Assessment of mineral composition and health status of five honey samples from southern Nigeria: drifting towards food quality control

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Abstract

Mineral composition, interaction and safety index of five honey samples from southern Nigeria was investigated. Agilent 720 ICP-OES was used for the determination of mineral element concentrations (mg/l). Honey sample from Akure was highest in concentration (570.06 mg/l) while Ijala-Ikeren was lowest (90.25 mg/l). The mean and ranges for minerals were: Akure; 27.15 (0.35–277.14), Ogunmakin; 13.13 (0.26–91.81), Ibadan; 9.05 (0.42–114.53), Agbor; 5.50 (0.01–58.02) and Ijala-Ikeren; 4.11 (0.14–39.81). The coefficient of variance ranged from 12.48–180.27% revealing variations in concentration. The most abundant mineral elements were Ca, 116.26 (39.82–277.14); Mg, 41.05 (3.43–173.05); Na, 33.67 (11.09–70.59); K, 30.08 (11.43–75.14); Zn, 4.60 (0.58–14.04); Al, 3.89 (1.05–8.03); U, 3.17 (2.11–4.53) and Fe, 2.85 (1.88–3.52). Heavy metals like Fe, Ba, Pb, Cr, Ni, Ag, As, Cu, Mn and Cd were above maximum permissible limits. The mineral ratio for Zn/Cu, Fe/Cu, Fe/Pb, and Zn/Cd indicated possible unhealthy interaction. The K/[Ca + Mg] values revealed potential hypomagnesaemic effect if continually consumed. The safety indices were all within recommended range except for Se. The principal component plot showed no particular mineral distribution pattern. Thallium had the highest hazard quotient (2.00–50.00) while the hazard indices were between 3.24 and 53.97 showing potential non-carcinogenic effect. The presence of essential trace and major minerals revealed potential usefulness of the honeys as food supplement; however, the heavy metals presence resonate the need for quality control, food safety and health risk assessment before open marketing.

Introduction

Honey is a sweet natural food substance produced by bees after foraging on brightly coloured and/or beautifully scented polliniferous and/or nectariferous flowers. They also sometimes collect exudates from succulent parts (leaves, trunk, stem or fruits) of plants (living or dead) as well as enzymatic excretions from plant-sucking insects (Kadri et al., 2017). These collections are then mixed and digested with intestinal enzymes from the honeybees for producing honey thus conferring numerous health or pharmaceutical benefits and functions. Honey is medicinal, therapeutic, economically valued, nutritional and widely used for numerous purposes, for examples remedy for curing cough, cold, treatment of wounds and respiratory tract infections (Haridy, 2020). It also has antimicrobial, antioxidative and anti-inflammatory properties (Adeonipekun et al., 2016). Depending on the botanical and geographical source, the chemical and biological composition of honey may vary in quality and quantity (Bogdanov et al., 2008). Biochemically, honey is made up of a complex mixture of carbohydrates, proteins, enzymes, amino acids (Kaur et al., 2015), vitamins, volatile chemicals (Pattamayutanon et al., 2017), organic acids, flavonoids and minerals (Wang & Li, 2011). Mineral elements are minor but essential components of honey as they can cause adverse and toxic health effect when consumed at high level/concentration (Aghamirlou et al., 2015).

The mineral content may also vary depending on plant source or location. These differences observed in the quantity and quality of minerals, chemical compounds and nutrients can also be a function of the flowering seasonality, number of plants foraged, nectar type, pollen chemistry, adulteration and environmental factors like pollution and contamination. This means, that although, honey is widely consumed as natural food having essential trace elements and minerals, it could as well as contain toxic metals. Heavy metals and other contaminants in honey have been reported (Gebremariam & Brhane, 2014) with suspected acts of adulterations while, Adugna et al. (2020) reported that the presence of trace and heavy metals in honey could be due to environmental pollution.

There are few other possibilities for the introduction of trace and heavy metals into honeys. The use of pesticides in farmlands can contribute to the presence of pesticides in honeys (El-Nahhal, 2020). Trace or heavy metals can be absorbed by plants from the soil or water, taken up through the use of pesticides near the apiary (Aghamirlou et al., 2015). The presence of toxic metals in honey have been reported for honeys from Ethiopia (Adugna et al., 2020), Romania (Oroian et al., 2016; Bartha et al., 2020), Turkey (Erbilir & Erdogrul 2005), China (He et al., 2013), Iran (Aghamirlou et al., 2015; Sobhanardakani & Kianpour, 2016), Pakistan (El-Nahhal, 2020; Yaqub et al., 2020) and results showed concentrations above the recommended limits. These recurrent observations call for urgent assessment of honey mineral quality in many developing countries including Nigeria.

Apart from finding out the presence of heavy metals and essential minerals, the bioavailability, and interaction between minerals is important. The nutritional relationship that exists between mineral elements is best determined through mineral ratios and this is because physiological factors that influences metabolism and interaction of minerals in the body can be deciphered accordingly (Gemede, 2020). These ratios are more useful in interpreting the nutritional quality of a food than the individual minerals.) Mineral ratios provide more information about the richness of a food supplement and how the diet can be related to its health outcomes (Owen et al., 2018). Since the minerals are not consumed in combined form in diets, mineral ratio analysis can predict if the minerals aligned to recommended optimal physiological conditions for nutrient interactions. These minerals multi-task during metabolism in tissues and do not target one tissue type, therefore, comparing one mineral over another is an important determinant of the health quality of food and in this research, honey.

