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Total Phenolic, flavonoids and some selected metal content in honey and propolis samples from South Wolo zone, Amhara region, Ethiopia

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Abstract

Ethiopia is endowed with a variety of multifloral origin of honey and propolis. However, there is paucity of information on the chemical composition of some of the regions honey and propolis. For this study, seven different type of honey and three propolis samples were collected from South Wolo zone districts of the Amhara region, Ethiopia. The total phenolic and flavonoids contents were estimated spectrophotometrically while eight metal contents were analyzed using ICP-OES. Both honey and propolis samples were found to be rich in total phenolic content expressed as gallic acid equivalent (GAE) ranged from (45.42 - 73.51) mg GAE/100 g) of honey and (204.4 - 262.5) mg GAE/100 g of propolis samples. The total flavonoids content expressed as catechin equivalent (CE) ranged from (ND - 55.73) mg CE/100 g of honey and (187.7 - 214.9) mg CE/100 g of propolis. The mineral content in the honey samples ranged from (25 - 65) µg/100 g, (10 - 113) µg/100 g, (3.75 - 17.5) µg/100 g (132.5 - 296.3) µg/100 g, (250 - 910) µg/100 g, (807.5 - 6860) µg/100 g respectively for Cr, Co, Cd, Mg, Ca and Fe However, Ni and Cu were not detected in the samples. Cd was found

below the maximum permissible limit. Thus, the honey samples collected from South Wolo zone of the Amhara region, Ethiopia are of good quality in terms of heavy metal contamination and contained relatively good composition of phenolic compounds as compared to some honey samples reported from overseas.

ABOUT THE AUTHORS

Dr Minaleshewa Atlabachew's research group is consisted of some staff members of the Bahir Dar University and postgraduate (MSc and PhD) students. Dr. Minaleshewa Atlabachew is a full time associate professor of analytical chemistry in Bahir Dar University. He graduated with B.Ed in Chemistry from Bahir Dar University, Ethiopia in 2004, M.Sc and PhD in Analytical Chemistry from Addis Ababa University in 2007 and 2013 respectively. From 2014-2016, he was a postdoctoral fellow at the Tshwane University of Technology, South Africa. Minaleshewa Atlabachew's research group spans the development of modern sample preparation techniques for bioactive molecules investigation and extensive use of the advanced analytical techniques together with multivariate data analysis for quality control of indigenous natural products. So far, he authored/co-authored more than 24 peer-reviewed original research articles.

Public Interest statement

Since honey and propolis are rich in phytochemical, their composition and bioactivity depends on the floral source, the method used to collect the nectar, seasonal and environmental factors, and geographic origin. Furthermore, honey may be useful as an environmental indicator of heavy metal pollution as honeybees may be continuously exposed to contaminants.

Even though, Ethiopia has diversified agro-ecological conditions and a variety of multifloral origin of honeys and propolis are available in the different areas of the country, their composition and quality have not been well investigated. Secondly, white honey is traditionally more valuable than colored honeys. Hence, this paper describes the dependence of phenolic compounds on the color of the honey and propolis as well as the mineral composition of some of the well knows honey samples. Thus, consumers' pharmacologists and nutritionist can now select the honey and propolis types of their interest for their consumption or concentration based studies.

Keywords: Phenolic compounds, Honey, Propolis, Flavonoids, Metal, Ethiopia

Introduction

Honey and propolis are natural substances of honeybee products which have potential role in contributing to human health (Chua et al., 2013). Honey is produced by honeybees (*Apis mellifera*) from nectar while Propolis is one of honeybee product with sticky and resinous nature. It is collected by honeybees from buds and barks of different trees and enriched in the beehive by addition of salivated secretions and wax (Bankova *et al*, 2000; Sime et al., 2015).

Honey and propolis contained several classes of phenolic acids, flavonoids, vitamins, enzymes, carbohydrate, pigments, aroma and minerals (Chua et al., 2013). The composition and the quantity of these phytochemicals are highly dependent on the influence of plants, climatic and environmental conditions, production methods, processing and storage conditions, as well as the nectar source of the honey (Sime et al., 2015; Kılıç Altun et al., 2017).

