Albore, 1998). We have also used our reference collection of pollen from plants with recognized scientific and local names. The pollen grains were classified as pollen type, as a genus, or as a single species whenever it was possible (Louveaux et al., 1977).

Taxa distribution frequencies have been calculated according to the number of honey samples in which they are found. As explained by Feller-Demalsy et al. (1989), they are classified as very frequent taxa (+ 50 %); fre-

quent taxa (20-50 %); rare taxa (10-20 %); sporadic taxa (-10 %).

2.2.2 Physicochemical analysis

Except for colour, all physicochemical parameters (water content, HMF, pH, free acidity, and electrical conductivity) were analysed following the analysis techniques recommended by the International Honey Commission (IHC) published by Bogdanov et al. (2004) and updated by Bogdanov (2009).

Water content was measured by refractive index using an ATAGO NAR-3T refractometer. Electrical conductivity was determined by using a conductivity meter (CORNING). pH and free acidity were measured by using a HANNA pH meter. HMF was measured by a spectral method using a CECIL CS-3041 UV-Vis spectrophotometer. Colour measurement was performed according to Bianchi method as described by Lacerda et al. (2010) and Ferreira et al. (2009), a 50 % (w/v) honey solution was prepared with warm water between 45 ° and 50 °C, then filtered to remove any coarse particles and measured by absorbance reading at 635 nm. Colour intensity was determined using the Pfund scale according to the equation (1):

$$Pfund = - 38.70 + 371.39 \times Abs \quad (1)$$

The carbohydrate profile was determined using an Agilent 1260 Infinity II HPLC equipped with a DAD detector and Open Lab CDS data processing software, sugars were separated using an Ammonia (NH₃) USP L8 analytical column (25 cm × 4.6 mm, 5 mm i. d.). Standard solutions of fructose (2 g %), glucose (2 g %), and sucrose (0.5 g %) were prepared in ultrapure water, and sample preparation was carried out by dissolving 2 g of honey in 20 ml distilled water. The analysis procedure was conducted following Aljohar et al. (2018). Identification of sugars, their peaks and their concentrations was made possible by comparing their chromatograms, retention times, and the surfaces of their peaks to those obtained from standard sugar solutions.

2.2.3 Trace and toxic elements analysis

Six minerals and three heavy metals were the subject of our study. 200 mg of previously homogenized sample to which 7 ml HNO₃ (65 %) and 1 ml H₂O₂ (30 %) were added, the mixture was placed in the digestion microwave (Ethos Easy - Milestone Connect microwave). The concentration of each analyte was determined in the sample and blank solutions using an ICP-MS inductively coupled plasma mass spectrometer (ICAP-RQ Thermo Scientific). The results were expressed as mg kg⁻¹ of honey for Zn, Mn, Fe, Cu, Cr, and Ni and per µg kg⁻¹ of honey for Cd, Pb, and As.

2.2.4 Statistical analysis

The results were reported as a mean ± standard de-

viation. To investigate correlations among several variables, including physicochemical parameters, mineral concentrations, and toxic metal levels, a Spearman correlation analysis was performed and a graphic was generated using a corrplot package. Multiple factor analysis (MFA) was performed using the FactoMineR on a data frame containing several sets of qualitative and quantitative variables structured into three groups; physical, mineral (trace elements), and heavy metal (toxic elements). This analysis aims to describe the characteristics of the honey samples spread over two groups: honey with pollen dominance (unifloral) and honey without pollen dominance (polyfloral). All statistical analyses were carried out using R software (version 4.2.2).

3 RESULTS AND DISCUSSION

3.1 QUALITATIVE POLLEN ANALYSIS

Sixty taxa were identified for the twenty honey samples analysed, of which 55 were nectariferous, five were nectarless, and of which 20 were sporadic, 22 were rare, 13 were frequent, and 5 were very frequent (Table 1).

From the distribution of identified taxa, the presence of 18 pollen types; corresponding to the classes frequent and very frequent; are represented in the four pollen frequency classes (Figure 2).

The pollen analysis (Tab. 2 and 3) highlighted the presence of 15 unifloral honeys: *Sinapis arvensis* L., *Citrus* sp., *Eucalyptus* sp., *Ziziphus lotus* (L.) Lam., *Echium* sp., *Bupleurum* sp., Fabaceae honey with the genera of *Dorycnium* sp. and *Lotus* sp. and one unifloral honey of *Peganum harmala* L.. The 05 remaining samples were polyfloral honey composed of secondary pollens from 1 to 3 taxa maximum.