Mineral bioavailability and accessibility in food substances can ameliorate the effect of malnutrition from chronic deficiency of micronutrients which affect growth, productivity and performance of people (WHO, 2012; Drago, 2017). Mineral elements are important components of human diets as they play vital roles in the moderating the health status of people. For example, a recommended consumption of Ca enhances bone growth, muscle contraction, and blood clotting, Cr helps to control blood sugar, Mg for nerve and muscle functions (WHO, 2004), Mn and Mo for enzyme functioning, K helps in dissolving kidney stone and controls blood pressure, Se helps thyroid hormone production, protects cells from damage, Na helps fluid balance while Zn is useful in boosting human immune system (Felson, 2020). Zinc plays a beneficial role in tissue repairs and immunity of cells, but at higher intake concentrations of 500 mg it is considered to be toxic (Drago, 2017). Its deficiency can lead to an increased risk of getting easily infected. Iron is essential component for cytochromes, enzymes metabolism, haemoglobin and oxygen transport in the blood.

The bioavailability of Iron can be influenced by its interaction with Ca when an insoluble polymineral ligand involving Fe is formed, thus Fe/Ca interaction is important (Drago, 2017). Despite the presence of minerals, low or excess bioavailability for absorption within intestinal tissues is modulated by mineral interaction (synergistic or antagonistic) within the matrix or during ingestion and the most significant interactions are Na/K, Ca/Mg, Mn/Fe, Fe/Cu, and Zn/Cu (O'dell, 1989). Hence, quantifying the presence in honey samples is pertinent to public health and quality control. Although, Zn, Cu, Mn, and Cr are useful, above permissible limits, they could have toxic effects (Ashenef, 2014). Therefore, a comprehensive analysis on quality and quantity of mineral element are needed. Many locals indulge in adulteration since they believe it boosts financial gains from sales, without considering the health implications.

In recent years, adulteration of honey has become a norm, and there is need for quality control. El-Nahhy (2020) study showed that most African countries do not have relevant information on honey quality control though efforts are now been geared towards providing relevant information on quality control. Only a few countries (like Ethiopia, Ghana and Kenya) have reputable work on contaminants and nutritional composition in honey. To our knowledge, very little or no empirical research on the elemental composition of honey has been carried out in Nigeria, and only recently, Obasi et al. (2020) reported the presence of heavy metals in apiary honeys from south-eastern Nigeria with Arsenic and Lead levels above recommended range in some of the samples posing potential health risk for children. Odoh et al. (2015) report also covered the middle belt region of Nigeria (Taraba, Nassarawa, FCT and Abuja) confirming the presence of honeys from that region.

There are indications that in southern Nigeria, most of the open market honeys are yet to be assessed of its quality parameters by regulatory bodies. This region accommodates over 90 million people who consume different food products (natural and synthetic) including honey with no detailed nutritional value label. Understanding the concentration of minerals in honey could shed light on the nutritional status and levels of environmental pollution in the location where the honey was produced (Pohl, 2009). It has been confirmed that high concentration of heavy metals could be related to adulteration (Odoh et al., 2015). This paper tries to conduit this perceived gap by attempting to add to the pool of data on honey quality assessment in Africa. The study objectively attempts to know the concentration of minerals in honey, find out the mineral safety indices for major mineral elements, determine the mineral ratio for ascertaining their contribution to consumers health, mineral bioavailability and macronutrient overdose, know if there are significant difference in the composition of mineral elements in the honeys and find out if there is any uniformity in the distribution of mineral elements based on locations.

Materials And Methods

Collection of samples

Five honey samples were randomly purchased from the four states and five different apiaries from Akure (Ondo State), Agbor and Ijala-Ikeren (Delta State), Ibadan (Oyo State) and Ogunmakin (Ogun State) in the southern part of Nigeria between 23rd January and 4th July, 2020. The Ijala-Ikeren honey was sourced from an apiary in Ondo State according to vendor. The vegetation types of the precise location of the apiaries were not ascertained, only the ecozone was established. The samples were preserved in a refrigerator in a properly labelled air-tight plastic container before taken to the Laboratory (Department of Biochemistry, College of Medicine, Lagos University Teaching Hospital, Idi-Araba, Surulere) for analysis. The acronyms – AKR (Akure), AGB (Agbor), IJK (Ijala-Ikeren), IBD (Ibadan) and OGM (Ogunmakin) were used to represent the samples respectively.

Mineral element determination

The identification and quantification of elements in the honey samples was carried out using the Agilent 720 Inductively Coupled Plasma - Optical Emission Spectrometer (ICP-OES) (Agilent Technologies Inc., Santa Clara, California, U.S.A) due to its accuracy in quantifying elements in food supplements (Aghanirlou *et al.* 2015; Sobhanardakani & Kianpour, 2016; Kilic-Altun *et al.*, 2017).

Mineral ratio and safety index

Mineral ratios and milliequivalent (K/(Ca + Mg) were calculated (Jacob et al. 2015; Adeyeye & Olaleye, 2016; Ajala et al. 2019). The mineral safety indices were calculated according to Adeyeye et al. (2012) and Adeyeye and Olaleye (2016). There are currently no mineral safety indices for K, Mn, Co and Pb (Adeyeye & Omolara, 2018), and were not calculated. Ideal range for mineral safety index and ratio were in accordance with Adeyeye & Omolara (2018), Oluwagbenle et al. (2019).

