

Physicochemical Properties, Minerals, Trace Elements, and Heavy Metals in Honey of Different Origins: A Comprehensive Review

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Abstract: Honey is a popular natural food product with a very complex composition mainly consisting of both organic and inorganic constituents. The composition of honey is strongly influenced by both natural and anthropogenic factors, which vary based on its botanical and geographical origins. Although minerals and heavy metals are minor constituents of honey, they play vital role in determining its quality. There are several different analytical methods used to determine the chemical elements in honey. These methods are typically based on spectroscopy or spectrometry techniques (including atomic absorption spectrometry, atomic emission spectrometry, inductively coupled plasma mass spectrometry, and inductively coupled plasma optical emission spectrometry). This review compiles available scientific information on minerals and heavy metals in honey reported from all over the world. To date, 54 chemical elements in various types of honey have been identified and can be divided into 3 groups: major or macroelements (Na, K, Ca, Mg, P, S, Cl), minor or trace elements (Al, Cu, Pb, Zn, Mn, Cd, Tl, Co, Ni, Rb, Ba, Be, Bi, U, V, Fe, Pt, Pd, Te, Hf, Mo, Sn, Sb, La, I, Sm, Tb, Dy, Sd, Th, Pr, Nd, Tm, Yb, Lu, Gd, Ho, Er, Ce, Cr, As, B, Br, Cd, Hg, Se, Sr), and heavy metals (trace elements that have a specific gravity at least 5 times higher than that of water and inorganic sources). Chemical elements in honey samples throughout the world vary in terms of concentrations and are also influenced by environmental pollution.

Keywords: composition, heavy metals, honey, minerals, trace elements

Introduction

Honey is a sweet natural product produced by honey bees (*Apis mellifera*) that collect the nectar from flowers and convert it into a delicious food product that is known to be a more health beneficial nutritional option than plain sugar (Vanhanen and others 2011). It is mainly composed of fructose and glucose (65%) as well as water (18%), with minimal protein and lipid contents (Khalil and others 2001; Silva and others 2009). Honey also contains minerals and heavy metals, which play important roles in determining honey qualities. The mineral content varies, ranging from 0.04% in pale honeys to 0.20% in darker honeys (Bogdanov and others 2007). The major minerals are mainly derived from the soil and nectar-producing plants, but they may also come from anthropogenic sources, such as environmental pollution. It has been reported that micro- or trace minerals originating from organic or plant sources are important for maintaining human health, while those which

originate from inorganic or metallic sources, such as heavy metals, can be toxic (Hernández and others 2005; Pohl 2009).

Honey's composition and properties depend on the botanical origin of the nectar or the bee secretions. The geographic and plant origin of honey is usually determined by using several techniques. Some of these techniques include the melissopalynological method (Anklam 1998), gas chromatography-mass spectrometry (Bianchi and others 2005), electronic nose technology (Lammer-tyn and others 2004), or quantification of selected chemical parameters (Anklam 1998; Conte and others 1998; Anupama and others 2003). Due to technological advancements, the determination of inorganic compounds in honey has become simpler, faster, and more economical.

Honey has various nutritional, medicinal, and prophylactic (preventative) properties contributed by its various chemical constituents. Nevertheless, in order to yield medicinal effects, honey should be free of any contaminants. Due to this fact and honey utility as a natural, effective, and pleasant sweetener, there is an increasing interest in the study of honey (Mckee 2003). Honeybees are estimated to forage on plants growing over a relatively large area (more than 7 km²) and, when going from flower to flower, they are also in contact with air, water, soil, branches, and leaves. Therefore, honey is the result of a bio-accumulative process that is useful for collecting information about the environment and may be considered as a bio-indicator of environmental pollution (Bratu and Georgescu 2005). Honey has also been used as an indicator

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for a variety of environmental contaminants, including heavy metals, low-level radioactivity, and pesticides (Nalda and others 2005; Bogdanov and others 2007).

Although different types of honey have been used since ancient times for diverse purposes, there are a considerable number of scientific data on the types and concentrations of minerals and heavy metals in honey, however, bitterly scattered. This review is aimed at reporting the information on the physicochemical properties of honey, particularly the mineral and heavy metal contents, from the past 15 y (2000 to 2014) and from countries all over the world, and at evaluating their importance and their correlations with various parameters. Broad analysis of these data can be used to promote more widespread use of honey with special properties. To our knowledge, this is the first detailed report on the minerals, trace elements, and heavy metals in honey and their medicinal effects.

Methods of Review

For data collection, Google Scholar, PubMed, and ScienceDirect were independently searched and the full text was accessed via Univ. Sains Malaysia library. Keywords including honey, physicochemical properties, minerals, trace elements, heavy metals, and the names of several different countries were applied in the search strategy.

Honey: its composition and physicochemical properties

According to the definition set by European Union Council Directive 2001/110/EC, "Honey is the natural sweet substance produced by the *Apis mellifera* bees from the nectar of plants or from the secretions of living parts of plants or excretions of plant-sucking insects on the living parts of plants which the bees collect, transform by combining with specific substances of their own, deposit, dehydrate, store and leave in honeycombs to ripen and mature." Honey can be classified according to its origin (such as nectar or honeydew honeys), mode of production and/or presentation (comb honey, chunk honey, drained honey, extracted honey, pressed honey, and filtered honey), and as baker's honey (suitable for industrial uses or as an ingredient in other foodstuffs) (European Union Council Directive 2001/110/EC of 20 December 2001 relating to honey 2002). Honey is the only food sweetener that can be used industrially without prior processing (Bogdanov and others 2008), and its production is increasing globally (Ma 2009).

Honey is a concentrated aqueous solution composed of a mixture of glucose and fructose, but it also contains at least 22 other complex carbohydrates, various amino and organic acids, proteins, antibiotic-rich inibine, enzymes, phenol antioxidants, aroma compounds, vitamins, minerals, pigments, waxes, and pollen grains (PGs) (White and Doner 1980). Honey is viscous and acidic in nature, with a pH ranging between 3.2 and 4.5. Because sugars are its main constituents, the physical features and behavior of honey are attributed to its sugars. Tests conducted on sugar will indicate its sweetness due to the high sugar content, with the most plentiful sugar being fructose. Minor constituents such as flavor compounds, minerals, acids, pigments, and phenols are largely responsible for defining each specific type of honey (White and Doner 1980; Ajibola and others 2007; Bogdanov and others 2008).

The U.S. Food and Drug Act defines honey as "the nectar and saccharine exudation of plants, gathered, modified and stored in the comb by the honey bees (*Apis mellifera* and *A. dorsata*) which contain no more than 25% water, 0.25% ash and 8% sucrose." This definition was based on a survey published in 1908.

In a more recent study, the composition of honey based on 490 samples of different floral origins indicated that honey composed of 17.2% moisture, 38.19% fructose, 31.28% glucose, 1.31% sucrose, and 0.17% ash (White and others 1980). Therefore, an updated physicochemical composition is given in Table 1, compiled from approximately 1000 honey samples from all over the world. The data were found to vary to some extent from those stated by the Codex standard for honey (Commission 1981).

Mineral content

From a nutritional standpoint, minerals are naturally occurring inorganic solid substances in the biosphere (including all foods) found following the degradation of plant and animal tissues (Belitz and others 2009). They are formed by geological processes (Nickel 1995) and are essential for the regulation of metabolic pathways in the living body (Gopalan and others 1989). Minerals are divided into 3 groups on the basis of body requirements: (1) major elements, (2) trace elements, and (3) ultra-trace elements. Major elements [sodium (Na), potassium (K), calcium (Ca), magnesium (Mg), phosphorus (P), sulfur (S), chlorine (Cl)] should be present at >50 mg/d, whereas trace elements [iron (Fe), iodine (I), fluorine (F), zinc (Zn), selenium (Se), copper (Cu), manganese (Mn), chromium (Cr), molybdenum (Mo), cobalt (Co), nickel (Ni)] are required in concentrations of <50 mg/d in human beings (Belitz and others 2009). The calculated dietary requirements for ultra-trace elements [aluminum (Al), arsenic (As), barium (Ba), bismuth (Bi), boron (B), bromine (Br), cadmium (Cd), cesium (Cs), germanium (Ge), mercury (Hg), lithium (Li), lead (Pb), rubidium (Rb), antimony (Sb), silicon (Si), samarium (Sm), tin (Sn), strontium (Sr), thallium (Tl), titanium (Ti), tungsten (W)] are usually less than 1 µg/g and often present at less than 50 ng/g in the dry matter of the diet (Nielsen 1984; Belitz and others 2009). Micro- or trace minerals are useful for good health, especially if they originate from an organic or plant source. On the other hand, if they originate from an inorganic or metallic source, they will have at least 5 times the specific gravity of water and can become toxic. In this case, they are then known as heavy metals (Ajibola and others 2012). The scientific literature indicates that Pb, Cd, Hg, Cr, Cu, Mn, Ni, Zn, and Ag are the most important heavy metals (Nielsen 1984). Heavy metals are toxic or poisonous at low concentrations because of their tendency to accumulate in living organisms (Zugravu and others 2009).

Honey also contains minerals and elements, with their contents differing in the types and amounts depending on the botanical and geographical origins of the honey (Vincēviča-Gaile 2010). We have listed 54 minerals reported to be present in honey by searching for articles on honey published in the last 15 y from all over the world, which include both major (Na, K, Ca, Mg, P, S, Cl) and minor elements (Al, Cu, Pb, Zn, Mn, Cd, Tl, Co, Ni, Rb, Ba, Be, Bi, U, V, Fe, Pt, Pd, Te, Hf, Mo, Sn, Sb, La, I, Sm, Tb, Dy, Sd, Th, Pr, Nd, Tm, Yb, Lu, Gd, Ho, Er, Ce, Cr, As, B, Br, Cd, Hg, Se, Sr (González Paramás and others 2000; Birge and Price 2001; Devillers and others 2002; Nanda and others 2003; Tuzen and Soylak 2005; Mendes and others 2006; Bogdanov and others 2007; Tuzen and others 2007; Baroni and others 2009; Taha and others 2010; Vincēviča-Gaile 2010; Vit and others 2010; Vanhanen and others 2011; Batista and others 2012; Ruschioni and others 2013).

A previous study has shown that the total mineral content in honey is relatively low and generally accounts for 0.1% to 0.2% of the composition of nectar honeys but can exceed 1% in other types of honey such as honeydew honeys (Hernández and others 2005).

Table 1—Composition and physicochemical properties of honey.

Constituents	Mean	SD ^a	Range	Values compiled from these sources
Moisture (%)	17.90	3.16	13.21–26.50	D'arcy and others (1997); Khalil and others (2001); Terrab and others (2003b); Nalda and others (2005); de Oliveira Alves and others (2005); Küçük and others (2007); Marchini and others (2007); Cantarelli and others (2008); Qamer and others (2008); Baroni and others (2009); Omafuvbe and Akanbi (2009); Al and others (2009); Chefrour and others (2009); Marghitas and others (2010); Kahraman and others (2010); Saxena and others (2010); Alvarez-Suarez and others (2010b); Islam and others (2012); Mahmoudi and others (2012); Buba and others (2013); Belay and others (2013); Mondragón-Cortez and others (2013); Moniruzzaman and others (2013); Akhtar and others (2014)
pH	3.96	0.33	3.40–4.71	
Fructose (%)	39.44	2.11	37.07–2.65	
Sucrose (%)	3.19	3.81	0.36–16.57	
Glucose (%)	28.15	5.74	18.20–32.10	
Reducing sugar (%)	68.31	6.51	54.50–77.10	
Minerals (%)	0.36	0.18	0.11–0.72	
Total protein (%)	1.13	1.22	0.22–2.93	
Lipid content (mg/g)	215.20	105.78	140.40–290.00	
HMF ^b (mg/kg)	15.49	12.43	0.86–39.08	
Vit-C (mg/g)	13.19	7.93	13.19–7.93	
Lactone (meq/kg)	8.57	5.61	1.56–14.87	
Proline content (mg/kg)	873.29	886.57	114.35–2131.47	
Diastase activity (DN)	14.27	9.72	0.40–22.08	
Electrical conductivity (ms/cm)	0.64	0.40	0.15–1.64	
ABS450 ^c (mAU; 50 w/v)	834.43	327.75	378.83–1143.71	
Free acidity (meq/kg)	35.32	28.75	6.45–124.20	
Total soluble solids (Brix)	127.70	105.79	78.77–316.92	
Refractive index	1.49	0.01	1.48–1.49	
Water-insoluble fraction (% w/w)	15.95	22.46	0.07–31.84	

^aStandard deviation.^b5-(Hydroxymethyl)furfural.^cColor intensity determined by taking absorbance at 450 nm.