Table 1: Distribution frequencies of taxa in the 20 samples of honey

Relative frequency classes (%)	Taxa
Sporadic < 10%	<i>Anthyllis</i> (5 %), <i>Araceae</i> (5 %), <i>Bubplerum</i> sp. (5 %), <i>Buxaceae</i> (5 %), <i>Castanea</i> sp. (5 %), <i>Chenopodiaceae</i> (5 %), <i>Cucurbitaceae</i> (5 %), <i>Daucus carota</i> L. (5 %), <i>Dipsacaceae</i> (5 %), <i>Dorycnium</i> sp. (5 %), <i>Ephedraceae</i> (5 %), <i>Geraniaceae</i> (5 %), <i>Melilotus</i> sp. (5 %), <i>Orobanchaceae</i> (5 %), <i>Palmeae</i> (5 %), <i>Ranunculaceae</i> (5 %), <i>Raffeciaceae</i> (5 %), <i>Rubiaceae</i> (5 %), <i>Salix</i> sp. (5 %), <i>Sophora</i> sp. (5 %)
Rare 10-20 %	<i>Anacardiaceae</i> (10 %), <i>Annonaceae</i> (10 %), <i>Betulaceae</i> (10 %), <i>Borago officinalis</i> L. (10 %), <i>Cupressaceae</i> (10 %), <i>Echium</i> sp. (10 %), <i>Erica</i> sp. (10 %) <i>Liliaceae</i> (10 %), <i>Lotus</i> sp. (10 %), <i>Malvaceae</i> (10 %), <i>Muscari</i> sp. (10 %), <i>Ononis</i> sp. (10 %), <i>Sinapis arvensis</i> L. (10 %), <i>Smilacaceae</i> (10 %), <i>Taraxacum</i> sp. (10 %), <i>Fagaceae</i> (15 %), <i>Hedysarum coronarium</i> L. (15 %), <i>Peganum harmala</i> L. (15 %), <i>Senecio vulgaris</i> L. (15 %), <i>Verbenaceae</i> (15 %), <i>Citrus</i> sp. (20 %), <i>Salicaceae</i> (20 %)
Frequent 20-50 %	<i>Diplotaxis erucoides</i> (L.) DC. (25 %), <i>Ziziphus lotus</i> (L.) Lam. (25 %), <i>Boraginaceae</i> (30 %), <i>Mimosaceae</i> (30 %), <i>Myrtaceae</i> (30 %), <i>Oxalis</i> sp. (30 %), <i>Brassicaceae</i> (35 %), <i>Ericaceae</i> (35 %), <i>Oleaceae</i> (35 %), <i>Polygonaceae</i> (35 %), <i>Poaceae</i> (40 %), <i>Eucalyptus</i> sp. (50 %), <i>Euphorbia</i> sp. (50 %)
Very frequent > 50 %	<i>Fabaceae</i> (60 %), <i>Lamiaceae</i> (60 %), <i>Asteraceae</i> (70 %), <i>Rosaceae</i> (75 %), <i>Apiaceae</i> (90 %)

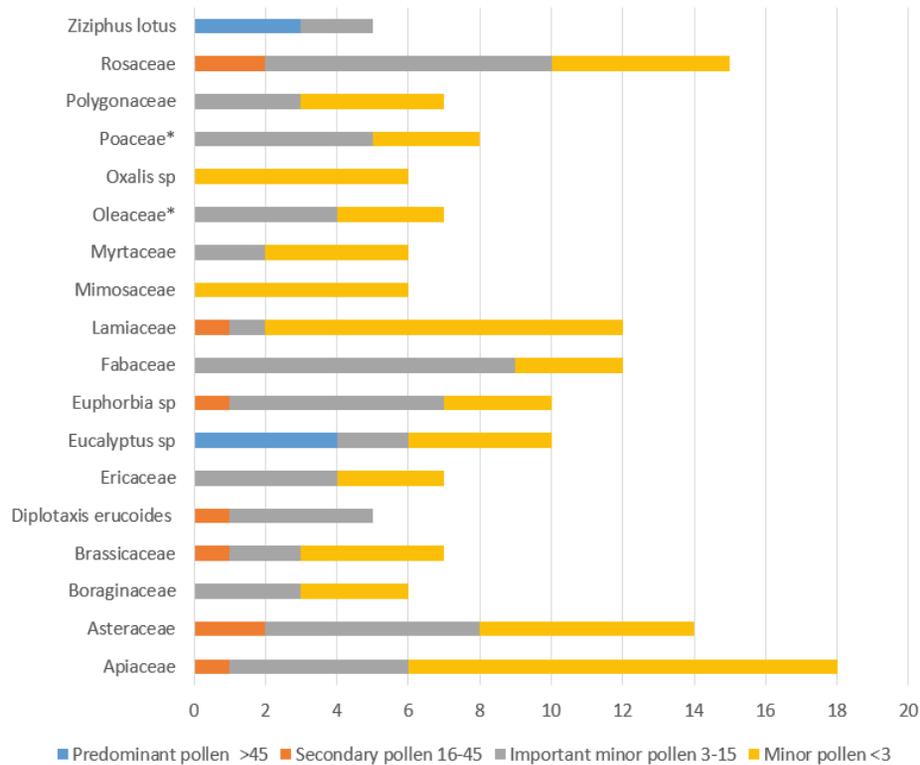


Figure 2: Presence of 18 pollen types; corresponding to frequent and very frequent classes; represented in four classes of pollen frequency: predominant pollen > 45 %, secondary pollen (16–45 %), important minor pollen (3–15 %) and minor pollen < 3 %. *Nectarless species

Samples E2 and E41 showed *Citrus* sp. frequencies of 30 % and 21 % respectively, Persano Oddo et al. (2004) reported that citrus pollen (*Citrus* sp.) is under-represented to a degree more or less important according to the different species or cultivars. A minimum frequency of citrus pollen varying from 10 to 20 % is accepted to be considered as citrus honey (Louveaux et al., 1977; Reyes, 2017; Seraglio et al., 2021). In addition to that, the physicochemical and sensory properties of these samples are in agreement with those described by Persano Oddo & Piro (2004) for citrus honey. In a study covering Mitidja zone, Benaziza-Bouchema et al. (2010) presented values ranging from 21 to 69 % for citrus honey, these values corroborate very well with our aforementioned values.

The eucalyptus pollen content ranged between 70 to 73 %. According to Persano Oddo & Piro (2004) eucalyptus pollen is overrepresented in honeys. Our results corroborate very well with those of (Benaziza-Bouchema & Schweitzer, 2010) and (Makhloufi et al., 2010) reporting both values of eucalyptus pollen more than 70 %.

The *Ziziphus lotus* pollen content ranged between 52 and 89 % of the pollen spectra. These values are consistent with those reported by Mekious et al. (2015); Zer-

rouk et al. (2017) for 45.75 to 97.12 % and 45.3 to 93.7% respectively.

3.2 PHYSICOCHEMICAL ANALYSES

The results of the physicochemical analyses are summarized in Table 4. They are presented according to the results of the pollen analysis. Hence, we distinguish two groups, the unifloral honey group (whose pollen spectrum includes dominant pollen) and the polyfloral honey group (whose pollen spectrum does not include any dominant pollen).