Mineral ratio = Mineral A/Mineral B, while

Mineral Safety Index (MSI) of each mineral =

$$\frac{\mathrm{MSIstandard}}{\mathrm{RequiredAverageIntake}}\mathrm{xConcentrationofindividualmineralelement}$$

Health Status Assessment of Honey

In assessing the health status of the honeys, the chronic daily intake (CDI), hazard quotient (HQ) and hazard index (HI) were calculated (Onyele & Anyanwu, 2018; Duru & Duru, 2021). The values were compared to standards set (USEPA, 2011; Elumalai et al., 2017; Moslen & Miebaka, 2017; Uche et al., 2017). The CDI is daily dose of heavy metal consumers may be exposed to. HQ is the non-carcinogenic effect of each of the heavy metals. Hazard index is the summation of HQ for each heavy metals considered as a representation of the overall toxic risk. HQ and HI > 1.0 poses health concerns.

$$\text{CDI} = \frac{\text{CxIRxEFxED}}{\text{BwxAT}} \text{andH}Q = \frac{\text{CDI}}{\text{RfD}}$$

Where; C = concentration of heavy metals, IR = ingestion rate (L/day) (Adult – 2, Children – 1), EF = exposure frequency (365 days/year), ED = exposure duration in years (adults – 30, children – 6) Bw = body weight (adults – 70 kg, children – 15 kg), AT = average time in days (adults − 10950, children − 2190) and RfD = oral toxicity reference dose. Since the CDI is in mg/kg, the concentrations of the trace metals were converted to mg/kg from mg/l using the specific gravities and volume of the honeys.

Specific gravity = Density of object (kg/l)/Density of water (kg/l)

Conc. of minerals (in mg/kg) = Conc. of minerals (mg/l)/Density of honey (kg/l)

Data analysis

Data was subjected to descriptive and inferential analysis. Calculated valued were compared with standardised values (USEPA, 2011; FAO 1997; WHO 2012; Hathcock, 2014). The percentage coefficient of variance (CV%) was used to ascertain the degree of variation of the standard deviation from the mean concentration of mineral element (Adeyeye & Omolara, 2018). Principal component plot (factorial analysis) used to determine the similarity in distribution of the metals in the honeys. Students' t-test was used to ascertain the level of difference in the concentration of metals across different locations at 0.05 level of significance. The analyses were performed using IBM SPSS version 21 (SPSS Inc., Chicago, IL, USA) and Microsoft Excel 2019.

Results And Discussion

The results for this study are presented in Figs. 1–3 and Tables 1–5 below. Figure 1 shows the total amount of mineral elements in the honey samples collected from the five different locations. Figure 1 showed that AKR honey had the highest (570.06 ± 66.07 mg/l) concentration of honey while IJK had the lowest (90.25 ± 8.55 mg/l) concentration.

Figure 2 shows the concentration of the individual mineral elements in the five honey samples. The values in Fig. 2 revealed that Ca was the most abundant in all samples followed by Mg, Na, K, and Al while, Table 1 shows the mean, standard deviation and range for the quantity of each of the mineral elements. Table 1 revealed that the mean and range for most abundant mineral were Ca, 116.26 (39.82–277.14) mg/l; Mg, 41.05 (3.43–173.05) mg/l; Na, 33.67 (11.09–70.59) mg/l; K, 30.08 (1143–75.14) mg/l; Zn, 4.60 (0.58–14.04) mg/l; Al, 3.89 (1.05–8.03) mg/l; U, 3.17 (2.11–4.53) mg/l and Fe; 2.85 (1.88–3.52) mg/l while, others were found in very minute quantities. Cobalt and Cu were detected in all samples but in insignificant and negligible quantity, however, in some of the samples, heavy metals (mg/l) like Se (0.34) in IJK and OGM and Zn (5.09 and 14.04 mg/l) in AKR and AGB respectively were above the permissible limit along with Ba, Pb, Cr, Ni, Ag, As, Cu, Cd and Mn. The mean and range (mg/l) for minerals showed Akure, 27.15 (0.35–277.14); Ogunmakin, 13.13 (0.26–91.81); Ibadan, 9.05 (0.42–114.53); Agbor, 5.50 (0.01–58.02) and Ijala-Ikeren, 4.11 (0.14–39.81). The ranges for CV% for all mineral elements were between 12.48% and 180.27%.

The PCA plot in Fig. 3 showed the relationship among the mineral elements from the region of sampling. The data showed no particular relationship or distribution pattern. Although, there were obvious clusters e.g., Ag, Tl, Th and Se; Be, Cr, and As; Zn, Fe, Mg, Ca and Ba; Na, Mn and Cu. Elements like Cd, V, Pb, Ni, U, Al, Co and K were found scattered around in separate points.

Table 2 shows the mineral interaction and the milliequivalent. It revealed that the ratios for Ca/Na, Ca/K, Na/Mg, Zn/Fe, K/Co, Mn/Fe, Zn/Cd, Fe/Zn were within recommended limits. Zn/Cu (3.4) was below the lower limits (4.0) indicating limitations in the bioavailability of Cu Fe/Co (13.71) was below the standard average of 440, which shows the antagonistic interaction of Fe over Co. The milliequivalent (0.29) was below the average (2.2). Fe/Cu (3.02) was above the limits (1.6). AKR honey had a significantly high ratio (236.88) while the average was within range. Iron/Pb (2.22) was below lower limits of 4.0. For mineral safety index, Se (25.60) was above the recommended value of 14 while others (Na, Mg, Ca, Fe, Zn, Se and Cu) were below set limits (Table 3). The t-values showed that there was no significant difference among the safety indices of Mg ($p = 0.282$), Ca ($p = 0.052$), Zn ($p = 0.139$), and Se ($p = 0.170$) while, difference was significant for Na ($p = 0.038$), Fe ($p = 0.001$) and Cu ($p = 0.004$) (Table 3).