Table 2—Major elements, trace elements, and heavy metals in honey.

Element	Units	Mean	SD	Range	Values compiled from these sources
Na ^a	mg/kg	96.48	80.58	3.23–236.80	Caroli and others (1999); González Paramás and others (2000); Birge and Price (2001); Conti and Botrè (2001); Devillers and others (2002); Matei and others (2004); Terrab and others (2004a); Tuzen and Soylyak (2005); Dag and others (2006); Dobrinas and others (2006); Fredes and Montenegro (2006); Conti and others (2007); Roman and others (2007); Osman and others (2007); Tuzen and others (2007); Yarsan and others (2007); Akram (2008); Farooq Khan and Maqbool (2008); Frías and others (2008); Omode and Ademukola (2008); Baroni and others (2009); Zugravu and others (2009); Taha and others (2010); Agbagwa and others (2011); Mbiri and others (2011); Vanhanen and others (2011); Atanassova and others (2012); Alvarez-Suarez and others (2012); de Alda-Garçilope and others (2012); Perna and others (2012); Berinde and Michnea (2013); Mondragón-Cortez and others (2013); Silici and others (2013); Corredera and others (2014); de Andrade and others (2014); Eissa and others (2014); Moniruzzaman and others (2014)
K ^a	mg/kg	742.43	453.88	39.66–1349.34	
Ca ^a	mg/kg	84.36	68.34	4.85–218.00	
Mg ^a	mg/kg	74.31	163.05	2.18–563.72	
P ^a	mg/kg	84.10	48.36	28.80–118.50	
S ^a	mg/kg	35.27	19.86	15.39–72.20	
Cl ^a	mg/kg	302.63	83.81	181.30–427.2	
Fe ^b	mg/kg	30.34	64.99	0.41–224.00	
Cu ^{b,c}	mg/kg	1.99	4.31	0.05–17.30	
Zn ^{b,c}	mg/kg	9.33	19.55	0.23–73.60	
As ^b	mg/kg	0.05	0.04	ND–0.10	
Ni ^{b,c}	mg/kg	1.24	2.62	ND–9.00	
Mn ^{b,c}	mg/kg	1.42	1.47	0.00–4.35	
Se ^b	mg/kg	0.01	0.00	0.01–0.01	
Al ^b	mg/kg	5.12	3.61	1.39–11.36	
Si ^b	mg/kg	23.52	16.13	8.70–40.70	
B ^b	mg/kg	5.36	1.33	4.42–6.30	
Mo ^b	mg/kg	0.23	0.30	0.01–0.44	
Sr ^b	mg/kg	1.63	1.03	0.90–2.36	
V ^b	mg/kg	0.03	0.03	0.01–0.05	
Ti ^b	mg/kg	43.40	17.30	25.00–71.00	
Cd ^{b,c}	μg/kg	89.69	129.75	0.17–373.00	
Pb ^{b,c}	μg/kg	424.57	839.65	0.63–3232.00	
Co ^b	μg/kg	171.78	291.72	0.01–800.00	
Hg ^{b,c}	μg/kg	5.09	6.82	0.27–9.91	
Cr ^{b,c}	μg/kg	152.84	139.74	ND–370.00	
Ag ^{b,c}	μg/kg	299.61	419.17	3.21–596.00	
Be ^b	μg/kg	9.93	0.05	9.83–10.00	

^aMajor elements.^bTrace elements.^{b,c}Trace elements/Heavy metals.

ND, not detected; SD, standard deviation.

Our study indicated that it ranged between 0.11% and 0.72% (Table 1), with K being present in higher amounts (742.43 mg/kg; Table 2). Some mineral types cannot be detected in some types of honey collected from certain areas but are nevertheless available in other regions. For example, the trace elements Ni and Cr cannot be detected in Egypt (Taha and others 2010) but are reported to be present in honey samples from Pakistan (Farooq Khan and Maqbool 2008), Turkey (Tuzen and others 2007; Silici and others 2008), New Zealand (Vanhanen and others 2011), Romania

(Matei and others 2004), and Spain (González Paramás and others 2000; Terrab and others 2004a), which may be attributed to differences in soil composition, floral type, and floral density as well as nectar and pollen (Abu-Tarboush and others 1993; Fernández-Torres and others 2005; Yarsan and others 2007; Santos and others 2008). Therefore, honey composition can actually be used to identify its geographical and botanical sources (Anklam 1998). Other trace elements (Pb, Cd, Hg, Cu, Mn, Zn, Ag), which are also heavy metals, play important roles as bio-indicators for

environmental pollution (Roman and others 2007; Vit and others 2010). Overall, the individual amount of minerals and metals may differ from one honey to another. In this study, we gathered the recent data from at least 37 original articles from 6 continents and tabulated the means, SD, and ranges of the reported different major elements and minor elements as well as heavy metals (Table 2).

Minerals and heavy metal contents vary with locations

The amount of different minerals and heavy metals in honey is largely dependent on the soil composition, as well as various types of floral plants, because minerals are transported into plants through the roots and are passed to the nectar and finally into the honey produced from it (Anklam 1998). In addition to these factors, the beekeeping practices, environmental pollution, and honey processing also contribute to the diversified mineral content found to be present in honey (Pohl 2009). Different studies have indicated that almost all macrominerals were commonly found in honeys from all countries, with the exception of Cl, which was only detected in honey samples from Spain (González Paramás and others 2000).

Among the major and minor elements reported to be present in honey, K is reported to be present in the highest concentrations (Abu-Tarboush and others 1993; Terrab and others 2004a; Fernández-Torres and others 2005; Dag and others 2006; Farooq Khan and Maqbool 2008), followed by Na (Nanda and others 2003; Santos and others 2008; Daniel and others 2011; Mbiri and others 2011; Moniruzzaman and others 2014), while Mg, Ca, Fe, Zn, and Cu are present in intermediate amounts (Tuzen and Soyak 2005; Dag and others 2006; Belouali and others 2008; Vit and others 2010; Atanassova and others 2012; Mondragón-Cortez and others 2013). Although the stated amount of minerals found in honeys is shown in apparently low percentage when compared with other honey constituents, some precise elements are found in similar or even higher amounts than some common fruits and vegetables. For instance, from 100 g of honey, 0.4 mg of iron is achievable, which is higher than that of apples (0.2 mg), cranberries (0.2 mg), grapefruits (0.2 mg), kiwis (0.3 mg), mangoes (0.2 mg), papayas (0.2 mg), pineapple (0.3 mg) (United States Department of Agriculture 2015). The floral types (botanical origins) characterize not only the differences between the mineral and toxic element contents, but also the composition, taste, color, and flavor of honey (Bogdanov and others 2008; Silva and others 2009).

Honey may be monofloral or the more common polyfloral type (Silici and others 2008). Mono-floral honey originates from areas in temperate climatic zones (especially from species such as heather, linden, buckwheat, or clover) to tropical climatic zones (from species such as orange acacia, chestnut, rhododendron, eucalyptus, and rosemary) (Devillers and others 2002; Fernández-Torres and others 2005; Silici and others 2008). Toxic trace elements or heavy metals have been reported to be found in higher concentrations in honeys found in close proximity to certain industrial areas (Bratu and Georgescu 2005). Similarly, traffic-related pollution and chemical-intensive agriculture, which contaminate the air, water, and soil can also contribute to the increasing levels of these elements in honey (Street and others 2009; Silici and others 2013). Higher Pb concentrations (up to 2370 mg/kg) were detected in honey samples collected from contaminated areas in Italy (D'ambrosio and Marchesini 1982). In another study, the highest concentrations of Pb, Cd, and Cr reported to be present in honey were 45.0, 630, and 102 mg/kg, respectively, which

were higher than the tolerable intake levels (Conti and Botre 2001).

Initially, Batista and others (2012) reported that honey samples from Brazil contained 42 chemical elements (toxic and essential elements) in which some unique components such as Pd, U, Tl, Te, Hf, Sn, Sb, La, I, Sm, Tb, Dy, Sd, Th, Pr, Nd, Tm, Yb, Lu, Gd, Ho, Er, Ce, and C were reported to be present. Another study, however, indicated that 32 chemical elements were found in honeys from several European countries (Vincēviča-Gaile 2010). The summarized major and minor elements, as well as heavy metals commonly found in different countries on several continents, are as reported in Table 3 and 4.

Significance of minerals and heavy metals in honey

Honey as food and medicine has many nutritional and medicinal benefits (Ajibola and others 2012). As honey is a source of nutrients in low quantities, it would be advisable for adults to take honey in larger quantities (70 to 95 g/d) to obtain the complete desired nutritional benefits (Al-Waili 2004; Yaghoobi and others 2008; Münstedt and others 2009). In a comparative study on the importance of honey relative to sucrose in children's nutrition, honey ingestion led to the development of hematological profiles and calcium uptake, lighter and thinner bowel contents, better skin color, and improved digestion, and the honey contributed to steady weight gain and reduced disease susceptibility (Alvarez-Suarez and others 2010a). Analysis of honey composition (Table 1) indicates that minerals and heavy metals are important constituents and play specific roles in human health. A study observed that the growth of rodents following honey feeding was significantly high due to bone growth and mineralization (Chepulis and Starkey 2008). Ca is an essential mineral because of its important contribution to several biological functions in the cardiac, nervous, and musculoskeletal systems, including contributing to the formation of bone and teeth. Furthermore, Ca is involved in mineral homeostasis and physiological performance as well as acting as a cofactor for many enzymes (Theobald 2005; Huskisson and others 2007; Morgan 2008).

Another study showed that honey is beneficial to individuals suffering from anemia because it helps to improve hemoglobin concentration, to increase erythrocyte count and to elevate hematocrit level. The researchers also attributed this ability to the presence of Fe in honey as well as the increased immunity found in honey eaters (Ajibola and others 2007). In an electron microscopy and microhardness study, honey consumption was safer and provided a better environment for oral health when compared with drinking fruit juice (Grobler and others 1994). It was reported that drinking fruit juice erodes the tooth enamel in just 10 min after consumption, while erosion of teeth was not prominent or visible even up to half an hour after honey consumption. It is plausible that honey constituents such as calcium, fluoride, phosphorus, and other colloidal honey components confer a protective role, thus making honey less cariogenic. Another important point is that Na plays a vital role in the maintenance of optimum blood pressure, kidney function, and nerve and muscle functions (Hall 2003; Sobotka and others 2008). The presence of K needs to be in balance with the circulatory system like Na, and K also assists in nerve functions, heart activity, and muscle contraction (Ko and others 2008; Lambert and others 2008; Sobotka and others 2008). Another key mineral in honey is Mg, which acts as cofactor for up to 300 enzymes most of which are related to antioxidant reactions. Mg deficiency contributes to aging and

Table 3—Major elements reported in honey samples from different countries.