Electrical conductivity values; for all samples; were between 0.12-0.68 mS cm⁻¹. A maximum of 0.8 mS cm⁻¹ is established by the Codex Alimentarius for nectar honey, which is the case for all our samples.

The highest pH mean value was registered for unifloral honey group with 4.09. According to Gonnet (1986), nectar honeys, or a mixture of nectar and honeydew honeys have a pH between 3.5 and 4.5, honeydew honeys have a pH of 4.5 and 5.5. Our samples were therefore nectar honey. The range of pH values for all samples was 3.41-5.07. The high pH values obtained were due to

Table 2: Predominant and secondary pollen types in 20 honey samples

Sample Number	Predominant pollen (>45 %)	Secondary pollen (16-45 %)
E2	-----	<i>Citrus</i> sp. (30 %), <i>Sinapis arvensis</i> L. (18 %)
E41	-----	<i>Citrus</i> sp. (21 %), <i>Diploptaxis eruroides</i> (L.) DC. (20 %), <i>Lotus</i> sp. (17 %)
E32	<i>Eucalyptus</i> sp. (71 %)	-----
E44	<i>Eucalyptus</i> sp. (70 %)	-----
E74	<i>Eucalyptus</i> sp. (73 %)	-----
E78	<i>Eucalyptus</i> sp. (71 %)	-----
E36	<i>Ziziphus lotus</i> (L) Lam. (52 %)	-----
E46	<i>Zyziphus Lotus</i> (L) Lam. (81 %)	-----
E69	<i>Zyziphus Lotus</i> (L.) Lam (89 %)	-----
E1	<i>Sinapis arvensis</i> L. (83%)	-----
E31	<i>Dorycnium</i> sp. (72 %)	-----
E33	<i>Echium</i> sp. (85 %)	-----
E35	<i>Bupleurum</i> sp. (62 %)	-----
E38	<i>Lotus</i> sp. (46 %)	-----
E39	<i>Peganum harmala</i> L. (81 %)	-----
E34	-----	<i>Peganum harmala</i> L. (33 %), Rosaceae (21 %), <i>Euphorbia</i> sp. (19 %)
E40	-----	<i>Melilotus</i> sp. (28 %)
E42	-----	<i>Anthyllis</i> sp. (38 %), <i>Senecio vulgaris</i> L. (27 %)
E51	-----	Apiaceae (23 %), Rosaceae (21 %), Brassicaceae (20 %)
E71	-----	Lamiaceae (38 %), <i>Erica</i> sp. (33 %)

the presence of jujube honey which is characterized by a high pH value (5.1 for E69) such a value is also reported by Mekious et al. (2015) and Zerrouk et al. (2017) with 5.17 ± 0.48 and 5.5 ± 0.6 for jujube honey respectively.

The highest free acidity mean value was registered for the polyfloral honey group with 25.03 meq kg⁻¹. Whereas, free acidity values for all samples ranged from 10.25-34 meq kg⁻¹. The (Codex Alimentarius, 2001; Council Directive, 2002) set a maximum limit of 50 meq kg⁻¹ of honey. Our samples, therefore, complied with in-

ternational standards showing an absence of undesirable fermentations.

Moisture content for all samples was in the range of 13.6 to 18.25 %. A high value was registered for unifloral honey group with 16.25 %. The water content provides information on the maturity of the honey and also determines its conservation (Terrab et al., 2003). A low water content preserves the honey against microbial development (Bogdanov, 2009). Whereas, with a high water content, honey tends to ferment easily (El Sohaimy et al., 2015).

The HMF values for all honey samples ranged between 0-184.7 mg kg⁻¹. Unifloral honey showed the highest mean value with 27.67 mg kg⁻¹. Indeed, three samples from this group showed values above the limit set by the European and international standards (40 mg kg⁻¹ of honey), namely E1 and E46 for 59.0 and 50.5 mg kg⁻¹ respectively, and even more than the recommended limit set for honeys from tropical climate and blends of these honeys (80 mg kg⁻¹ of honey) (Council Directive, 2002) for E44 with 184.7 mg kg⁻¹. The HMF does not occur in newly harvested honey but its content rises through conditioning and storage. To prevent the granulation of honey and also to decrease its viscosity, beekeepers usually, tend to warm it during the harvesting process. The quality of honey is not affected at temperatures of 32–40 °C. However, the application of higher temperatures tends to increase the HMF levels in honey (Anklam, 1998). HMF is therefore considered an indicator of freshness and/or overheating of honey.

In general, and for high-quality honey, it is recommended a maximum moisture content of 18 % and an HMF rate of no more than 15 mg kg⁻¹ of honey. At these rates, the risk of fermentation is avoided and the honey remains fresh until its final consumption (Schweitzer, 1998). The results of these two parameters for our samples showed a maximum water content of 18.3 % which indicates respect for the honey maturity process before harvesting and compliance with international standards. Whereas, the HMF values were above the level for three samples reaching a maximum value of 184.7 mg kg⁻¹ of honey. Knowing that consumption of HMF may create certain health problems such as irritation of the mucous membranes of the upper respiratory tract, eyes, skin, etc... if it is consumed beyond the recommended limits (Pastoriza de la Cueva et al., 2017; Shapla et al., 2018). HMF can also be metabolized to 5-sulfooxymethylfurfural (SMF) by sulfotransferases (Pastoriza de la Cueva et al., 2017), the SMF is an intermediate molecule that can bind to DNA and induce mutagenic effects. Svendsen et