Table 4 shows the chronic daily intake level of the trace metals in the five honey samples. When compared with the set standards for oral reference toxicity dose, As in AKR (children) and IJK (adults and children) were 0.0003, 0.0003 and 0.0006 mg/kg respectively. Selenium for AGB adults (0.0093 mg/kg) was also higher. The CDI for Thallium for all samples for both adult and children had toxic dose. For U, only AKR adult (0.0002) had toxic oral dosage. Table 5.0 shows the hazard quotient of the trace metals in the five honey samples. Usually the limit of < 1.0, the result revealed that As (1.0) in AKR honey was above the limit. Similarly, As in IJK honey for children (1.0) and adults (2.0), and Se in AGB for children (1.86) were also above the limits. Thallium in all samples was significantly above the limits ranging from 2.0 through 20.0 to 50.0. Uranium in AKR was also above limits (1.0) for adults. The overall hazard indices for children were higher than that of adults and it ranged from 3.24 to 53.97.

Discussion

The overall concentration of mineral elements showed that the location where the honeys were produced affected the presence and concentration of minerals. The honey from Akure (Fig. 1), a lowland rainforest zone had the highest, followed by those from rainforest/guinea savanna mosaic regions (IBD and OGM) and IJK was least. It is insufficient however to conclude that the ecozone have direct effect on the mineral (ash) content of honey based on the number of samples examined even though there was observed differences for geographical locations. But it is safe to say honeys from same geographical region may have close similarity in mineral content while the slight differences could be due to the diversity of foraged botanicals (pollen and nectar) within the ecozone which correlates with the assertion of Bogdanov et al. (2008). The most abundant minerals were Ca, Mg, Na, K, Zn, Fe, and Al in all honeys (Fig. 2) showed that the honeys are good for skeletal/nerve/muscle function, kidney, boosting immune, bone formation and for blood related metabolic process (WHO, 2004; Felson, 2020). Calcium, magnesium, sodium and potassium were found to be most abundant. This is slightly in conformity with Olaitan et al. (2007) report which says K is usually the most abundant mineral in honey along with Ca, Cu, Mn and Fe in good quantities. The variation may be due to the limited number of samples for this study used or location where they were sourced in comparison with that of Olaitan et al. (2007).

The uniform occurrence of heavy metals like As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn indicated that the honeys were polyfloral (Oroian et al. 2016). The availability of most of the essential minerals showed that the honeys are rich in nutrients. Copper and Iron are essential components for the energy metabolism of cytochrome oxidase enzyme, while Co prevents anaemia. Iron can be sourced from plants like unripe plantain in meals and also prevents anaemia. In addition, minerals from honey serve as food supplement and their bioavailability in the body. Magnesium is an enzyme activator while, Na helps to normalise pH and K acts as cation intercellularly (Fig. 2). Zinc is found in almost all tissues in humans and a component of many enzymes, Mn is useful for nerve functions, reproductive health and bone formation. Zn helps to reduce the circulation of HDL high density lipoproteins which causes heart disease. When the honeys are consumed, Ca and P can help to avoid osteomalacia and rickets in children, making honey important for children.

Fe can support carriage of oxygen from lungs to the tissues once it found in the blood stream. The presence of Ag, Co, Ba, Cr, Pb, Ni and Zn could be a source of concern even though they are in small quantities. These minerals have been classified as heavy metals and could be toxic following continuous consumption.

Table 1
Mineral composition and concentrations (mg/l) in the five honey samples from Nigeria