Country	Na (mg/kg)	K (mg/kg)	Ca (mg/kg)	Mg (mg/kg)	P (mg/kg)	S (mg/kg)	Cl (mg/kg)	References
Asia								
India	163.30–304.31	489.52–932.56	32.60–84.63	18.50–204.60	—	—	—	(Nanda and others 2003)
Israel	26.60–132.50	189.00–3768.30	58.40–137.10	21.83–199.33	47.00–651.00	22.40–188.30	—	(Dag and others 2006)
Malaysia	83.17–732.16	413.63–4026.40	65.80–567.27	18.00–39.00	—	—	—	(Moniruzzaman and others 2014)
Pakistan	71.00–89.00	340.00–1480.00	51.00–71.00	18.39–23.21	—	—	—	(Farooq Khan and Maqbool 2008)
Saudi Arabia	—	—	—	—	—	—	—	(Osman and others 2007)
Turkey	—	350.00–7030.00	15.20–232.00	21.90–67.50	—	—	—	(Silici and others 2008)
Europe								
Bulgaria	7.22–16.30	105.00–1628.00	32.00–110.00	6.00–97.00	24.00–124.00	12.00–41.00	—	(Atanassova and others 2012)
France	—	—	8.90–130.90	3.62–68.78	84.39–354.45	9.61–118.10	—	(Devillers and others 2002)
Italy	6.10–62.50	205.00–2639.00	9.10–402.60	3.90–65.00	—	—	—	(Conti and others 2007)
Poland	6.80–131.00	12.00–736.00	27.60–61.10	2.80–145.00	35.70–571.00	—	—	(Grembecka and Szefer 2013)
Spain	55.80–151.65	183.30–5570.73	15.14–202.30	23.90–1078.95	67.70–205.00	27.80–53.80	181.30–427.2	(González Paramás and others 2000; de Alda-Garcilope and others 2012)
Africa								
Kenya	98.04–269.10	172.83–564.13	19.33–70.17	12.64–41.88	—	—	—	(Mbiri and others 2011; Maiyo and others 2014)
Morocco	—	30.65–5097.91	22.32–228.57	5.02–62.80	—	—	—	(Belouali and others 2008)
Nigeria	289.00	28770.00	144.00–270.00	2660.00	—	—	—	(Omode and Ademukola 2008; Daniel and others 2011)
Sudan	14.11–28.24	17.60–74.66	35.63–82.92	23.67–177.15	28.10–204.60	46.10–131.50	—	(Mohammed and Babiker 2007)
North America								
Mexico	20.50–302.00	276.70–1760.00	38.00–127.30	13.10–61.30	47.80–104.80	13.10–62.90	—	(Mondragón-Cortez and others 2013)
South America								
Argentina	6.10–89.98	90.92–1955.75	18.60–136.14	6.01–46.57	1.17–100.66	—	—	(Cantarelli and others 2008)
Brazil	113.08–205.70	489.43–555.94	32.48–49.94	13.86–16.88	—	—	—	(Santos and others 2008)
Venezuela	—	—	98.26–214.01	12.59–79.00	—	—	—	(Vit and others 2010)
Australia								
New Zealand	1.10–110.00	34.80–3640.00	7.21–94.30	7.52–86.30	29.50–255.00	13.40–93.90	—	(Vanhanen and others 2011)

Table 4–Trace elements and heavy metal levels reported in honey samples from different countries.

Country	Fe (mg/kg)	Cu (mg/kg)	Zn (mg/kg)	Cd (μg/kg)	Pb (μg/kg)	As (mg/kg)	Ag (μg/kg)	Si (mg/kg)	B (mg/kg)	Co (μg/kg)	Ni (mg/kg)	Hg (μg/kg)	Cr (μg/kg)	Mn (mg/kg)	Se (mg/kg)	Al (mg/kg)	Mo (mg/kg)	Sr (mg/kg)	V (μg/kg)	Ti (mg/kg)	Be (μg/kg)	References
Asia																						
China	0.04–0.31	0.59–22.85	0.59–22.85	BDL–4.20	7.15–85.78	BDL–0.08	–	–	–	–	–	0.80–4.00	–	–	–	–	–	–	–	–	–	(Ru and others 2013)
India	8.86–13.25	1.74–2.90	2.50–16.77	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	(Nanda and others 2003)
Iran	2.13–9.24	0.05–2.70	0.31–7.75	330.00–530.00	60.00–340.00	0.08–0.40	–	–	–	–	–	2370.00–3510.00	–	0.15–0.62	0.06–0.26	1.85–16.12	–	–	–	–	–	(Samimi and others 2001)
Israel	0.90–9.30	9.00–3.18	0.80–11.50	–	150.00–8220.00	–	–	4.20–18.00	0.17–12.60	–	–	–	–	–	–	–	–	–	–	–	–	(Dag and others 2006)
Malaysia	55.83–233.00	ND–2.93	4.7–173.77	ND–1030.00	ND–1017.00	0.03–0.13	–	–	–	ND–107.00	–	–	–	–	–	–	–	–	–	–	–	(Moniruzzaman and others 2014)
Pakistan	4.35–7.54	1.46–1.94	1.98–2.94	–	–	–	–	–	–	840.00–1120.00	1.02–1.48	–	–	–	–	–	–	–	–	–	–	(Farooq Khan and Maqbool 2008)
Palestine	ND–7.60	2.06–8.36	–	–	–	–	–	–	–	–	ND–0.13	–	ND–350.00	–	–	–	–	–	–	–	–	(Swaileh and Abdul Khaliq 2013)
Saudi Arabia	0.31–3.19	0.24–0.39	0.21–0.75	2.00–37.00	38.00–80.00	–	–	–	–	–	–	–	–	0.19–0.37	–	–	–	–	–	–	–	(Osman and others 2007)
Turkey	0.57–8.74	0.06–0.80	0.01–5.39	0.28–240.00	1.54–106.00	–	–	–	–	1.25–30.00	1.35–1.44	–	1.24–1040.00	0.02–1.56	0.01–0.45	0.18–13.68	–	–	–	–	–	(Tuzen and Soyjak 2005; Tuzen and others 2007; Yarsan and others 2007; Leblebici and Aksoy 2008; Silici and others 2008; Silici and others 2013)
Europe																						
Bulgaria	0.35–4.37	<0.01–0.45	0.08–1.17	<10.00–310.00	<80.00–310.00	<0.1–0.320	–	–	–	<10.00–	<0.01–1.00	–	<10.00–20.00	0.06–12.70	–	0.24–1.58	–	0.12–0.40	<50	–	–	(Jivan and others 2008; Atanassova and others 2012)
Croatia	–	1.24–1272	–	1000.00–4000.00	4130.00–21590.00	10.0–499.00	–	–	–	–	–	130.00–900.00	–	–	–	–	–	–	–	–	–	(Bilandžić and others 2012)
Czech Republic	–	–	–	0.95–32.35	22.80–117.85	<0.01–0.04	–	–	–	–	–	3.24–11.31	–	–	–	–	–	–	–	–	–	(Bateklová and others 2012)
France	0.56–86.76	0.06–1.71	0.17–6.42	80.00–250.00	280.00–1080.00	–	90.00–160.00	–	–	100.00–230.00	0.09–0.34	NA	80.00–360.00	0.11–42.81	–	0.18–9.72	0.15–0.33	–	–	–	–	(Devillers and others 2002)
Italy	2.00–35.10	0.19–2.98	0.178–8.120	12.00–274.00	100.00–1533.00	–	–	–	–	–	–	–	33.30–60.00	0.08–1.17	–	–	–	–	–	–	–	(Sanna and others 2000; Conti and Botrè 2001; Conti and others 2007)
Poland	0.60–5.70	0.10–0.90	0.30–8.40	ND–	ND–	–	–	–	–	ND–300.00	ND–0.50	–	ND–500.00	0.10–8.00	–	–	–	–	–	–	–	(Grembecka and Szefer 2013)
Romania	2.04–2.91	0.01–1.20	–	10.00–1600.00	20.00–6000.00	–	–	–	–	–	0.90–2.50	–	30.00–800.00	1.75–6.51	–	–	–	–	–	–	–	(Matei and others 2004; Dobrinas and others 2006)
Spain	ND–7.59	0.36–1.36	1.19–2.76	2.70–54.20	31.50–46.32	–	–	0.01–0.04	–	BDL–136.2	0.05–0.75	–	50.10–843.30	0.98–10.9	–	3.04–8.31	–	–	–	–	–	(González Paramás and others 2000; Frías and others 2008; de Alda-Garcilope and others 2012)

(continued)

Table 4–Continued.

Country	Fe (mg /kg)	Cu (mg /kg)	Zn (mg /kg)	Cd (μg/kg)	Pb (μg/kg)	As (mg/kg)	Ag (μg/kg)	Si (mg/kg)	B (mg/kg)	Co (μg/kg)	Ni (mg/kg)	Hg (μg/kg)	Cr (μg/kg)	Mn (mg/kg)	Se (mg/kg)	Al (mg/kg)	Mo (mg/kg)	Sr (mg/kg)	V (μg/kg)	Ti (mg/kg)	Be (μg/kg)	References		
Africa																								
Egypt	21.50–38.82	BLD	1.63–2.57	BLD	860.00–1880.00	–	–	–	–	–	BLD	–	BLD	0.46–1.17	–	–	–	–	–	–	–	–	(Taha and Ali 2012)	
Ethiopia	6.33–13.61	0.37–1.124	0.370–1.124	22.00–29.00	–	–	–	–	–	0.00–249.00	1.99–2.61	–	493.00–624.00	0.99–1.49	–	–	–	–	–	–	–	–	(Nigussie and others 2012)	
Kenya	0.08–1.12	0.02–0.05	0.05–0.35	BLD–30.00	BLD–280.00	BLD–0.03	–	–	–	–	–	–	N.D.–0.15	–	–	–	–	–	–	–	–	–	(Mbiri and others 2011; Maiyo and others 2014)	
Libya	–	0.80–10.40	–	N.D.–5.00	N.D.–60.00	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	(Ahmida and others 2013)	
Morocco	0.88–207.65	0.51–4.75	0.04–2.74	1.30–24.9	36.00–1880.00	–	–	–	–	–	–	–	–	0.08–9.76	0.26–17.39	–	–	–	–	–	–	–	(Belouali and others 2008)	
Nigeria	0.12–3.42	0.01–0.02	0.04–0.11	–	N.D.–0.90	–	–	–	–	–	0.01–0.02	–	–	–	–	–	–	–	–	25.0–71.0	–	–	(Omode and Ademukola 2008; Daniel and others 2011)	
Sudan	2.05–33.65	2.94–58.12	4.86–9.61	N.D.–100.00	<450.00	–	–	–	–	5.00–1172.00	N.D.–4.06	–	<10.00	0.12–1.02	–	–	–	–	–	–	–	–	(Mohammed and Babiker 2007)	
North America																								
Cuba	4.04–10.24	0.04–0.59	<0.20–1.71	<0.10–0.50	76.70–102.00	<0.01	–	–	–	–	0.09–0.19	<10	28.2–56.9	<0.10–1.51	<10	–	–	–	<10	–	–	–	(Alvarez-Suarez and others 2012)	
Mexico	0.82–4.72	–	1.51–6.80	–	–	–	–	23.7–90.9	–	–	–	–	–	–	–	–	–	–	–	–	–	–	(Mondragón-Cortez and others 2013)	
United State	<983.30	0.08–0.13	0.31–1.27	704.20–1585.40	<983.30–1534.80	–	<2.46–6.14	–	–	–	<0.01–0.13	–	<9.83–<10.00	–	–	–	–	–	–	–	–	<9.83–<10.00	(Birge and Price 2001)	
South America																								
Argentina	1.13–10.32	0.05–0.68	0.14–3.87	–	–	–	–	–	–	–	–	–	–	0.07–0.68	–	0.02–13.04	–	–	–	–	–	–	–	(Cantarelli and others 2008)
Brazil	3.98–21.99	N.D.–2.10	0.25–0.79	<LOQ–8.00	141.00–228.00	–	–	–	–	–	–	–	83.00–94.00	0.90–3.27	–	–	–	–	–	–	–	–	–	(Santos and others 2008; de Andrade and others 2014)
Chile	0.78–7.66	0.09–4.32	0.19–4.93	N.D.–10.00	N.D.–100.00	–	–	–	–	10.00–620.00	0.01–1.48	–	10.00–1980.00	0.02–6.97	–	0.04–22.06	–	0.04–22.06	–	–	–	–	–	(Fredes and Montenegro 2006)
Venezuela	1.09–4.59	0.00–0.97	0.60–2.52	–	–	–	–	–	–	–	–	–	–	0.50–10.70	–	–	–	–	–	–	–	–	–	(Vit and others 2010)
Australia																								
New Zealand	0.67–3.39	0.70–0.09	0.20–2.46	10.00–450.00	10.00–40.00	0.04–0.17	–	–	0.05–0.49	–	0.02–0.65	–	120.00–550.00	0.18–4.75	–	0.21–21.3	N.D.–0.01	–	–	–	–	–	–	(Vanhanen and others 2011)

N.D., not detected; LOQ, limit of quantification; BDL, below detection limit.

age-related disorders (Durlach and others 1998; Huskisson and others 2007).