Table 3: Important minor and minor pollen types in 20 honey samples

Sample Number	Important minor pollen (3-15 %)	Minor pollen (< 3 %)
E2	Oleaceae (13 %), Polygonaceae (11 %), Dipsacaceae and Rosaceae (7 %), Fabaceae (4 %), Apiaceae and Asteraceae (3 %)	<i>Salix</i> sp., Poaceae, Boraginaceae, Ericaceae, Lamiaceae, Liliaceae, Malvaceae, Myrtaceae, Orobanchaceae, <i>Oxalis</i> sp.,
E41	Boraginaceae (7 %), Oleaceae (6 %), <i>Senecio vulgaris</i> L. and Fabaceae (5 %), Rosaceae (4 %), <i>Eucalyptus</i> sp. and Polygonaceae (3 %)	<i>Oxalis</i> sp., Rubiaceae, Smilacaceae, Annonaceae, Apiaceae, Malvaceae, Geraniaceae,
E32	<i>Hedysarum coronarium</i> L. (10 %), Ericaceae (7 %), <i>Euphorbia</i> sp. (5 %), Asteraceae (4 %)	Brassicaceae, Apiaceae, Lamiaceae, Mimosaceae
E44	Oleaceae (10 %) Salicaceae and Poaceae (5%), <i>Hedysarum coronarium</i> L. (4 %), Brassicaceae (3 %)	Apiaceae, Mimosaceae, Asteraceae, Betulaceae, Ephe-draceae, Fagaceae, Lamiaceae, <i>Oxalis</i> sp., Rosaceae, Verbenaceae,
E74	Oleaceae (13 %), <i>Erica</i> sp. (8 %), Fabaceae (5 %)	Apiaceae, Asteraceae
E78	Fabaceae (13 %), Apiaceae (6 %), Rosaceae (5 %), Ericaceae (3 %)	<i>Taraxacum</i> sp., <i>Castanea</i> sp., Lamiaceae
E36	Rosaceae (14 %), Poaceae (13 %), Fabaceae (6 %), Asteraceae and Myrtaceae (3 %)	Apiaceae, Araceae, Brassicaceae, <i>Euphorbia</i> sp., Boraginaceae, Lamiaceae, Mimosaceae
E46	Apiaceae (8 %), <i>Hedysarum coronarium</i> L. (6 %), Asteraceae (5 %)	-----
E69	<i>Ononis</i> sp. (5 %), <i>Euphorbia</i> sp. (3 %)	Apiaceae, Fagaceae, Rosaceae, Asteraceae, <i>Borago officinalis</i> L.
E1	<i>Citrus</i> sp. (11 %), Fabaceae (4 %)	Apiaceae, Oleaceae, Rosaceae, Myrtaceae, Poaceae, <i>Taraxacum</i> sp.
E31	<i>Echium</i> sp. and Polygonaceae (8 %), Palmeae and Rhamnaceae (3 %)	<i>Euphorbia</i> sp., Poaceae, Apiaceae, Asteraceae, Brassicaceae, Lamiaceae
E33	Fabaceae, Salicaceae (4 %), Asteraceae (3 %)	Ericaceae, <i>Eucalyptus</i> sp., Lamiaceae, <i>Oxalis</i> sp., Apiaceae, Brassicaceae
E35	Rosaceae (13 %), <i>Eucalyptus</i> sp. (11 %), Asteraceae (5 %), Brassicaceae, Ericaceae (3 %)	Lamiaceae, <i>Oxalis</i> sp., Boraginaceae, Buxaceae, Mimosaceae
E38	Poaceae (13 %), <i>Diplotaxis eruroides</i> (L) DC. (9 %), <i>Sophora</i> sp. (8%), <i>Muscari</i> sp., Rosaceae (5 %), Apiaceae, Asteraceae and Boraginaceae (4 %)	Annonaceae, Fabaceae, <i>Euphorbia</i> sp., Betulaceae, Lamiaceae, Myrtaceae
E39	<i>Diplotaxis eruroides</i> (L) DC. (6 %), Anacardiaceae (5 %), <i>Euphorbia</i> sp. (4 %)	Oleaceae, Smilacaceae, Asteraceae, Fabaceae, Apiaceae, Rosaceae
E34	<i>Ziziphus lotus</i> (L) Lam. (6 %), <i>Diplotaxis eruroides</i> (L) DC. (5 %), Poaceae (4%), Ericaceae (3 %)	Asteraceae, Fabaceae, Apiaceae, Cucurbitaceae, Polygonaceae, Cupressaceae, Liliaceae, Salicaceae, Verbinaceae
E40	Fabaceae, Myrtaceae (13 %), Apiaceae, Boraginaceae (9 %), Anacardiaceae (7 %), Lamiaceae (5 %), <i>Diplotaxis eruroides</i> (L) DC., Rosaceae (4 %), <i>Euphorbia</i> sp., Ranunculaceae (3 %)	Asteraceae, Polygonaceae, Ericaceae, Fagaceae, Mimosaceae
E42	<i>Euphorbia</i> sp., Rosaceae (8 %), Brassicaceae (5 %), Chenopodiaceae, Poaceae (4 %)	<i>Borago officinalis</i> , Apiaceae, Cupressaceae, <i>Oxalis</i> sp., Polygonaceae
E51	Salicaceae (10 %), <i>Euphorbia</i> sp., <i>Peganum harmala</i> (L). (6 %), <i>Senecio vulgaris</i> L.(5%), <i>Muscari</i> sp. (3 %)	Lamiaceae, Myrtaceae, Oleaceae, Rafflesciaceae, <i>Citrus</i> sp., <i>Ononis</i> sp., Polygonaceae, Verbenaceae
E71	<i>Eucalyptus</i> sp. (12 %), Asteraceae, Fabaceae (5 %), <i>Daucus carota</i> L. (3 %)	Rosaceae, Mimosaceae