Mineral		Wavelength (nm)	AGB	AKR	IJK	IBD	OGM	Mean	σ	CV%	Min	Max	MPL for water
Ca	Calcium	396.847	58.02	277.14	39.81	114.53	91.81	116.26	94.50	81.28*	39.81	277.14	1500
Mg	Magnesium	279.553	4.42	173.05	3.43	7.16	17.17	41.05	73.99	180.27**	3.43	173.05	300
Na	Sodium	589.592	17.52	47.53	11.10	21.64	70.59	33.67	24.85	73.79*	11.09	70.59	1500
K	Potassium	766.491	11.43	29.33	14.47	20.01	75.14	30.08	26.10	86.77*	11.43	75.14	4700
Zn	Zinc	213.857	5.09	14.04	0.58	0.75	2.52	4.60	5.58	121.48**	0.58	14.04	15
Al	Aluminium	396.152	2.28	8.03	1.05	2.68	5.43	3.89	2.81	72.23*	1.05	8.03	10
U	Uranium	385.957	4.11	2.22	2.11	2.85	4.53	3.17	1.10	34.80	2.11	4.53	0.03
Fe	Iron	238.204	2.88	3.44	1.88	3.52	2.55	2.85	0.68	23.70	1.88	3.52	15
Ba	Barium	455.403	2.05	3.45	2.10	3.19	2.36	2.63	0.65	24.79	2.05	3.45	1.8
Pb	Lead	220.353	0.55	1.17	1.48	3.80	3.56	2.11	1.47	69.65*	0.55	3.80	0.3
Cr	Chromium	267.716`	2.07	1.82	1.36	1.48	2.12	1.77	0.34	19.28	1.36	2.12	0.05
Th	Thorium	283.730	1.62	1.27	1.84	0.79	1.52	1.41	0.40	28.59	0.79	1.84	0.3
Ni	Nickel	231.604	2.15	0.53	1.02	1.21	1.64	1.31	0.62	47.01	0.53	2.15	0.09
Ag	Silver	328.068	1.52	1.06	1.32	1.32	1.28	1.30	0.16	12.48	1.06	1.52	0.007
Tl	Thallium	190.794	1.86	0.88	1.56	0.55	1.11	1.19	0.53	43.97	0.55	1.86	0.0002
Cu	Copper	327.395	1.30	1.55	0.74	0.57	1.32	1.09	0.42	38.31	0.57	1.55	1
Mn	Manganese	257.610	0.96	1.19	0.68	0.76	1.27	0.97	0.26	26.66	0.68	1.19	0.4
Co	Cobalt	238.892	0.03	0.89	0.41	1.50	1.42	0.85	0.63	74.64*	0.03	1.50	0.01
V	Vanadium	292.401	0.31	0.72	1.00	0.75	0.37	0.63	0.29	46.06	0.31	1.00	-
Cd	Cadmium	214.439	0.38	0.46	1.89	0.51	-0.16□	0.62	0.76	123.88**	-0.16*	1.89	0.003
As	Arsenic	188.980	0.50	0.35	0.14	0.42	0.54	0.39	0.16	40.05	0.14	0.54	0.01
Se	Selenium	196.026	0.01	-0.01□	0.34	-0.04□	0.34	0.13	0.19	152.78**	-0.04*	0.34	0.035
Be	Beryllium	313.042	-0.05□	-0.05□	-0.04□	-0.05□	0.26	0.02	0.14	878.27**	-0.05*	0.26	-
	Mean		5.50	27.15	4.11	9.05	13.13			48.07			
	σ		12.42	68.81	8.70	24.88	27.31			41.12			
	Range		0.01–58.02	0.35-277.14	0.14–39.81	0.42-114.53	0.26–91.81			12.48-180.27			

□BDL = below detection limits, **σ** = standard deviation, Coefficient of variance (CV)%: low variance (1% – 50%), * = moderate to high variance (51% – 100%), ** = very high variance (101% and above). Maximum permissible limits (MPL) (FAO, 1997; WHO, 2012; Hathcock, 2014; USEPA, 2015). Results are arranged in decreasing order of abundance.

Table 2
Mineral interactions/ratio in the honeys

Ratios	AGB	AKR	IJK	IBD	OGM	Mean	σ	CV%	t-value	Sig.	Range	Standard ratio (range)
Ca/Mg	13.13	1.6	11.61	15.99	5.34	9.53	5.9	61.93*	3.61	0.023	14.35	7.0 (3.0–11.00)
Na/K	1.53	1.62	0.77	1.08	0.94	1.19	0.37	31.25	7.154	0.002	0.85	2.40 (1.4–3.4)
Ca/Na	3.31	5.83	3.59	5.29	1.3	3.86	1.79	46.39	4.82	0.009	4.53	-
Ca/K	5.08	9.44	2.75	5.72	1.22	4.84	3.14	64.87*	3.447	0.026	8.22	4.20 (2.2–6.2)
Na/Mg	3.96	0.28	3.24	3.02	4.11	2.92	1.55	53.08*	4.222	0.013	3.83	4.0 (2.0–6.0)
Zn/Cu	3.92	9.06	0.78	1.32	1.91	3.4	3.38	99.49*	2.248	0.088	8.28	8.0 (4.0–12.0)
Zn/Fe	1.77	4.08	0.31	0.21	0.99	1.47	1.59	107.77**	2.075	0.019	3.87	2.0 (0.8–3.5)
Fe/Cu	2.22	2.22	2.54	6.18	1.93	3.02	1.78	59.00*	3.79	0.019	4.25	0.90 (0.2–1.6)
Ca/Pb	105.49	236.88	26.90	30.14	25.79	100.28	145.17	144.76**	2.082	0.106	211.09	126 (84–168)
Fe/Pb	5.24	2.94	1.27	0.93	0.72	2.22	1.90	86.37*	2.611	0.059	4.52	4.40 (4.0–8.8)
Mn/Fe	1.08	0.33	0.35	0.22	0.50	0.50	0.34	68*	1.861	0.134	0.86	-
K/Co	381	32.96	35.29	13.34	52.92	11.93	15.15	126.97**	1.478	0.213	367.66	2000 (NA)
Zn/Cd	13.39	30.52	0.31	1.47	-	9.05	11.32	125.05**	1.550	0.196	30.21	500 (450–1000)
Fe/Co	96	3.87	4.59	2.35	1.80	13.71	21.65	157.94**	1.169	0.307	94.2	440 (NA)
Fe/Zn	0.57	0.25	3.24	6.07	1.01	2.23	2.45	109.86**	2.037	0.111	5.82	2.0 (1.0–3.0)
K/[Ca + Mg]	0.18	0.07	0.34	0.16	0.69	0.29	0.24	85.03*	2.63	0.058	0.62	2.2 (NA)

NA = not available, CV%: low variance (1% – 50%), * = moderate to high variance (51% – 100%), ** = high variance (above 100%).