Trace elements are helpful when present in optimum concentrations, but at higher concentrations of trace elements (heavy metals) contribute to toxicity in humans. The toxicity occurs due to the inability of the heavy metal to be metabolized by the body, leading to accumulation in human or animal soft tissues without being fully inactivated or destroyed (Ajibola and others 2012). Health problems caused by heavy metals include headaches, metabolic abnormalities, respiratory disorders, nausea, and vomiting. For instance, Pb can cause damage to the brain, kidney, nervous system, and red blood cells (Garcá-Fernández and others 1996). According to the Agency for Toxic Substances and Disease Registry, the most hazardous heavy metals include As, Pb, and Cd. As and Cd poisonings are comparatively less common with honey, because of lower use, but contamination with Pb is frequently reported (Bogdanov 2006). Other heavy metals in honey include Cr, Zn, Hg, Mn, and Ag, all of which are toxic to human health, and they play important roles as bio-indicators of environmental pollution (González Paramás and others 2000; Celli and Maccagnani 2003; Vit and others 2010).

Determination of different minerals using different techniques

A number of techniques are used to determine chemical elements, such as minerals, metals, and nonmetals, at low concentrations in honey samples. Different studies on detecting minerals and heavy metals in honey samples have utilized spectrometry and several other instrumental methods. These analyses are important to determine the nutritional value and also the potential effect of honey on human well-being, and they can be called upon to ensure the general safety and purity of honey. However, analysis of minerals in honey is challenging due to the complex organic matter of the matrix (Ajtony and others 2007). The commonly used techniques are flame emission spectrometry, flame atomic absorption spectrometry, inductively coupled plasma optical emission spectrometry, and inductively coupled plasma mass spectrometry. In some cases, ethylenediaminetetraacetic acid titration or precipitation titrations are used to identify Ca and Ag, respectively (Table 5).

There are also some reported marginal uses of ion chromatography (Buldini and others 2001), total reflection X-ray fluorescence spectrometry (Kump and others 1996; Golob and others 2005), potentiometric stripping analysis (Muñoz and Palmero 2006), and anodic stripping voltammetry (Khammas and others 2012). However, the application of these diverse techniques is dependent on instrumental availability in the laboratory. Although different processes can determine the aforementioned elements, they differ in speed, precision, sensitivity, specificity, and accuracy (Table 5). Acid digestion is usually required in these analyses for honey sample preparation. Then, the absorption of radiation is measured at specific wavelengths corresponding to the mineral of interest. The instruments must be calibrated and standardized using relevant working standards. The concentrations of the individual elements are then measured after the instrument is accurately calibrated.

Color correlation between minerals and honey

The appearance of food is one of the factors that defines its quality and is actually a consumer's first impression. As a part of appearance, color has to meet consumer expectations and be within a critical range. Honey varies in color, ranging from white or pale yellow to dark red or even black. According to the USDA (1985),

honey samples with Pfund values of less than 8 mm are categorized as "water white," between 9 and 17 mm as "extra white," between 18 and 34 mm as "white," between 35 and 50 mm as "extra light amber," between 51 and 85 mm as "light amber," between 86 and 114 mm as "amber," and greater than 114 mm as "dark amber." Honey color mainly differs based on its floral origin, industrial processing methods, temperature, and storage duration (Salas and others 1993; Gonzales and others 1999). Some studies have considered that, in addition to the mineral content, the morphology and color of PGs (Terrab and others 2004b) in honey also have some effects on honey color and even taste (González-Miret and others 2005).

The mineral composition of honey also correlates with its color (Downey and others 2005; González-Miret and others 2005; Pohl 2009). Dark and amber honeys (avocado, chestnut, honeydew, heather) contain higher amounts of certain major, minor, and trace metals (such as Al, Ca, Cd, Cu, Fe, K, Mg, Mn, Na, Ni, Zn) when compared to pale-colored honeys (González-Miret and others 2005; Nalda and others 2005; Osman and others 2007; Pisani and others 2008). Amber light honeys (eucalyptus and thyme honeys) have shown rather intermediate quantities of total minerals. Interestingly, the darker-colored honeys have been reported to be mostly associated with higher concentrations of Cd, Fe, and Pb, whereas light- and brown-colored honeys are related to Al and Mg concentrations (González-Miret and others 2005). Several studies have indicated that darker honeys (such as avocado, chestnut, honeydew, and heather) contain higher levels of minerals (between 13 and 1879 mg/kg), which correlates with other studies performed on Spanish, Italian, and Moroccan honeys (Rodríguez-Otero and others 1994; Poiana and others 1996; Terrab and others 2003a). Lighter honeys (citrus, rosemary, and lavender) have lower levels of minerals (542, 588.5, and 672 mg/kg), and amber light honeys (eucalyptus and thyme) have medium quantities of total minerals (921 and 983 mg/kg, respectively; Figure 1).

Mineral levels in honey are correlated with its electrical conductivity (EC)

EC is the measure of the amount of electrical current that a material can carry. Honey is a mixture of water, minerals, acids, and mainly organic materials and is measured for its EC, which indicates the mineral and acid contents in a honey sample (Kropf and others 2008). There is a relationship between both the ash (minerals) and acid content of honey and its EC. The higher the concentration of minerals is, the greater the EC values (Louveaux and others 1973; Bogdanov and others 2007; Kropf and others 2008; Silva and others 2009; Pohl 2009). Kropf and others (2008) demonstrated a linear regression model for the relationship between ash and EC for Slovenian honey using 290 samples (including acacia, lime, chestnut, spruce, fir, multifloral, and mixed forest honeydew honeys).

Another study performed by Bogdanov and others (2007) also revealed significant positive correlations between the EC and most of the mineral elements (with the exception of Pb and Cr) on the basis of an examination of 95 honey samples from production areas in Switzerland. As mineral content determines botanical origins and the color of honey, individual mineral measurements can be replaced by EC. For example, lighter honeys usually contain fewer elements than darker ones and, consequently, the value of K is lower in light honeys than in dark ones. In addition, EC, along with measurement of the trace elements Ni, Fe, Mn, and Cd, could be used for the classification and isolation of unifloral honeys from polyfloral honeys (Bogdanov and others 2004).

Table 5–Determination of different elements using various techniques.

Method	Elements	References
Flame emission photometry or spectrometry (FES)	Na, K, Ca, Li	Mbiri and others (2011)
Inductively coupled plasma optical emission spectrometry (ICP–OES)	Na, K, Ca, Li, Mg, Fe, Zn, Mn, Cu, Al, Cd, Co, Cr, Ni, Pb	Caroli and others (1999); Devillers and others (2002); Fernández-Torres and others (2005); Dobrinias and others (2006); Belouali and others (2008); Cantarelli and others (2008); Akram (2008); Leblebici and Aksoy (2008); Demirezen and Aksoy (2010); Akbari and others (2012); Ruschioni and others (2013)
Inductively coupled plasma mass spectrometry (ICP-MS)	Al, Cu, Pb, Zn, Mn, Cd, Tl, Co, Ni, Rb, Ba, Be, Bi, U, V, Fe, Pt, Pd, Te, Hf, Mo, Sn, Sb, P, La, Mg, I, Sm, Tb, Dy, Sd, Th, Pr, Nd, Tm, Yb, Lu, Gd, Ho, Er, Ce, Cr	Caroli and others (1999); Bogdanov and others (2007); Musgrove and others (2010); Batista and others (2012)
Flame atomic absorption spectrometry (FAAS)	Na, K, Ca, Li, Mg, Fe, Zn, Hg, Mn, Cu, Cd, Co, Cr, Ni, Pb	Osman and others (2007); Farooq Khan and Maqbool (2008); Omode and Ademukola (2008); Belouali and others (2008); Agbagwa and others (2011); Mbiri and others (2011); Nigusse and others (2012); Taha and Ali (2012); Saghaei and others (2012); Ahmida and others (2013); Grembecka and Szefer (2013); Ru and others (2013); Moniruzzaman and others (2014); Akhtar and others (2014)
Electrothermal atomic absorption spectrometry (ET-AAS)	Fe, Zn, Mn, Cu, Al, Cd, Co, Cr, Ni, Pb	Vinas and others (1997); de Andrade and others (2014)
Graphite furnace atomic absorption spectrometry (GF-AAS)	Cd, Pb	Silici and others (2013); Ru and others (2013)
Hydride generation-atomic fluorescence spectrometry (HG-AAS)	As, Hg	Muñoz and others (1999); Mbiri and others (2011); Ru and others (2013)
Ion chromatography	Fe, Cu, Zn, Pb, Cd, Co	Buldini and others (2001)
EDTA titration	Ca, Mg	Farooq Khan and Maqbool (2008)

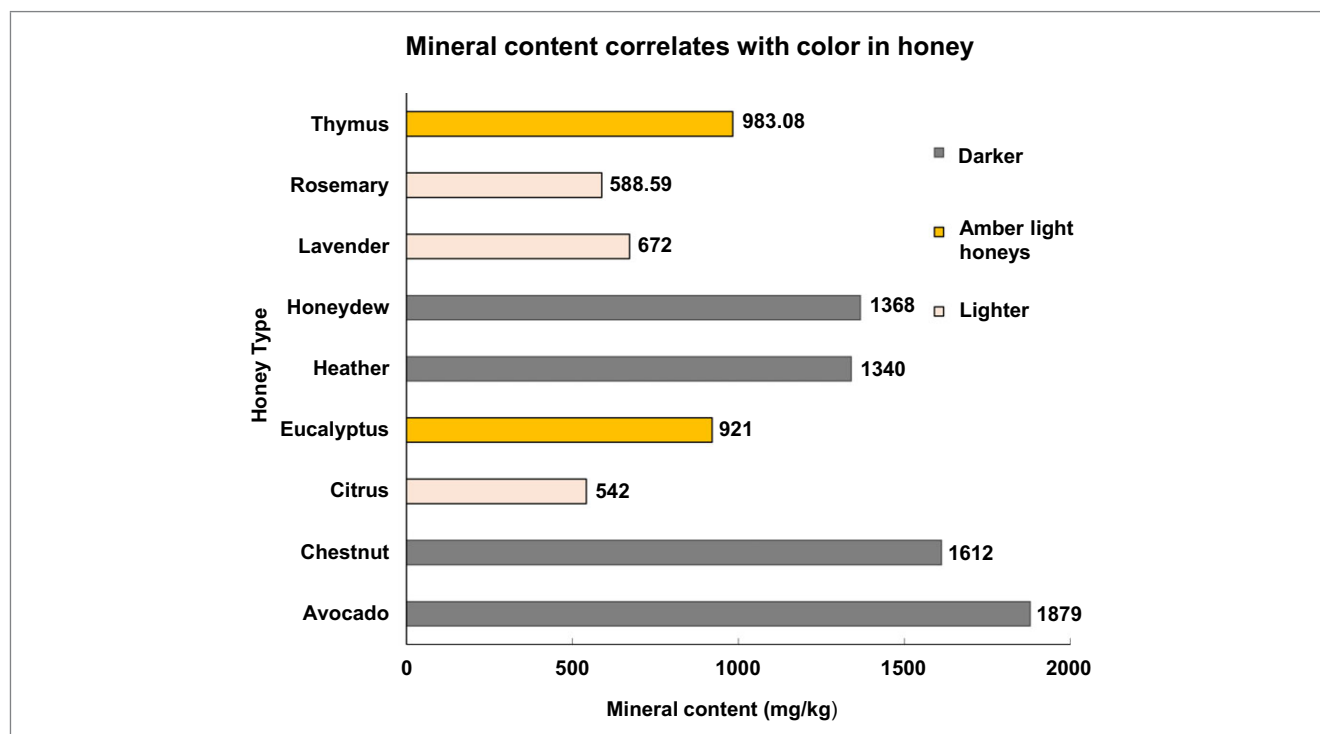


Figure 1–Reported concentrations of minerals and metals in darker honeys (avocado, chestnut, honeydew, and heather honeys) compared to amber light honeys (eucalyptus and thyme honeys) or lighter honeys (citrus, rosemary, and lavender honeys).