Table 4: Results of physicochemical parameters and sugar profile

Physicochemical Parameters	Unifloral honey (n = 15)	Polyfloral honey (n = 5)	Min-Max
EC (mS cm ⁻¹)	0.30 ± 0.16	0.26 ± 0.14	0.12 - 0.68
pH	4.09 ± 0.44	3.81 ± 0.31	3.41 - 5.07
Free acidity (meq kg ⁻¹)	22.88 ± 7.16	25.03 ± 8.45	10.25 - 34.40
Moisture (%)	16.25 ± 1.36	15.42 ± 1.41	13.60 - 18.25
HMF (mg kg ⁻¹)	27.67 ± 46.42	7.37 ± 4.5	0.00 - 184.70
Color (PFund)	86.78 ± 67.34	95.07 ± 86.1	5.68 - 210.13
	(Amber)	(Amber)	(Water white - dark amber)
Fructose (%)	34.21 ± 2.49	33.28 ± 1.34	28.90 - 39.51
Glucose (%)	32.24 ± 2.99	33.40 ± 2.21	27.63 - 37.49
F+G (%)	66.45 ± 3.87	66.69 ± 4.58	61.38 - 76.06
Sucrose (%)	3.15 ± 2.26	2.44 ± 2.2	0.68 - 6.39

al. (2009) reported in their study on rats that HMF and SMF could be initiators of colon cancer.

Hence the interest in drawing beekeepers' attention to this parameter and the importance of respecting the levels proposed for those who want to distinguish their honey by a qualitative approach.

Color values are presented in Pfund values (mm) and classified in a scale going from water white to dark amber. The unifloral and polyfloral honey groups showed mean values of 86.78 and 95.07 mm, corresponding both of them to amber. The extreme values of color ranged from 5.68–210.13 mm corresponding to water white to dark amber. The variation in color for the different honey samples is due to several factors including among others: the variation in the sources of nectar (Kuś et al., 2014), the electrical conductivity, the richness of the honey in minerals, and the storage conditions (González-Miret et al., 2005; Naab et al., 2008), as well as the composition of honey in phenolic compounds and their antioxidant power (Bertoncelj et al., 2007).

The study of the carbohydrate profile provides several information, such as possible fraud attempts through adulteration, which results in an increase in HMF, the classification of monofloral honeys (Persano Oddo & Piro, 2004), or the tendency of honey to crystallize; in fact, the ratios F/G (Fructose/Glucose) and G/E (Glucose / Water) are considered criteria for predicting the tendency of honeys to crystallize. High F/G levels suggest that honeys are more likely to remain liquid. For the G/E ratio, results equal to or less than 1.7 indicate liquid

honey, while values equal to or greater than 2.1 predict rapid granulation (Doner, 1977).

The extreme values of fructose were 28.90–39.51 % with a high value registered for unifloral honey group (34.21 %), whereas glucose extreme values obtained ranged from 27.63–37.49 % with a high value registered for polyfloral honey group (33.4 %). In general, our data for glucose corroborate with those quoted by Makhloufi et al. (2010), Haderbache et al. (2013); Ouchemoukh et al. (2010), Mekious et al. (2015) and Zerrouk et al. (2017) who mentioned values ranging from 25.47–33.89 %. Contrariwise, the mean values of fructose are lower compared to those of the aforementioned authors who provided values ranging from 35.50–42.10 %, regardless of the type of honey studied. Nevertheless, fructose and glucose values obtained in our study corroborate with those of Gonnet (1971) who specifies that the sugar content of honey varies from 32–46 % for fructose and from 26–41 % for glucose. Molan (1996) reported that the nectar composition of plants influences the proportions of these two major sugars. Also, Mateo et al. (1998) reported that the sugar profile of honey depends greatly on the types of flora foraged by the bees, by regional and climatic conditions. In general and according to White et al. (1979), fructose predominates over glucose. This finding is confirmed in our study.

The total sugar content ranged from 61.38 %–76.06 %, with similar mean values to both groups (66.45 and 66.69 %). These values are in agreement with the standards of Codex Alimentarius (2001); Council Directive (2002) requiring a rate of more than 60 % for nectar honey.

Sucrose content oscillated between 0.68 %–6.39 %. The highest value was recorded for unifloral honey group with 3.15 %. (Anklam, 1998) explained that honeys of the same floral source can vary due to seasonal climatic variations or to a different geographical origin. The Codex alimentarius standard specifies 5 % of sucrose for all varieties of honey, with the exception of 10 % for *Banksia*, *Citrus*, *Hedysarum*, *Medicago*, and honeys, and of 15% for *Lavandula* honey. However, the high sucrose content (6.39 %) found in our study corresponds to *Ziziphus* honey. This latter is not among the honeys mentioned as exempted. Indeed, its high value could be due to different reasons such as overfeeding bees with sucrose syrup, adulteration, or harvesting honey early, where the sucrose has not been fully transformed into glucose and fructose (Anklam, 1998; Azeredo et al., 2003; Guler et al., 2007). However, all samples showed values corroborating to those of (Benaziza-Bouchema & Schweitzer, 2010) (between 0–7.6 %) and also remain below 10 %, a limit mentioned by Bocquet (1997) for sucrose.

3.3 TRACE AND TOXIC ELEMENTS ANALYSIS

All the minerals and heavy metals identified in honey samples are listed in Table 5. The mean mineral concentrations in the different honey groups were expressed by mg kg^{-1} for Cr, Mn, Fe, Ni, Cu, and Zn and by $\mu\text{g kg}^{-1}$ for As, Pb, and Cd, the concentrations of the two later were compared to the maximum allowable contaminant levels established by the Commission Regulation (EU) (2015) and proposed by Bogdanov et al. (2003).

Considering the average value of all samples, the most abundant trace elements were Fe followed by Zn, Ni, Cu, Cr, and Mn. To the best of our knowledge, few studies were conducted on minerals and toxic elements in honeys in Algeria. (Haderbache et al. in 2013, Yaiche Achour and Khali (2014) and Zerrouk et al. (2017).

Yaiche Achour et al. (2014) and Zerrouk et al. (2017) reported higher values than ours for Fe (6.37 and 6.3 mg kg^{-1}) respectively for jujube honey. Whereas, (Haderbache et al., 2013) reported values of (0.923 and 0.969 mg kg^{-1}) for jujube and multifloral honeys. These results are quite lower than our results.