Table 3
Mineral safety index of the honey samples

Honeys	Na	Mg	Ca	Fe	Zn	Se	Cu
AGB	0.17	0.17	0.48	1.29	11.20	2.00	14.30
AKR	0.46	6.49	2.30	1.54	30.89	NA	17.05
IJK	0.11	0.13	0.33	0.84	1.28	68.00	8.14
IBD	0.21	0.27	0.95	1.57	1.65	NA	6.27
OGM	0.68	0.64	0.76	1.14	5.54	68.00	14.52
Mean ± σ	0.32 ± 0.24	1.54 ± 2.77	0.96 ± 0.78	1.27 ± 0.30	10.11 ± 12.28	**25.60** ± **38.87**	12.06 ± 4.61
Range	0.11–0.68	0.13 – 6.49	0.48 – 2.3	0.84 – 1.57	1.28 – 30.89	2.0–68.0	6.27 – 17.05
CV%	73.79*	180.27**	81.28*	23.69	121.47**	151.83**	38.20
t-value	3.055	1.241	2.747	9.446	1.841	1.673	5.853
sig.	0.038	0.282	0.052	0.001	0.139	0.170	0.004
MSI standard	4.8	15	10	6.7	33	14	33
RAI	500	400	1200	15	15	0.07	3

NA = not available, CV%: low variance (1% – 49%), * = moderate to high variance (50% – 100%), ** = very high variance (above 100%), MSI = mineral safety index, RAI = recommended average intake.

The cause of this presence is yet to be known, however, Aghamirlou et al. (2015) reported that the closeness of an apiary to an industry or pesticide sprayed farm is one factor responsible. Table 1 showed that there were very high variations in the distribution and concentration of the mineral elements as seen from their coefficient of variance. Invariably, no particular pattern of distribution or concentration of the minerals was observed. It points to the fact that the botanical and geographical source of honey to a very large extent determined the kind of minerals that can be found in it. Despite having essential minerals, the honeys had heavy metals with concentrations above maximum permissible limits. Ba, Fe, Pb, Cr, Ni, Ag, Mn, Cd and As were above recommended limits similar to the reports of Obasi et al. (2020) in the South east, and Odoh et al. (2015) in the middle belt. This calls for further public health concern and detailed investigation. It also showed that these honeys were probably harvested or farmed near contaminated environments and this may pose food safety risk for the growing population of the south which depends largely on honey for some domestic purpose and may negatively impact on the export potentials of these Nigerian honeys.

It is important to consider mineral interactions since it determines the bioavailability of individual minerals in honey. Knowledge of these ratios helps to prevent chronic diseases. This study showed that most of the minerals interacted within recommended range apart from Zn/Cu, Fe/Cu, Fe/Pb, and Zn/Cd. Thomson *et al*. (1971) reported that Cu can have negative interaction with Iron leading to the decline in the reduced bioavailability of Cu, the K/[Ca + Mg] value showed that there may be a possible hypomagnesaemia. Adeyeye et al. (2012) reported that K (Ca + Mg) interactions help to decipher the potential of developing hypomagnesaemia especially when it is below 2.2. The IJK and OGM honey sample will be excellent for reducing blood pressure since Na/K ratio was lower than 1. The ratio of Fe/Zn or Zn/Fe was not close to one for AGB and AKR honeys which may affect the absorption of the lower one. Calcium is found in body fluids and helps to correct the excess availability of K, Mg and Na. thus Ca/K, Ca/Mg and Ca/Na ratios in the honeys make them important for human health. There was a competitive inhibition with an iron overload for IBD and IJK honeys since Fe/Zn is greater than 2.1.

The mineral safety index showed that all essential minerals except for Se are in safe concentration despite the high variance among the honeys (Table 3) and no overloading in the body if consumed. This again calls for concern on the food safety index of the honeys especially the IJK and OGM honey samples. The PCA plots re-iterated the fact that mineral elements can be distributed in no particular order in honey samples (Fig. 3) as each honey mineral contents are influenced by the source or location of plant. Clusters of elements have not provided any pointer to the possible cause; however, there seem to be some closely related mineral elements in the five samples. It was not coincidental to have found Mn and Zn to be positively correlated (Fig. 3) which was reportedly found even for ground water (Elumalai et al., 2017). The time of collection was also different hence; it may be due to seasonality in nectar production and sourcing. The honey samples were found to have low chronic daily intake (Table 4). However, Arsenic was above the recommended reference toxic dose for AKR and IJK for children and adults and also for Selenium for children who may have consumed AGB honey. Thallium CDI is of grave concern for all the honeys for both adults and children, while U was only chronic for adults for AKR honey which makes it not safe for consumption. Table 5 further expatiated that As (AKR for children, IJK for both), Se (AGB for children). Thallium (for all honeys and age group) and U (AKR for adults) may pose non-carcinogenic threat to anyone who consumes the honeys. The hazard index for all honeys revealed that the cumulative hazard quotient for all the honey was above the recommended and it is an indication of the food safety concerns of the honeys.