Currently, EC is one of the most beneficial quality parameters and has been included in the new international standards for honey (Commission 1981; Alimentarius 2001).

Correlation of mineral concentrations in honey with the age of honeycombs

The mineral contents in honey depend on not only botanical origin (Abu-Tarboush and others 1993; Anklam 1998; Nanda

and others 2003; Fernández-Torres and others 2005; Bogdanov and others 2007), geographical origin, and environment conditions (Anklam 1998; González Paramás and others 2000; Bratu and Georgescu 2005; Bogdanov and others 2007; Kropf and others 2010), but also the length of storage (Badei and Shawer 1986) and the age of the honeycombs (Taha and others 2010). Honeycombs are masses of hexagonal wax cells built by honey bees to store honey and pollen, and also contain their larvae. Taha and

others (2010) showed the levels of K, Na, Ca, Mg, Fe, Zn, Pb, and Mn in clover honey significantly depended on the age of combs. They observed significant ($P < 0.01$) positive correlations between all determined mineral elements in clover honey with the age of honeycombs. They found the highest mineral values in honey and wax from combs more than 4 y old. Ni was not detected in younger combs, but in combs more than 4 y old, the content was 1.00 mg/kg. Taha and El-Sanat (2007) found the color intensity and the EC of honey significantly correlated to the age of the comb. The color intensity and the EC of honey increases with a rise in mineral level (Lynn and others 1936; González-Miret and others 2005; Kropf and others 2008). These results are similar to those of another study performed by Bogdanov and others (2007) who established significant positive correlations between chemical elements (Fe, Zn, Mn, and Pb) in honey and the age of the honeycomb.

Mineral contents: honey antioxidant activity

Free radicals are atoms, molecules, or ions having one or more unpaired electrons and are very reactive and toxic to cells (Surai 2003). The harmful effects of free radicals are lessened by the body's antioxidant defenses. These defense mechanisms against free-radical damage usually involve vitamin C, vitamin E, and beta-carotene (and other carotenoids) as the main vitamin antioxidant sources. Furthermore, several metal-containing enzymes, which include glutathione peroxidase (Se), catalase (Fe), and superoxide dismutase (Zn, Cu, and Mn), are also critical in defending the internal cellular components from oxidative damage (McDowell and others 2007). The dietary and tissue balance of all these nutrients (vitamins and trace elements) are important in protecting tissues against free-radical damage as well as participating in immune functions. However, when there is a lack of equilibrium in the organism between the production of free radicals and the antioxidant protective activity, then an "oxidative stress" condition is developed (Bogdanov 2011).

A varied range of elements is present in honey, many of which are known to have antioxidant properties. The naturally occurring antioxidants present in honey are flavonoids, phenolic acids, and some enzymes (such as glucose oxidase, catalase), ascorbic acid, carotenoid-like substances, organic acids, Maillard reaction products, amino acids, and proteins (Gheldof and others 2002; Aljadi and Kamaruddin 2004; Nagai and others 2006; Pérez and others 2007; Vela and others 2007; Khalil and others 2011; Alam and others 2014). Pernaet and others (2012) showed that there was a statistically significant correlation ($P < 0.05$) between metal content and both total phenolic and antioxidant activities, based on 78 samples of southern Italian honeys from 5 different floral origins (chestnut, eucalyptus, citrus, multifloral, and sulla). They observed correlations between total phenolic and Co, Cr, Fe, Pb, and Zn contents ($r = 0.34, 0.48, 0.58, 0.37, \text{ and } 0.38$, respectively). They also showed that the correlation coefficients between total flavonoid and metal contents were low and not significant, except for the correlations between total flavonoid and both Co and Fe contents, with $r = 0.22$ ($P < 0.05$) and 0.42 ($P < 0.001$), respectively. Another study by Ferreira and others (2009) concluded that dark honey had better antioxidant activity and phenolic content than the other studied honey (amber and light) samples. From the above studies, it can be concluded that with an increase in metal content in honey (color intensity is positively correlated with metal content in honey), there is an increase in its antioxidant properties. There is a significant relationship between the mineral contents and antioxidant properties of honey, but the necessary data are not

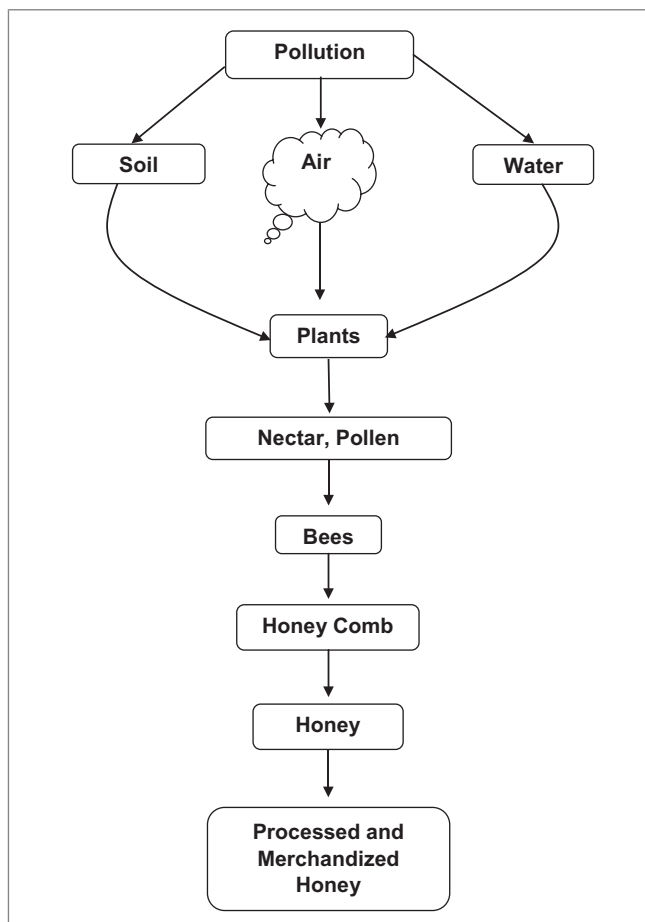


Figure 2—Schematic overview of natural and anthropogenic sources of heavy metal pollutants in honey which can influence honey composition.

currently available. Therefore, appropriate studies are warranted to verify and support this correlation.

Honey as a bio-indicator of environmental pollution

Honey's composition, flavor, and aroma are derived from the plant utilized by the bees, as well as regional and climatic conditions (Singh and Bath 1997; Mendes and others 1998). Therefore, the specific composition of honey and the possible presence of contaminants are also dependent on the crops surrounding the beehives (Kujawski and Namieśnik 2008; Aliferis and others 2010). The bees are considered biological indicators due to their important morphological, ecological, and behavioral characteristics. They act as detectors of environmental pollution in 2 ways: (1) high mortality rates in the presence of toxic molecules and (2) the presence of residues in honey, pollen, and larvae due to the excessive existence of heavy metals, fungicides, and herbicides that are normally harmless to bees (Samimi and others 2001; Celli and Maccagnani 2003). Therefore, honey can be considered as a bio-indicator for assessing the possible presence of environmental pollutants like heavy metals (Conti and Botrè 2001; Przybyłowski and Wilczyńska 2001).

The heavy metals present in honey may be of environmental origin (soil, water, and air) or result from various anthropogenic activities such as agricultural practices, industries nearby, and waste dumps (Figure 2) (Przybyłowski and Wilczyńska 2001; Stankovska and others 2008; Pohl 2009; Vincēviča-Gaile 2010). Many studies

have shown that there is a direct relationship between the heavy metal contents in honey samples and the industry-related environmental pollution in the area from which the honey is harvested. For example, the city of Baia Mare and its surroundings are known as one of the most polluted areas with heavy metals both in Romania and worldwide, and Cu, Zn, Cd, and Pb are the main industrial pollutants there. Berinde and others (2013) investigated the honey samples harvested from that area and revealed a positive correlation between the metal contents in honey and those in the air, settling dust, and soils from that region. Cimino and others (1984) showed the effects of volcanic activity on honey, while Jones and others (1987) successfully established the potential use of honey as an indicator of trace elements in the United Kingdom.

In fact, chemical wastes and exhaust fumes emitted or produced by the mines and steelworks, industrial and urban areas, or highway vehicles near the bees' foraging area have been shown to increase the concentrations of certain metals in honey (Cd, Al, Ca, Ba, Cu, Mn, Ni, Pb, Zn, or Pd) (Üren and others 1998; Przybyłowski and Wilczyńska 2001; Bratu and Georgescu 2005; Tuzen and others 2007; Stankovska and others 2008) depending on the type of flowers visited by the bees. For instance, honeys produced from the nectar of pungent plants are categorized by high concentrations of heavy metals because these plants tend to concentrate pollutants more easily than herbaceous plants. In addition, honeys derived from caduceus trees usually contain fewer pollutants than honeys of other botanical origins (Devillers and others 2002). Honey can also be contaminated during its processing by the beekeepers, by equipment and tools used, and by the process itself. Materials such as aluminum, stainless steel, and galvanized steel used in tools and equipment for the processing of honey may release some polluting metals (including Al, Cd, Co, Cr, Cu, Fe, Pb, Ni, and Zn) into honey (Üren and others 1998; González Paramás and others 2000; Santos and others 2008; Stankovska and others 2008; Tuzen and Soylak 2009).

Future Directions

The analysis of minerals and heavy metals in honey gives an indication of its geographical and botanical origins, as well as types, source, and degree of contamination, and also an overall measure of honey purity. Such information, where displayed, can help consumers make informed decision when purchasing honey and also help beekeepers to avoid possible contamination. In addition, honey can be used as a monitor of environmental pollution in an area. Heavy metals in honey indicate the level of pollution and the type of toxic metals, which could assist regulatory agencies in taking proper measures for environmental protection. Moreover, the minerals in honeys have medicinal and nutritional value that correlates with antioxidants and antioxidant activities. Considering these benefits, future studies are needed to obtain more extensive data from honey samples from all over the world to emphasize honey's medicinal and nutritional properties.

Interest in the analysis of the mineral and heavy metal contents in honey has been increasing to demonstrate their nutritive roles and correlate the data with some important antioxidant parameters. Different studies from all over the world have reported on the huge number of minerals containing heavy metals, with the common objective of characterizing various types of honeys to ensure their safety. Such information is important to guide honey production away from areas near highways, railways, and volcanic emissions and into areas, where the environment is clean. This information will also help in selecting the equipment used in the processing and storing of honey after harvesting, as this equipment

can be a possible source of product contamination with heavy metals. In addition, such information can assist in the development of a quality control protocol for maintaining the honey trade in local and international markets. This would help to prevent health problems associated with honey toxicity, especially poisoning with As, Cd, and Pb. Furthermore, minerals in honey correlate with color, EC, storage time, and even antioxidant activities. Therefore, determination of the mineral contents in honey will assist consumers in choosing a honey of interest based on its quality. It can also encourage local consumption as well as provide more income to the producing country. Additionally, to ensure its high quality and purity, it would be interesting to establish appropriate techniques to remove toxic contaminants and heavy metals from honey.

Conclusion

Honey consumption, as a nutraceutical agent, is associated with various nutritional benefits and therapeutic potential. The biological activity of honey is affected by its complex and crucial components. The composition of honey is strongly influenced by both natural and anthropogenic factors, which vary based on botanical and geographical origins. Although minerals and heavy metals are minor constituents of honey, they play vital roles in determining honey quality. Chemical elements in honey samples throughout the world vary in terms of concentrations and are also influenced by environmental pollution.

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Author Contributions

Md. Solayman, Md. Asiful Islam, Sudip Paul, and Yousuf Ali searched the literature, compiled data, interpreted the data, and wrote the manuscript part by part. Sudip Paul and Md. Asiful Islam additionally contributed with precise designing and critical revision of the manuscript. Nadia Alam, Ibrahim Khalil, and Siew Hua Gan provided the overall concept, critically edited, and approved the final version of the manuscript.