Regarding Zn, Yaiche Achour & Khali (2014) and Zerrouk et al. (2017) reported values of 11.04 mg kg^{-1} for all types of honey and 1.8 mg kg^{-1} for jujube honey respectively. Although, unifloral honey group; in our study; showed a mean value quite similar to that of jujube honey.

The values of Ni reported by Haderbache et al. (2013) and Yaiche Achour & Khali (2014) are (0.0234 , $0.0307 \text{ mg kg}^{-1}$) and (0.32 mg kg^{-1}) for jujube and multifloral honeys and for all types of honeys respectively. The Ni concentration value obtained in our study for unifloral honey group was quite similar to that reported by Yaiche Achour & Khali (2014). Whereas, polyfloral honey group exhibited a higher value than the previous studies.

Cu was studied only by Yaiche Achour & Khali (2014), their obtained values were in the range of 2.72 -

3.22 mg kg^{-1} , these values are higher than our results for both honey groups.

The obtained concentration values of Cr were higher than the reported one by Yaiche Achour and Khali (2014) 0.023 mg kg^{-1} for all types of honey. Whereas, for the Mn, the observed values were lower than the values of Haderbache et al. (2013) and Yaiche Achour & Khali (2014) (0.077 , 0.069 mg kg^{-1}) for jujube and multifloral honeys and (3.06 mg kg^{-1}) for all types of honey respectively.

Regarding the toxic elements Haderbache et al. (2013) reported for Pb values lower than ours. While, Yaiche Achour and Khali (2014) reported higher values. (9.2 , 16.3 mg kg^{-1}) for jujube and multifloral honeys and (0.22 mg kg^{-1}) for all types of honey respectively.

The Cd concentration values were much lower than those observed for Haderbache et al. (2013) and Yaiche Achour and Khali (2014) with (10.7 , 13.9 mg kg^{-1}) for jujube and multifloral honeys and (0.018 - 0.019 mg kg^{-1}) for all types of honey respectively.

As concentration mean values; obtained in our study; were different from the values reported by Yaiche Achour and Khali (2014), (0.020 - 0.024 mg kg^{-1}) as a mean range for all type of honeys.

Considering previous investigations on honeys conducted in different country in Europe and China, the average value of Fe was 1.89 mg kg^{-1} (ranging from 0.25 - 21.54 mg kg^{-1}). Quite similar values were observed in Italy with 1.265 and 1.75 mg kg^{-1} for polyfloral and sweet chestnut honey (Buldini et al., 2001). Bilandžić et al. (2014); Hernández et al. (2005) reported higher values of Fe (4.85 and 3.61 mg kg^{-1}) comparatively to our results for honey produced in Spain and Croatia respectively.

Zn was the second most abundant trace element, with an average of 0.90 mg kg^{-1} (ranging from 0 - 4.46 mg kg^{-1}). The mean value of Zn was lower than those found in previous investigations in Croatia (1.69 and 1.17 mg kg^{-1}) (Bilandžić et al., 2014; Lachman et al., 2007), Italy (2.64 and 3.205 mg kg^{-1}) (Buldini et al., 2001), Spain (1.57 and 1.441 - 4.496 mg kg^{-1}) (Fernandez-Torres et al., 2005; Hernández et al., 2005) respectively and China ($1329.5 \mu\text{g kg}^{-1}$) (Ru et al., 2013).

The Ni mean concentration for all honeys was 0.54 mg kg^{-1} (ranging from 0 - 3.57 mg kg^{-1}). The observed value was higher than the reported values in Italy (0.10 - 03.22 mg kg^{-1}) (Squadrone et al., 2020).

The Cu mean concentration for all honeys was 0.17 mg kg^{-1} (ranging from 0 - 2.92 mg kg^{-1}). This mean level was much lower than those observed in Italy ($890 \mu\text{g kg}^{-1}$ and 0.30 - 0.95 mg kg^{-1}) (Buldini et al., 2001; Squadrone et al., 2020) respectively, Spain (0.37 , < 0.531 - 0.693 mg kg^{-1}) (Fernandez-Torres et al., 2005; Hernández et al., 2005) respectively, Croatia (0.42 and 14.4 mg kg^{-1}) (Bilandžić et al., 2014; Lachman et al., 2007) respectively. The mean

Table 5: Results of minerals and heavy metals analyses

	Unifloral honey (n = 15)	Polyfloral honey (n = 5)	Min-Max
Cr (mg kg^{-1})	0.07 ± 0.06	0.13 ± 0.11	0.00 - 0.33
Mn (mg kg^{-1})	0.04 ± 0.06	0.02 ± 0.01	0.01 - 0.24
Fe (mg kg^{-1})	2.15 ± 5.38	1.09 ± 0.65	0.25 - 21.54
Ni (mg kg^{-1})	0.31 ± 0.47	1.22 ± 1.39	0.00 - 3.57
Cu (mg kg^{-1})	0.22 ± 0.75	0.03 ± 0.01	0.00 - 2.92
Zn (mg kg^{-1})	1.14 ± 1.38	0.19 ± 0.16	0.00 - 4.46
As ($\mu\text{g kg}^{-1}$)	16.48 ± 37.05	191.16 ± 303.45	0.00 - 718.09
Cd ($\mu\text{g kg}^{-1}$)	0.99 ± 2.82	3.48 ± 3.64	0.00 - 11.00
Pb ($\mu\text{g kg}^{-1}$)	75.11 ± 98.46	42.69 ± 22.33	3.17 - 357.19

Cu content obtained was within the range of that found in honey from Croatia (0.14-1.39 mg kg⁻¹) (Bilandžić et al., 2017).