Table 4: CDI (mg/kg) of the five honey samples

		AGB		AKR		IJK		IBD		OGM		Mean	Stdev
	RfD	**Adult**	**Child**	**Adult**	**Child**	**Adult**	**Child**	**Adult**	**Child**	**Adult**	**Child**		
Ag	0.005	0.0003	0.0001	0.0003	0.0001	0.00003	0.0001	0.00003	0.0001	0.00003	0.0001	0.0001	0.0001
As	0.0003	0.0001	0.0002	0.0001	0.0003	0.0003	0.0006	0.0001	0.0002	0.0001	0.0002	0.0002	0.0002
Cd	0.0005	0.0001	0.0003	0.0001	0.0002	0.00002	0.0001	0.0001	0.0002	0.0000	0.000	0.0001	0.0001
Co	0.043	0.0013	0.0031	0.0004	0.0001	0.0001	0.0002	0.00003	0.0001	0.00003	0.0001	0.0005	0.0010
Cr	0.003	0.0001	0.0005	0.0002	0.0001	0.00003	0.0001	0.00003	0.0001	0.00002	0.00004	0.0001	0.0001
Cu	0.001	0.0003	0.0001	0.0002	0.0001	0.00001	0.0001	0.0001	0.0002	0.00003	0.0001	0.0001	0.0001
Mn	0.046	0.0004	0.0001	0.00003	0.0001	0.0001	0.0001	0.0001	0.0001	0.00003	0.0001	0.0001	0.0001
Ni	0.02	0.0001	0.0005	0.0001	0.0002	0.00004	0.0001	0.00003	0.0001	0.00002	0.0001	0.0001	0.0001
Pb	0.0035	0.0001	0.0002	0.00003	0.0001	0.00003	0.0001	0.00001	0.00002	0.00001	0.00003	0.0001	0.0001
Se	0.005	0.004	0.0093	0.0000	0.0000	0.0001	0.0003	0.0000	0.0000	0.00011	0.0003	0.0014	0.0030
Tl	0.00001	0.0002	0.0005	0.00004	0.0001	0.00002	0.0001	0.0001	0.0002	0.00003	0.0001	0.0001	0.0001
U	0.0002	0.0001	0.00002	0.0002	0.00004	0.00002	0.00004	0.00001	0.00003	0.00001	0.00002	0.0000	0.0001
V	0.001	0.0001	0.0003	0.0001	0.0001	0.00004	0.0001	0.0001	0.0001	0.0001	0.00023	0.0001	0.0001
Zn	0.3	0.00001	0.00002	0.000002	0.00001	0.00006	0.0002	0.0001	0.0001	0.00001	0.00003	0.0001	0.0001

Table 5: Hazard quotient and index of mineral elements in the five honey samples

	RfD	AGB Adult	AGB Child	AKR Adult	AKR Child	IJK Adult	IJK Child	IBD Adult	IBD Child	OGM Adult	OGM Child	Mean	Stdev	Min	Max
Ag	0.005	0.06	0.02	0.06	0.02	0.01	0.02	0.01	0.02	0.01	0.02	0.02	0.02	0.01	0.06
As	0.0003	0.33	0.67	0.33	1.00	1.00	2.00	0.33	0.67	0.33	0.67	0.73	0.52	0.33	2.00
Cd	0.0005	0.20	0.60	0.20	0.40	0.04	0.20	0.20	0.40	0.00	0.00	0.22	0.19	0.00	0.60
Co	0.043	0.03	0.07	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.02	0.00	0.07
Cr	0.003	0.03	0.17	0.07	0.03	0.01	0.03	0.01	0.03	0.01	0.01	0.04	0.05	0.01	0.17
Cu	0.001	0.30	0.10	0.20	0.10	0.01	0.10	0.10	0.20	0.03	0.10	0.12	0.09	0.01	0.30
Mn	0.046	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01
Ni	0.02	0.01	0.03	0.01	0.01	0.00	0.01	0.00	0.01	0.00	0.01	0.01	0.01	0.00	0.03
Pb	0.0035	0.03	0.06	0.01	0.03	0.01	0.03	0.00	0.01	0.00	0.01	0.02	0.02	0.00	0.06
Se	0.005	0.80	1.86	0.00	0.00	0.02	0.06	0.00	0.00	0.02	0.06	0.28	0.61	0.00	1.86
Tl	0.00001	20.00	50.00	4.00	10.00	2.00	10.00	10.00	20.00	3.00	10.00	13.90	14.13	2.00	20.00
U	0.0002	0.50	0.10	1.00	0.20	0.10	0.20	0.05	0.15	0.05	0.10	0.25	0.30	0.05	1.00
V	0.001	0.10	0.30	0.10	0.10	0.04	0.10	0.10	0.10	0.10	0.23	0.13	0.08	0.04	0.30
Zn	0.3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
HI	1.0	**22.40**	**53.97**	**5.98**	**11.90**	**3.24**	**12.75**	**10.81**	**21.59**	**3.55**	**11.21**	**15.74**	**14.94**	**3.24**	**53.97**

It is significant to note that this study is one of the first to show that Tl can be found in honey in quantities that is considered toxic to health and needs to be closely observed in honeys produced in Nigeria. Inclusively, the cumulative heavy metal hazard quotient of all the honeys have been found to be above the recommended 1.0 which calls for concern and intervention.

Conclusions And Recommendations

Honey is widely used for pharmaceutical or medical purpose and consumed either as food or supplement. Just like every other essential phytochemical compound, mineral elements have been found in honeys. The preliminary assessment of the mineral element composition will provide valuable insight into its safety indices and quality for human as food. This research has shown that although honey is medicinal, contains essential macro and micronutrients and widely preferred for different ethno-medicinal purpose by locals, traces of heavy metals with concentrations above the recommended limits for chronic daily intake and hazard quotient may pose health risk. Mineral interactions in the investigated honeys have shown healthy interactions except for Selenium (Se). There were also some minerals that were above maximum permissible limits. The hazard quotient and index for Tl which were above the set limit and in some cases for As, Se and U were also above the recommended RfD. Hence, there are health concerns that need immediate review.