References

- Abu-Tarboush HM, Al-Kahtani HA, El-Sarrage M. 1993. Floral-type identification and quality evaluation of some honey types. *Food Chem* 46:13–7.
- Agbagwa O, Otokunefor T, Frank-Peterside N. 2011. Quality assessment of Nigeria honey and manuka honey. *J Microbiol Biotechnol Res* 1:20–31.
- Ahmida MH, Elwerfali S, Agha A, Elagori M, Ahmida NH. 2013. Physicochemical, heavy metals and phenolic compounds analysis of Libyan honey samples collected from Benghazi during 2009–2010. *Food Nutr Sci* 4:33–40.
- Ajibola A, Idowu G, Amballi A, Oyefuga O, Iqout I. 2007. Improvement of some haematological parameters in albino rats with pure natural honey. *J Biol Sci Res* 2:67–9.
- Ajibola A, Chamunorwa JP, Erlwanger KH. 2012. Nutraceutical values of natural honey and its contribution to human health and wealth. *Nutr Metab* 9:1–12.
- Ajtony Z, Bencs L, Haraszti R, Szigeti J, Szoboszlai N. 2007. Study on the simultaneous determination of some essential and toxic trace elements in honey by multi-element graphite furnace atomic absorption spectrometry. *Talanta* 71:683–90.
- Akbari B, Gharanfali F, Khayyat MH, Khashyarmansh Z, Rezaee R, Karimi G. 2012. Determination of heavy metals in different honey brands from Iranian markets. *Food Addit Contam Part B* 5:105–11.

- Akhtar S, Ali J, Javed B, Hassan S, Abbas S, Siddique M. 2014. Comparative physicochemical analysis of imported and locally produced Khyber Pakhtunkhwa honey. *GJBB* 9:55–9.
- Akram S. 2008. Characterization of Saudi Arabian floral honeys by their physicochemical characteristics and heavy metal contents. *FEB* 17:877–81.
- Al ML, Daniel D, Moise A, Bobis O, Laslo L, Bogdanov S. 2009. Physico-chemical and bioactive properties of different floral origin honeys from Romania. *Food Chem* 112:863–7.
- Alam F, Islam MA, Gan SH, Khalil MI. 2014. Honey: a potential therapeutic agent for managing diabetic wounds. *Evid Based Complement Alternat Med* 2014:1–16.
- Aliferis KA, Tarantilis PA, Harizanis PC, Alissandrakis E. 2010. Botanical discrimination and classification of honey samples applying gas chromatography/mass spectrometry fingerprinting of headspace volatile compounds. *Food Chem* 121:856–62.
- Alimentarius C. 2001. Codex standard 12. Available from: <http://www.codexalimentarius.net>. Accessed 2015 January 14.
- Aljadi A, Kamaruddin M. 2004. Evaluation of the phenolic contents and antioxidant capacities of two Malaysian floral honeys. *Food Chem* 85:513–8.
- Alvarez-Suarez JM, Tulipani S, Romandini S, Bertoli E, Battino M. 2010a. Contribution of honey in nutrition and human health: a review. *Med J Nut Metab* 3:15–23.
- Alvarez-Suarez JM, Tulipani S, Díaz D, Estevez Y, Romandini S, Giampieri F, Damiani E, Astolfi P, Bompadre S, Battino M. 2010b. Antioxidant and antimicrobial capacity of several monofloral Cuban honeys and their correlation with color, polyphenol content and other chemical compounds. *Food Chem Toxicol* 48:2490–9.
- Alvarez-Suarez JM, Giampieri F, Damiani E, Astolfi P, Fattorini D, Regoli F, Quiles JL, Battino M. 2012. Radical-scavenging activity, protective effect against lipid peroxidation and mineral contents of monofloral Cuban honeys. *Plant Foods Hum Nutr* 67:31–8.
- Al-Waili NS. 2004. Natural honey lowers plasma glucose, C-reactive protein, homocysteine, and blood lipids in healthy, diabetic, and hyperlipidemic subjects: comparison with dextrose and sucrose. *J Med Food* 7:100–7.
- Anklam E. 1998. A review of the analytical methods to determine the geographical and botanical origin of honey. *Food Chem* 63:549–2.
- Anupama D, Bhat K, Sapna V. 2003. Sensory and physico-chemical properties of commercial samples of honey. *Food Res Int* 36:183–1.
- Atanassova J, Yurukova L, Lazarova M. 2012. Pollen and inorganic characteristics of Bulgarian unifloral honeys. *Czech J Food Sci* 30: 520–6.
- Badei A, Shower M. 1986. Effect of long storage on the chemical composition and flavour constituents of Egyptian clover honey. *J Agric Res Tanta Univ* 12:166–75.
- Baroni MV, Arrua C, Nores ML, Fayé P, del Pilar Díaz M, Chiabrando GA, Wunderlin DA. 2009. Composition of honey from Córdoba (Argentina): assessment of North/South provenance by chemometrics. *Food Chem* 114:727–3.
- Batelková P, Borkovcová I, Čelechovská O, Vorlová L, Bartáková K. 2012. Polycyclic aromatic hydrocarbons and risk elements in honey from the South Moravian region (Czech Republic). *Acta Vet Brno* 81:169–4.
- Batista B, Da Silva L, Rocha B, Rodrigues J, Berretta-Silva A, Bonates T, Gomes V, Barbosa R, Barbosa F. 2012. Multi-element determination in Brazilian honey samples by inductively coupled plasma mass spectrometry and estimation of geographic origin with data mining techniques. *Food Res Int* 49:209–5.
- Belay A, Solomon W, Bultossa G, Adgaba N, Melaku S. 2013. Physicochemical properties of the Hareenna forest honey, Bale, Ethiopia. *Food Chem* 141:3386–92.
- Belitz H, Grosch W, Schieberle P. 2009. *Food Chemistry*. 4th revised and extended ed. Berlin/ London: Springer, 1070 pages.
- Belouali H, Bouaka M, Hakkou A. 2008. Determination of some major and minor elements in the east of Morocco honeys through inductively coupled plasma optical emission spectrometry. *Apiacta* 43:17–24.
- Berinde ZM, Michnea AM. 2013. A comparative study on the evolution of environmental and honey pollution with heavy metals. *J Sci Arts* 2:173–80.
- Bianchi F, Careri M, Musci M. 2005. Volatile norisoprenoids as markers of botanical origin of Sardinian strawberry-tree (*Arbutus unedo* L.) honey: characterisation of aroma compounds by dynamic headspace extraction and gas chromatography–mass spectrometry. *Food Chem* 89:527–32.
- Bilandžić N, Đokić M, Sedak M, Varenina I, Kolanović BS, Končurac A, Šimić B, Rudan N. 2012. Content of five trace elements in different honey types from Koprivnica-Kri Evci County. *Slov Vet Res* 49:167–75.
- Birge W, Price D. 2001. Analysis of metals and polychlorinated biphenyl (PCB) residues in honey bees, honey and pollen samples collected from the Paducah gaseous diffusion plant and other areas. Draft report submitted to Jon Maybriar, Division of Waste Management Kentucky Department for Environmental Protection. Available from: http://www.uky.edu/R_research/Superfund/images/pdf/22%20PCB%20Metals%20in%20bees%20honey%20pollen%20030901%20Report.pdf. Accessed 2011 September.
- Bogdanov S, Haldimann M, Luginbuhl W, Gallmann P. 2007. Minerals in honey: environmental, geographical and botanical aspects. *J Apic Res* 46:269–75. Available from: <http://www.bee-hexagon.net>; Accessed 2015 January 20.
- Bogdanov S. 2006. Contaminants of bee products. *Apidologie* 37:1–18.
- Bogdanov S. 2011. Honey as nutrient and functional food: a review. *Bee Product Sci* 1–31. Available from: <http://www.bee-hexagon.net>; Accessed 2015 January 20.
- Bogdanov S, Ruoff K, Persano Oddo L. 2004. Physico-chemical methods for the characterisation of unifloral honeys: a review. *Apidologie* 35:54–17.
- Bogdanov S, Jurendic T, Sieber R, Gallmann P. 2008. Honey for nutrition and health: a review. *J Am Coll Nutr* 27:677–89.
- Bratu I, Georgescu C. 2005. Chemical contamination of bee honey—identifying sensor of the environment pollution. *JCEA* 6:95–8.
- Buba F, Gidado A, Shugaba A. 2013. Analysis of biochemical composition of honey samples from North-East Nigeria. *Biochem Anal Biochem* 2:3–10.
- Buldini PL, Cavalli S, Mevoli A, Sharma JL. 2001. Ion chromatographic and voltammetric determination of heavy and transition metals in honey. *Food Chem* 73:487–95.
- Cantarelli M, Pellerano R, Marchevsky E, Camiña J. 2008. Quality of honey from Argentina: study of chemical composition and trace elements. *J Argent Chem Soc* 96:33–41.
- Caroli S, Forte G, Iamiceli A, Galoppi B. 1999. Determination of essential and potentially toxic trace elements in honey by inductively coupled plasma-based techniques. *Talanta* 50:327–36.
- Celli G, Maccagnani B. 2003. Honey bees as bioindicators of environmental pollution. *Bull Insectol* 56:137–9.
- Chefrour C, Draiaia R, Tahar A, Kaki YA, Bennadja S, Battesti M. 2009. Physicochemical characteristics and pollen spectrum of some north-east Algerian honeys. *AJFAND* 9:1276–93.
- Chepulis L, Starkey N. 2008. The long-term effects of feeding honey compared with sucrose and a sugar-free diet on weight gain, lipid profiles, and DEXA measurements in rats. *J Food Sci* 73:H1–7.
- Cimino G, Ziino M, Panuccio MR. 1984. Heavy metal pollution. Part X: impact of volcanic activity on Etnean honey. *Environ Technol* 5:453–6.
- Commission CA. 1981. Revised Codex Standard for Honey Codex Stan 12–1981, Rev. 1 (1987), Rev. 2 (2001).
- Conte LS, Miorini M, Giomo A, Bertacco G, Zironi R. 1998. Evaluation of some fixed components for unifloral honey characterization. *J Agric Food Chem* 46:1844–9.
- Conti ME, Botrè F. 2001. Honeybees and their products as potential bioindicators of heavy metals contamination. *Environ Monit Asses* 69:267–82.
- Conti ME, Stripeikis J, Campanella L, Cucina D, Tudino MB. 2007. Characterization of Italian honeys (Marche Region) on the basis of their mineral content and some typical quality parameters. *Chem Cent J* 1:1–10.
- Corredera L, Bayarri S, Pérez-Arquillué C, Lázaro R, Molino F, Herrera A. 2014. Evaluation of heavy metals and polycyclic aromatic hydrocarbons in honeys from different origins. *J Food Prot* 77:504–9.
- Dag A, Afik O, Yeselson Y, Schaffer A, Shafir S. 2006. Physical, chemical and palynological characterization of avocado (*Persea Americana* Mill.) honey in Israel. *Int J Food Sci Technol* 41:387–94.
- D’ambrosio M, Marchesini A. 1982. Research on contamination by heavy metals in honey sample. *Atti della Societa Italiana di Scienze Naturali* 123:342–8.
- Daniel V, Daniang I, Nimyel N. 2011. Phytochemical analysis and mineral elements composition of *Ocimum basilicum* obtained in JOS METROPOLIS, Plateau State, Nigeria. *IJET-IJENS* 11:161–5.
- D’arcy BR, Rintoul GB, Rowland CY, Blackman AJ. 1997. Composition of Australian honey extractives. 1. Norisoprenoids, monoterpenes, and other natural volatiles from blue gum (*Eucalyptus leucocylon*) and yellow box (*Eucalyptus melliodora*) honeys. *J Agric Food Chem* 45:1834–43.
- de Alda-Garcilope C, Gallego-Picó A, Bravo-Yagüe J, Garcinuño-Martínez R, Fernández-Hernando P. 2012. Characterization of Spanish honeys with protected designation of origin “Miel de Granada” according to their mineral content. *Food Chem* 135:1785–8.