The Cr mean concentration for all honeys was 0.09 mg kg⁻¹ (ranging from 0-0.33 mg kg⁻¹). This mean level fell within the range reported from Italy (0.068-0.093 mg kg⁻¹) (Squadrone et al., 2020) but was higher than the range reported for honey from Croatia (4.97-27.6 µg kg⁻¹) (Bilandžić et al., 2017).

The Mn mean concentration for all honeys was 0.04 mg kg⁻¹ (ranging from 0.01-0.24 mg kg⁻¹). This mean level was much lower than the ranges reported from Croatia and Italy (0.19-1.98 and 0.61-3.2 mg kg⁻¹) (Bilandžić et al., 2017; Squadrone et al., 2020) respectively.

In general, mineral elements come from the soil and end up in honey through plant nectar (Solayman et al., 2016). The variability in mineral content can be attributed to environmental, botanical, and geographical factors, or even beekeeping practices (Bogdanov, 2006; Sixto et al., 2019). The mean values of the total Cr, Mn, Fe, Ni, Cu and Zn concentrations were in the range of 2.7 mg kg⁻¹ (polyfloral)-3.93 mg kg⁻¹ (unifloral). These results were also in the range reported by (Squadrone et al., 2020) who mentioned values of 3.4 mg kg⁻¹ (acacia)-7.0 mg kg⁻¹ (multifloral). Such values indicate the contribution of these essential elements in the nutritional aspects of honey.

It is well known that lead is the most widespread heavy metal in the environment with potential toxicity. It has the potential to induce gradual poisoning and health complications like exhaustion, insomnia and body mass loss. Based on our study, the lead concentrations were high compared to As and Cd. The Pb mean concentration for all honey was 77.54 µg kg⁻¹ (ranging from 3.17-357 µg kg⁻¹). The obtained Pb value in our study was lower than those found in honey from Italy (620 µg kg⁻¹ for polyfloral honey) (Buldini et al., 2001), Croatia (530 µg kg⁻¹). While Bilandžić et al. (2017); Ru et al. (2013); Squadrone et al. (2020) reported values lower than ours with 33.98, 5.03-66.3, and 12.71 µg kg⁻¹ for honeys from China, Croatia and Italy respectively.

The Cd mean concentration for all honeys was 1.02 µg kg⁻¹ (ranging from 0.0-11 µg kg⁻¹). This obtained Cd value was almost similar to those obtained from China and Croatia (1.34 and 1.84 µg kg⁻¹) (Bilandžić et al., 2014; Ru et al., 2013) respectively. The Cd content found in this study was lower than those found in honey from Italy, (305 µg kg⁻¹) (Buldini et al., 2001).

The As mean concentration for all honeys was 60.15 µg kg⁻¹ (ranging from 0.0-718.09 µg kg⁻¹). The obtained As concentration was much higher than those reported

for honeys from Italy (7.7-17 µg kg⁻¹) (Squadrone et al., 2020), China (13.44 µg kg⁻¹) (Ru et al., 2013) and Croatia (0.62-6.95 µg kg⁻¹) (Bilandžić et al., 2017) and lower than those reported from Croatia (140.7 µg kg⁻¹) (Bilandžić et al., 2014).

Pb and Cd are considered the most toxic heavy metals. The Codex Alimentarius (2001) stipulates that "honey should only contain heavy metals at levels that do not pose a risk to human health". The European Union proposed limits of 1.0 and 0.1 mg kg⁻¹ for Pb and Cd respectively (Bogdanov et al., 2003).

High heavy metal values have several causes. Lead, as the most widespread metal, is mainly released into the air and then found in many products after being mixed with soil and thus penetrates plants, but in general, Pb is not transported by plants.

Cadmium and due to its use in a wide different industrial processes; notably the metallurgical industry and incinerators (Yao et al., 2019); is released into the environment, and through its absorption by plants from contaminated soil or water reaches the food chain. That said, several parameters influence the concentration of Cd in different locations, and consequently its concentration in honey. However, only a small proportion of Cd can reach honey by air, mainly in the proximity of incinerators.

Arsenic can also come from non-ferrous metallurgy and factories, but it can also be present in the environment through the use of agrochemicals such as arsenic-based fertilizers and pesticides. As a result, arsenic is found in water, soil, and air, and as it is absorbed by all plants, it finds its way into the food chain, including honey. Hence the importance of limiting the use of arsenic-based pesticides and introducing quality control measures for honey.

Poor beekeeping practices applied in the extraction and storage of honey can also cause a significant source of contamination in toxic elements, the acidic characteristic of honey also helps to release certain metals such as Pb from metal containers.

These results indicate that Algerian honey is not far from European and Chinese honeys in terms of quality and food safety. Even with regard to the European regulations, the levels of Pb and Cd are below the maximum limit, which suggests studying the possibility of establishing a national standard specific to Algerian honeys and also encouraging beekeepers to export their honeys without the risk of rejection due to non-compliance with heavy metals. But attention should be drawn to the specific limit of 0.1 mg kg⁻¹, beekeepers may think of introducing a variety of honey for children and persons with particular dietary needs.

3.4 STATISTICAL ANALYSIS

Several physicochemical parameters exhibited notable correlations. Specifically, Pb demonstrated strong positive associations with Cd ($r = 0.708, p < 0.01$), Cu ($r = 0.62, p < 0.05$), and Fructose ($r = 0.52, p < 0.05$). Similarly, Fe and Cr displayed a significant positive correlation ($r = 0.7, p < 0.001$). Cd and Ni also, showed a significant positive correlation ($r = 0.6, p < 0.01$). On the other hand, negative correlations were identified between Cr and pH ($r = -0.52, p < 0.05$), as well as between Cd and Zn ($r = -0.52, p < 0.05$).

Figure 3 shows a representation of the samples on the first two components which represent 36.4 % of the variability, with 19.8 % explained by the first axis and 16.6 % explained by the second axis.