The abundance of essential mineral elements showed that the honey samples are of good quality but was contaminated with of heavy metals with concentrations above the recommended. This made them relatively unfit for consumption and raises food safety concerns. Repeated reliable and standard quality control test needs to be adopted for other honey samples sold in the open market. Hence, there is need for quality assessment of honey samples in open markets by the appropriate food and drug regulatory bodies. The data from this study can be used for the standardisation of honey samples in southern Nigeria. Advocacy on the use of pesticides and environmental pollutants within the vicinity of apiaries should be done with caution. It has become necessary for agencies of government to be part of this quality regularisation agenda since one of its attendant effects is the introduction of heavy metals into pure honeys. It is pertinent to assess safety indices and quality for export and domestic purposes and competitive advantage.

Declarations

Originality: This study is amongst the maiden studies investigating mineral elements interaction and safety status in honey from southern Nigeria. It comprehensively investigated the presence of 23 mineral elements including trace and heavy metals and safety concern associated with pure honeys sold in the open market in some densely populated areas of southern Nigeria in an attempt to call the attention of regulatory bodies. This is the first report showing the presence of Thallium, Thorium, and Beryllium inside honeys from Nigeria.

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Figures

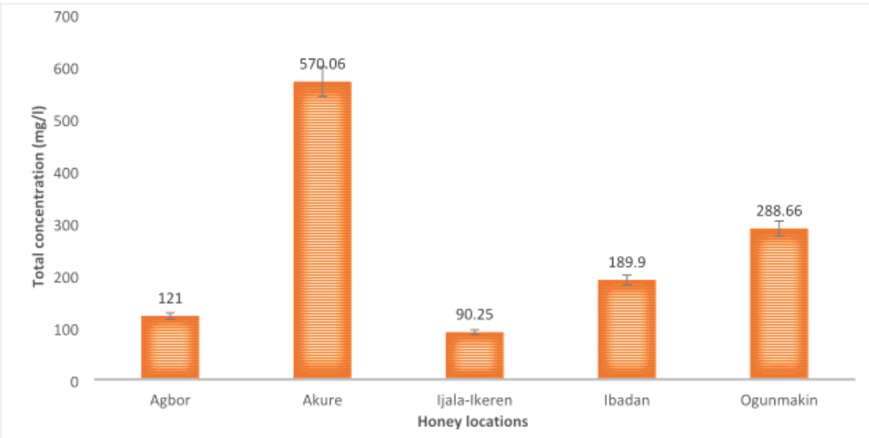

Figure 1

Total concentration (mg/l) of mineral elements in the five honey samples

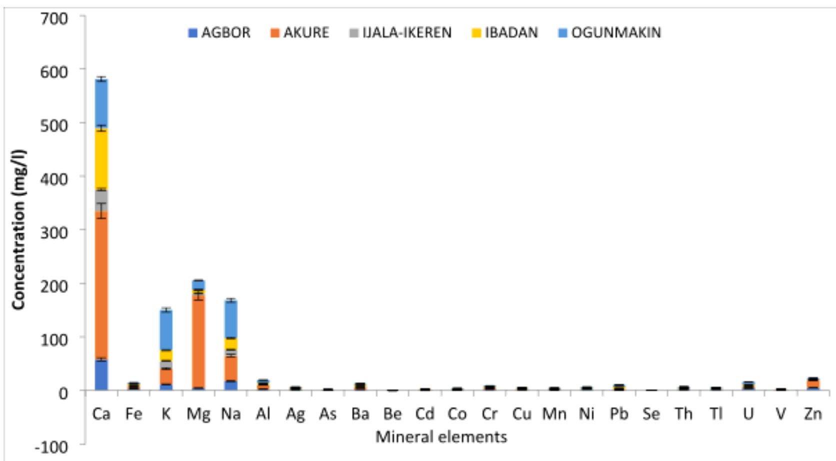

Figure 2

Aggregate concentration of mineral elements in the honey samples based on locations

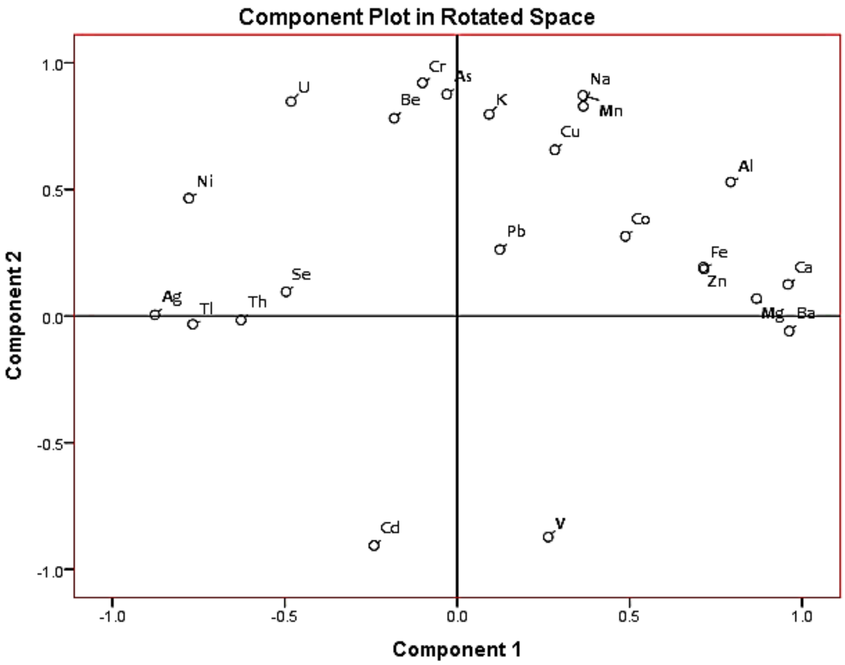

Figure 3

PCA for the relationship among mineral elements in the five honey samples