- de Andrade CK, dos Anjos VE, Felsner ML, Torres YR, Quinaia SP. 2014. Direct determination of Cd, Pb and Cr in honey by slurry sampling electrothermal atomic absorption spectrometry. *Food Chem* 146:166–73.
- de Oliveira Alves RM, de Carvalho CAL, de Almeida Souza B, da Silva Sodre G, Marchini LC. 2005. Caractersticas fsico-qumicas de amostras de mel. *Cinc Tecnol Aliment* 25:644–50.
- Demirezen D, Aksoy A. 2010. Determination of heavy metals in bee honey using by inductively coupled plasma optical emission spectrometry (ICP-OES). *GUJ Sci* 18:569–75.
- Devillers J, Dore J, Marengo M, Poirier-Duchene 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.
- Dobrinas S, Matei N, Soceanu A, Birghila S, Popescu V. 2006. Estimation of vitamin C and Cd, Cu, Pb content in honey and propolis. *Scientific Study Res VII*:729–34.
- Downey G, Hussey K, Kelly JD, Walshe TF, Martin P. 2005. Preliminary contribution to the characterisation of artisanal honey produced on the island of Ireland by palynological and physico-chemical data. *Food Chem* 91:347–54.
- Durlach J, Bac P, Durlach V, Rayssiguier Y, Bara M, Guiet-Bara A. 1998. Magnesium status and ageing: an update. *Magnesium Res* 11:25–42.
- Eissa AA, Hassan AS, El Rahman TAA. 2014. Determination of total aflatoxins and carbamate pesticide residues in some bee honey samples using QuEChERS method and high performance liquid chromatography. *Food Public Health* 4:209–13.
- European Union Council Directive. 2001/110/EC of 20 December 2001 relating to honey. 2002.
- Farooq Khan Z, Maqbool T. 2008. Physical and spectroscopic characterization of Pakistani honey. *Cinc Investig Agrar* 35:199–204.
- Fernandez-Torres R, Perez-Bernal JL, Bello-Lopez MA, Callejon-Mochon M, Jimenez-Sanchez JC, Guiram-Perez A. 2005. Mineral content and botanical origin of Spanish honeys. *Talanta* 65:686–91.
- Ferreira IC, Aires E, Barreira JC, Estevinho LM. 2009. Antioxidant activity of Portuguese honey samples: different contributions of the entire honey and phenolic extract. *Food Chem* 114:1438–43.
- Fredes C, Montenegro G. 2006. Heavy metal and other trace elements contents in honey bee in Chile. *Cinc Investig Agrar* 33:50–8.
- Fras I, Rubio C, Gonzalez-Iglesias T, Gutierrez J, Gonzalez-Weller D, Hardisson A. 2008. Metals in fresh honeys from Tenerife Island, Spain. *Bull Environ Contam Toxicol* 80:30–3.
- Garca-Fernandez A, Sanchez-Garca J, Gomez-Zapata M, Luna A. 1996. Distribution of cadmium in blood and tissues of wild birds. *Arch Environ Contam Toxicol* 30:252–8.
- Gheldof N, Wang X-H, Engeseth NJ. 2002. Identification and quantification of antioxidant components of honeys from various floral sources. *J Agri Food Chem* 50:5870–7.
- Golob T, Dobersek U, Kump P, Necemer M. 2005. Determination of trace and minor elements in Slovenian honey by total reflection X-ray fluorescence spectroscopy. *Food Chem* 91:593–600.
- Gonzales AP, Burin L, del Pilar Buera MA. 1999. Color changes during storage of honeys in relation to their composition and initial color. *Food Res Int* 32:185–91.
- Gonzalez Paramas AM, Gomez Barez JA, Garcia-Villanova RJ, Rivas Pala T, Ardanuy Albajar R, Sanchez J. 2000. Geographical discrimination of honeys by using mineral composition and common chemical quality parameters. *J Sci Food Agric* 80:157–65.
- Gonzalez-Miret ML, Terrab A, Hernanz D, Fernandez-Recamales MA, Heredia FJ. 2005. Multivariate correlation between color and mineral composition of honeys and by their botanical origin. *J Agric Food Chem* 53:2574–80.
- Gopalan C, Rama Sastri B, Balasubramanian S. 1989. Nutritive value of Indian foods, Hyderabad, India: National Institute of Nutrition, Indian Council of Medical Research, 156 pages.
- Grembecka M, Szefer P. 2013. Evaluation of honeys and bee products quality based on their mineral composition using multivariate techniques. *Environ Monit Assess* 185:4033–47.
- Grobler S, Du Toit I, Basson N. 1994. The effect of honey on human tooth enamel in vitro observed by electron microscopy and microhardness measurements. *Arch Oral Biol* 39:147–53.
- Hall JE. 2003. The kidney, hypertension, and obesity. *Hypertension* 41:625–33.
- Hernandez O, Fraga J, Jimenez A, Jimenez F, Arias J. 2005. Characterization of honey from the Canary Islands: determination of the mineral content by atomic absorption spectrophotometry. *Food Chem* 93:449–58.
- Huskisson E, Maggini S, Ruf M. 2007. The role of vitamins and minerals in energy metabolism and well-being. *J Int Med Res* 35:277–89.
- Islam A, Khalil I, Islam N, Moniruzzaman M, Mottalib A, Sulaiman SA, Gan SH. 2012. Physicochemical and antioxidant properties of Bangladeshi honeys stored for more than one year. *BMC Complement Altern Med* 12:1–10.
- Jivan A, Patruica S, Popescu G. 2008. Researches concerning the heavy metal content of the rape honey originating from the Banat area in the years 2006–2007. *SPASB* 41:302–8.
- Jones K. 1987. Honey as an indicator of heavy metal contamination. *Water Air Soil Pollut* 33:179–89.
- Kahraman T, Buyukunal SK, Vural A, Altunatmaz SS. 2010. Physico-chemical properties in honey from different regions of Turkey. *Food Chem* 123:41–4.
- Khalil M, Alam N, Moniruzzaman M, Sulaiman S, Gan S. 2011. Phenolic acid composition and antioxidant properties of Malaysian honeys. *J Food Sci* 76:C921–8.
- Khalil MI, Mottalib MA, Anisuzzaman A, Sathi ZS, Hye M, Shahjahan M. 2001. Biochemical analysis of different brands of unifloral honey available at the northern region of Bangladesh. *J Med Sci* 1:385–8.
- Khammas ZA, Ghali AA, Kadhim KH. 2012. Combined cloud-Point extraction and spectrophotometric detection of lead and cadmium in honey samples using a new ligand. *Int J Chem Sci* 10:1185–204.
- Ko EA, Han J, Jung ID, Park WS. 2008. Physiological roles of K⁺ channels in vascular smooth muscle cells. *J Smooth Muscle Res* 44:65–81.
- Kropf U, Jamnik M, Bertoneclj J, Golob T. 2008. Linear regression model of the ash mass fraction and electrical conductivity for Slovenian honey. *Food Technol Biotechnol* 46:335–40.
- Kropf U, Korosec M, Bertoneclj J, Ogrinc N, Necemer M, Kump P, Golob T. 2010. Determination of the geographical origin of Slovenian black locust, lime and chestnut honey. *Food Chem* 121:839–46.
- Kucuk M, Kolaylı S, Karaođlu , Ulusoy E, Baltacı C, Candan F. 2007. Biological activities and chemical composition of three honeys of different types from Anatolia. *Food Chem* 100:526–34.
- Kujawski M, Namiesnik J. 2008. Challenges in preparing honey samples for chromatographic determination of contaminants and trace residues. *TRAC* 27:785–93.
- Kump P, Necemer M, Snajder J. 1996. Determination of trace elements in bee honey, pollen and tissue by total reflection and radioisotope X-ray fluorescence spectrometry. *Spectrochim Acta Part B Spectrosc* 51:499–507.
- Lambert I, Hoffmann E, Pedersen S. 2008. Cell volume regulation: physiology and pathophysiology. *Acta Physiol* 194:255–82.
- Lammertyn J, Veraverbeke EA, Irudayaraj J. 2004. zNoseTM technology for the classification of honey based on rapid aroma profiling. *Sens Actuators B Chem* 98:54–62.
- Leblebici Z, Aksoy A. 2008. Determination of heavy metals in honey samples from Central Anatolia using plasma optical emission spectroscopy (ICP-OES). *Pol J Environ Stud* 17:549–55.
- Louveau J, Pourtallier M, Vorwohl G. 1973. Methodes d’analyses des miels. *Conductivit Bull Apic* 16:1–3.
- Lynn E, Englis D, Milum V. 1936. Effect of processing and storage on composition and color of honey. *J Food Sci* 1:255–61.
- Ma L. 2009. International comparison of the export competitiveness of Chinese honey. *Asian Agric Res* 1:17–20.
- Mahmoudi R, Zare P, Tajik H, Shadfar S, Nyiazpour F. 2012. Biochemical properties and microbial analysis of honey from north-western regions of Iran: seasonal effects on physicochemical properties of honey. *African J Biotechnol* 11:10227–31.
- Maiyo W, Kituyi J, Mitei Y, Kagwanja S. 2014. Heavy metal contamination in raw honey, soil and flower samples obtained from Baringo and Keiyo Counties, Kenya. *IJESE* 2:5–9.
- Marchini LC, Moreti ACDC, Otsuk IP, Sodre GDS. 2007. Physicochemical composition of *Apis mellifera* honey samples from Sao Paulo State, Brazil. *Quim Nova* 30:1653–7.
- Marghitas LA, Dezmierean DS, Pocol CB, Marioara I, Bobis O, Gergen I. 2010. The development of a biochemical profile of acacia honey by identifying biochemical determinants of its quality. *Notulae Botanicae Horti Agrobotanici Cluj-Napoca* 38:84–90.