The multiple factor analysis (Axes 1 and 2) distinct patterns within the two groups of honey samples. The polyfloral honey samples were characterized by a high concentration of Cr, As, and Ni. On the other hand, the

unifloral group showed a high concentration of pH, Zn, and HMF.

The multiple factor analysis showed that the sample (E39) of *P. harmala* L. was characterized by a particularly high concentration of Cu and Cd. A study on the germination characteristics of *P. harmala* seeds exposed to heavy metals and their impact on rehabilitating polluted arid lands showed that *P. harmala* had a high germination ability even in highly contaminated soils (Schweitzer, 2001). Another study also showed the effectiveness of *P. harmala* seeds to remove Pb^{2+} , Zn^{2+} and Cd^{2+} ions from aqueous solutions (EIC, 2015). These findings suggest that *P. harmala* is well-suited to growing in polluted environments and may be an effective adsorbent for removing heavy metals (Schweitzer, 2001) and thus may explain the high levels of Cu and Cd in *P. harmala* honey.

The multiple-factor analysis also showed that eucalyptus samples (E32, E44, E78) had a high moisture content and high levels of free acidity. Eucalyptus honey is known for its high electrical conductivity. This latter re-

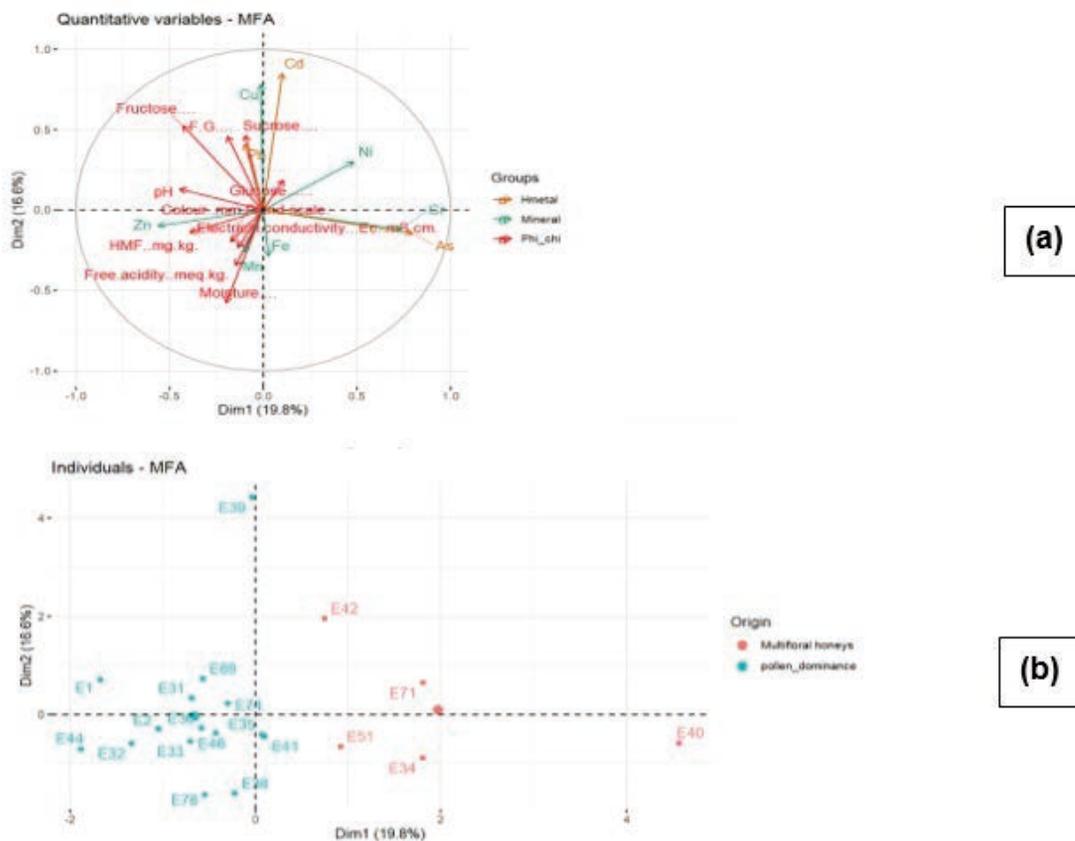


Figure 3: Multiple factor analysis (a) Loading biplot of the variables included in the analysis, (b) Score biplot of the samples regarding component 1 and 2

flects the richness of the honey in mineral elements and organic acids. Hence, a high content of organic acid and salts increases the free acidity present in honey (Ghorab et al., 2021). Moreover, Bogdanov et al. (2004) reported that honey's acidity is the result of the transformation of glucose into gluconic acid, and this transformation is more favored by high water content.

4 CONCLUSIONS

This study was carried out to analyze the pollen characteristics, physicochemical properties, and mineral and heavy metal composition of 20 types of honey from 13 different locations throughout Algeria. The study revealed the presence of several honey types, with the predominant pollen of *Citrus* sp., *Eucalyptus* sp., *Ziziphus lotus* (L.) Lam., *Sinapis arvensis* L., *Dorycnium* sp., *Echium* sp., *Bupleurum* sp., *Lotus* sp. and *Peganum harmala* L., and polyfloral honey. All types of honey meet the quality standards required by the Codex Alimentarius, the European Directive. Except HMF, for which we have noted non-conformity for three samples. Thus, it is noteworthy to mention that improving beekeepers' knowledge of honey harvesting techniques, processing, and storage is essential to produce high-quality honey that meets both national and international market standards.

The analyzed honey also complied with the standards of the European Directive for heavy metals. The concentrations of Pb and Cd in the honey samples were found to be below the maximum residue levels.

The obtained results are highly relevant to programs aimed at enhancing the value of honey and protecting it with a sign of quality or a geographical indication. This approach provides a real opportunity to maintain and improve the quality of local honey. The identified characteristics contribute to the creation of a reference database in order to establish regulations in Algeria regarding the physicochemical and heavy metals composition of honey.

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