- Matei N, Birghila S, Dobrinas S, Capota P. 2004. Determination of C vitamin and some essential trace. *Acta Chim Slov* 51:169–75.
- Mbiri A, Onditi A, Oyaro N, Murago E. 2011. Determination of essential and heavy metals in Kenyan honey by atomic absorption and emission spectroscopy. *JAST* 13:107–15.
- McDowell LR, Wilkinson N, Madison R, Felix T. 2007. Vitamins and minerals functioning as antioxidants with supplementation considerations. The Proceedings of the Florida Ruminant Nutrition Symposium Best Western Gateway Grand Gainesville, Fla. January 30–31. Available from: <http://dairy.ifas.ufl.edu/files/rns/2007/Mcdowell.pdf>. Accessed 2015 January 20.
- Mckee B. 2003. Prevention of residues in honey: a future perspective. *Apiacta* 38:173–7.
- Mendes E, Proença EB, Ferreira I, Ferreira M. 1998. Quality evaluation of Portuguese honey. *Carbohydr Polym* 37:219–23.
- Mendes TM, Baccan SN, Cadore S. 2006. Sample treatment procedures for the determination of mineral constituents in honey by inductively coupled plasma optical emission spectrometry. *J Braz Chem Soc* 17:168–76.
- Mohammed S, Babiker EE. 2007. Mineral elements and some toxic metals content in a single source Alaasal. Available from: <http://saudibi.com/>. Accessed 2015 January 3.
- Mondragón-Cortez P, Ulloa J, Rosas-Ulloa P, Rodríguez-Rodríguez R, Resendiz Vázquez J. 2013. Physicochemical characterization of honey from the West region of México. *CyTA-J Food* 11:7–13.
- Moniruzzaman M, Khalil MI, Sulaiman SA, Gan SH. 2013. Physicochemical and antioxidant properties of Malaysian honeys produced by *Apis cerana*, *Apis dorsata* and *Apis mellifera*. *BMC Complement Altern Med* 13:1–12.
- Moniruzzaman M, Chowdhury MAZ, Rahman MA, Sulaiman SA, Gan SH. 2014. Determination of mineral, trace element, and pesticide levels in honey samples originating from different regions of Malaysia compared to Manuka honey. *BioMed Res Int* 2014:1–10.
- Morgan KT. 2008. Nutritional determinants of bone health. *J Nutr Elder* 27:3–27.
- Muñoz E, Palmero S. 2006. Determination of heavy metals in honey by potentiometric stripping analysis and using a continuous flow methodology. *Food Chem* 94:478–83.
- Muñoz O, Vélez D, Montoro R. 1999. Optimization of the solubilization, extraction and determination of inorganic arsenic [As (III)+ As (V)] in seafood products by acid digestion, solvent extraction and hydride generation atomic absorption spectrometry. *Analyst* 124:601–7.
- Münstedt K, Hoffmann S, Hauenschild A, Bütle M, von Georgi R, Hackethal A. 2009. Effect of honey on serum cholesterol and lipid values. *J Med Food* 12:624–8.
- Musgrove M, Stern L, Banner J. 2010. Springwater geochemistry at Honey Creek State Natural Area, central Texas: implications for surface water and groundwater interaction in a karst aquifer. *J Hydrol* 388:144–56.
- Nagai T, Inoue R, Kanamori N, Suzuki N, Nagashima T. 2006. Characterization of honey from different floral sources. Its functional properties and effects of honey species on storage of meat. *Food Chem* 97:256–62.
- Nalda MN, Yagüe JB, Calva JD, Gómez MM. 2005. Classifying honeys from the Soria Province of Spain via multivariate analysis. *Anal Bioanal Chem* 382:311–9.
- Nanda V, Sarkar B, Sharma H, Bawa A. 2003. Physico-chemical properties and estimation of mineral content in honey produced from different plants in northern India. *J Food Compos Anal* 16:613–9.
- Nickel EH. 1995. The definition of a mineral. *Mineral J* 17:346–9.
- Nielsen FH. 1984. Ultratrace elements in nutrition. *Annu Rev Nutr* 4:21–41.
- Nigusie K, Subramanian P, Mebrahtu G. 2012. Physicochemical analysis of Tigray honey: an attempt to determine major quality markers of honey. *Bull Chem Soc Ethiop* 26:127–33.
- Omafuybe B, Akanbi O. 2009. Microbiological and physico-chemical properties of some commercial Nigerian honey. *Afr J Microbiol Res* 3:891–6.
- Omode P, Ademukola S. 2008. Determination of trace metals in southern Nigerian honey by use of atomic absorption spectroscopy. *Spectrosc Lett* 41:328–31.
- Osman KA, Al-Doghairi MA, Al-Rehiyani S, Helal MI. 2007. Mineral contents and physicochemical properties of natural honey produced in Al-Qassim region, Saudi Arabia. *JFAE* 5:142–46.
- Pérez RA, Iglesias MT, Pueyo E, González M, de Lorenzo C. 2007. Amino acid composition and antioxidant capacity of Spanish honeys. *J Agric Food Chem* 55:360–5.
- Perna A, Simonetti A, Intaglietta I, Sofo A, Gambacorta E. 2012. Metal content of southern Italy honey of different botanical origins and its correlation with polyphenol content and antioxidant activity. *Int J Food Sci Technol* 47:1909–17.
- Pisani A, Protano G, Riccobono F. 2008. Minor and trace elements in different honey types produced in Siena County (Italy). *Food Chem* 107:1553–60.
- Pohl P. 2009. Determination of metal content in honey by atomic absorption and emission spectrometry. *TRAC* 28:117–28.
- Poiana M, Fuda S, Manziu E, Postorino S, Mincione B. 1996. Ricerche sui mieli commercializzati in Italia: la componente minerale. *Industrie alimentari* 35:522–30.
- Przybyłowski P, Wilczyńska A. 2001. Honey as an environmental marker. *Food Chem* 74:289–91.
- Qamer S, Ahmad F, Latif F, Ali SS, Shakoori AR. 2008. Physicochemical analysis of *Apis dorsata* honey from Terai forests, Nepal. *Pak J Zool* 40:53–8.
- Rodríguez-Otero J, Paseiro P, Simal J, Cepeda A. 1994. Mineral content of the honeys produced in Galicia (north-west Spain). *Food Chem* 49:169–71.
- Roman A, Bartkowiak A, Regina M, Aland A. 2007. Proceedings of the 13th International Congress in Animal Hygiene, Tartu, Estonia, 2007 June 17–21, Vol. 2. The cumulation of selected chemical elements of toxic properties in bee honey originating from the industrial and rural-forest areas. Estonian Univ. of Life Sciences, Jõgeva Plant Breeding Inst., Estonian Research Inst. of Agriculture, p 877–81.
- 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* 53:256–62.
- Ruschioni S, Riolo P, Minuz RL, Stefano M, Cannella M, Porrini C, Isidoro N. 2013. Biomonitoring with honeybees of heavy metals and pesticides in nature reserves of the Marche region (Italy). *Biol Trace Elem Res* 154:226–33.
- Saghaei S, Ekici H, Demirbas M, Yarsan E, Tumer I. 2012. Determination of the metal contents of honey samples from Orumieh in Iran. *Kafkas Univ Vet Fak Derg* 18:281–4.
- Salas J, Echávarri J, Negueruela A. 1993. Influencia de la temperatura en la medida del color de la miel. *Óptica pura y aplicada* 26:549–58.
- Samimi A, Maymand OE, Mehratababaei M. 2001. Determination of cadmium and arsenic pollution by bee honey based on the study on Ja'far abad area from Saveh city from Iran. *Water Geosci* 199–2. Available from: <http://www.wseas.us/e-library/conferences/2010/Cambridge/WHGE/WHGE-32.pdf>.
- Sanna G, Pilo MI, Piu PC, Tapparo A, Seeber R. 2000. Determination of heavy metals in honey by anodic stripping voltammetry at microelectrodes. *Anal Chim Acta* 415:165–73.
- Santos JSD, Santos NSD, Santos MLPD, Santos SND, Lacerda JDDJ. 2008. Honey classification from semi-arid, Atlantic and transitional forest zones in Bahia, Brazil. *J Brazilian Chem Soc* 19:502–8.
- Saxena S, Gautam S, Sharma A. 2010. Physical, biochemical and antioxidant properties of some Indian honeys. *Food Chem* 118:391–7.
- Silici S, Uluozlu OD, Tuzen M, Soylak M. 2008. Assessment of trace element levels in rhododendron honeys of the Black Sea region, Turkey. *J Hazard Mater* 156:612–8.
- Silici S, Uluozlu OD, Tuzen M, Soylak M. 2013. Honeybee and honey as monitors for heavy metal contamination near the thermal power plants in Mugla, Turkey. *Toxicol Industrial Health*. In press. 1–10.
- Silva LR, Videira R, Monteiro AP, Valentão P, Andrade PB. 2009. Honey from Luso region (Portugal): physicochemical characteristics and mineral contents. *Microchem J* 93:73–7.
- Singh N, Bath PK. 1997. Quality evaluation of different types of Indian honey. *Food Chem* 58:129–33.
- Sobotka L, Allison S, Stanga Z. 2008. Basics in clinical nutrition: water and electrolytes in health and disease. *E Spen Eur E J Clin Nutr Metab* 3:e259–66.
- Stankovska E, Stafilov T, Šajin R. 2008. Monitoring of trace elements in honey from the Republic of Macedonia by atomic absorption spectrometry. *Environ Monit Assess* 142:117–26.
- Street R, Kulkarni M, Stirk W, Southway C, Abdillahi H, Chinsamy M, Van Staden J. 2009. Effect of cadmium uptake and accumulation on growth and antibacterial activity of *Merwillia plumbea*—An extensively used medicinal plant in South Africa. *S Afr J Bot* 75:611–6.

- Surai PF. 2003. Selenium–vitamin E interactions: does 1+1 equal more than 2. In: Lyons TP, Jacques KA, editors. Nutritional biotechnology in the feed and food industries. Nottingham, UK: Nottingham University Press. p 51–8.
- Swaileh KM, Abdulkhaliq A. 2013. Analysis of aflatoxins, caffeine, nicotine and heavy metals in Palestinian multifloral honey from different geographic regions. *J Sci Food Agric* 93:2116–20.
- Taha E, Ali M. 2012. Determination of heavy metals content in cotton honey in Kafr Elshiekh Province, Egypt. *J Plant Protec Pathol* 3:1211–9.
- Taha EA, El-Sanat SY. 2007. Proceedings of the 2nd International Conference of the Entomological Society of Egypt. Effect of combs age on honey production and its physical and chemical properties; Vol. 11, p 9–18.
- Taha E-KA, Manosur HM, Shower MB. 2010. The relationship between comb age and the amounts of mineral elements in honey and wax. *J Apic Res* 49:202–7.
- Terrab A, Gonzalez AG, Díez MJ, Heredia FJ. 2003a. Mineral content and electrical conductivity of the honeys produced in northwest Morocco and their contribution to the characterisation of unifloral honeys. *J Sci Food Agric* 83:637–43.
- Terrab A, González AG, Díez MJ, Heredia FJ. 2003b. Characterisation of Moroccan unifloral honeys using multivariate analysis. *Eur Food Res Technol* 218:88–95.
- Terrab A, Hernanz D, Heredia FJ. 2004a. Inductively coupled plasma optical emission spectrometric determination of minerals in thyme honeys and their contribution to geographical discrimination. *J Agric Food Chem* 52:3441–5.
- Terrab A, Escudero ML, González-Miret ML, Heredia FJ. 2004b. Colour characteristics of honeys as influenced by pollen grain content: a multivariate study. *J Sci Food Agric* 84:380–6.
- Theobald H. 2005. Dietary calcium and health. *Nutr Bull* 30:237–77.
- Tuzen M, Soylak M. 2005. Trace heavy metal levels in microwave digested honey samples from middle Anatolia, Turkey. *J Food Drug Anal* 13: 343–47.
- Tuzen M, Soylak M. 2009. Column solid-phase extraction of nickel and silver in environmental samples prior to their flame atomic absorption spectrometric determinations. *J Hazard Mater* 164:1428–32.
- Tuzen M, Silici S, Mendil D, Soylak M. 2007. Trace element levels in honeys from different regions of Turkey. *Food Chem* 103:325–30.
- Üren A, Şerifoğlu A, Sarıkahya Y. 1998. Distribution of elements in honeys and effect of a thermoelectric power plant on the element contents. *Food Chem* 61:185–90.
- USDA. 1985. United states standards for grades of extracted honey. May 23. Washington, DC: USDA, Agricultural Marketing Service.
- USDA. 2015. United States National Nutrient Database for Standard Reference, Release 28. U.S. Dept. of Agriculture, Agricultural Research Service; September 2015. Nutrient Data Laboratory. Available from: <http://www.ars.usda.gov/ba/bhnrc/ndl>. Accessed 2015 January 20.
- Vanhanen LP, Emmertz A, Savage GP. 2011. Mineral analysis of mono-floral New Zealand honey. *Food Chem* 128:236–40.
- Vela L, de Lorenzo C, Perez RA. 2007. Antioxidant capacity of Spanish honeys and its correlation with polyphenol content and other physicochemical properties. *J Sci Food Agric* 87:1069–75.
- Vinas P, López-García I, Lanzón M, Hernández-Córdoba M. 1997. Direct determination of lead, cadmium, zinc, and copper in honey by electrothermal atomic absorption spectrometry using hydrogen peroxide as a matrix modifier. *J Agric Food Chem* 45:3952–6.
- Vincēviča-Gaile Z. 2010. Macro- and trace elements in honey. *LLU-Raksti* 25:54–66.
- Vit P, Rodríguez-Malaver A, Rondón C, González I, Di Bernardo ML, García MY. 2010. Bioactive indicators related to bioelements of eight unifloral honeys. *Arch Latinoam Nutr* 60:405–10.
- White J, Doner LW. 1980. Honey composition and properties. *Beekeeping in the US Agriculture. USA Handbook* 335:82–91.
- Yaghoobi N, Al-Waili N, Ghayour-Mobarhan M, Parizadeh S, Abasalti Z, Yaghoobi Z, Yaghoobi F, Esmaili H, Kazemi-Bajestani S, Aghasizadeh R. 2008. Natural honey and cardiovascular risk factors; effects on blood glucose, cholesterol, triacylglycerols, CRP, and body weight compared with sucrose. *Scientific World J* 8:463–9.
- Yarsan E, Karacal F, Ibrahim I, Dikmen B, Koksall A, Das Y. 2007. Contents of some metals in honeys from different regions in Turkey. *Bull Environ Contam Toxicol* 79:255–8.
- Zugravu C-A, Parvu M, Patrascu D, Stoian A. 2009. Correlations between lead and cadmium pollution of honey and environmental heavy metal presence in two Romanian counties. *Bull UASVM Agric* 66:230–3.