Although honey is best known by its sugar content and other phytochemicals, the presence of essential and toxic metals have also been reported in several papers (Mondragón-Cortez et al., 2013; Kılıç Altun et al., 2017). As indicated elsewhere, the source of these minerals are the soil where by the plant uptake it and translocate it in the nectar and afterwards inculpated in to the honey (Rodríguez García et al., 2006; Liberato et al., 2013; Stankovska et al, 2008). Thus, the soil chemistry and geological feature together with type of the flowing plant have influence on the overall mineral composition of honey (Pohl, 2009).

Minerals such as Co, Zn, Fe, Ni, Cu and Mn are required for human's metabolism at a certain concentration level. However, above the permissible limits, they are considered as toxic and hazardous to human. The levels of Pb and Cd are unacceptable owing to their carcinogenic and cytotoxic influences (Kılıç Altun et al., 2017).

In addition to natural sources, polluted environments have an impact on the quality of honey in terms of its mineral content. Heavy metals might be accumulated in the nectar through emissions of gases and particles or through translocation from the root (Liberato et al., 2013). Thus, the level of heavy metals in honey is an indicative of the environmental pollution of the region and geographical origin of the honey samples as well as quality of the honey (Lachman et al., 2007; Liberato et al., 2013).

Even though Sime et al., (2015) have investigated the phenolic composition and anti-oxidant activity of honey and propolis samples collected from Eastern, Western and Sothern parts of Ethiopia, the data available about the organic and inorganic contents of Ethiopian honey as well as propolis is insufficient. Particularly, the phenolic constituents were not studied in honey samples from the northern parts of the country. Furthermore, there is paucity of information on the mineral content of Ethiopian honey unlike honey samples from overseas countries.

It has to be noted that the Ethiopian natural honey and propolis are thought to be of different varieties due to the unique and highly diverse flora of the country because of its rich variety of environmental features ranging from semi-desert to mountain forests and its wide range of ecological, edaphic, and climatic conditions. There are over 7000 flowering plants species recorded, of which 12% are probably endemic to Ethiopia and most of them are bee plants (Fikru 2015; Simie et al., 2015). Ethiopia has the largest bee population in Africa with over 10 million bee colonies, out of which about 5 to 7.5 million are estimated to be hived while the remaining exist in the wild (Legesse, 2014; MoARD, 2007;). The annual honey production of Ethiopia is estimated to be 45,300 metric tons which makes the country to rank first honey producing country in Africa and ninth in the world (FAO, 2010; Fikru 2015). Ethiopia exports honey to France, Japan, Korea republic, Norway, Sweden, UK, Somalia and Sudan (http://www.ethiopianimporter.com/ethiopia-export-data/honey.html).

The test, aroma and texture of honey from the northern part of the country are recognizably different since the vegetation is different from the rest of the regions of the country. Therefore, this study was designed to determine the mineral composition and to estimate the phenolic content and antioxidant activity of honey and propolis samples collected from Amhara region of Ethiopia, more specifically from south Wolo zone districts.

Experimental

Materials and Equipment

Double beam UV (lambda-35) spectrometry, inductively coupled plasma optic emission spectrophotometer (ICP-OES, optima 8000 I-Perkin Elmer).

Chemical and reagents

Anhydrous sodium carbonate, Orthophosphoric acid (85%) and sodium molybdate dehydrate (98%), Anhydrous $AlCl_3$, Na_2NO_2, Anhydrous sodium tungstate ($Na_2WO_4.2H_2O$), phosphomolybdic acid, ethanol and methanol, gallic acid, catechin, Anhydrous Na_2CO_3, NaOH, HCl (37%), Br_2, Li_2SO4, 69.5% HNO_3, H_2O_2 (30%) and metal standards.

Description of the Study Area

Samples were collected from the northern part of Ethiopia called Amhara regional state and more specifically from South Wollo zone (Figure 1). The state is one of the major honey producing area of the country. Honeys from this area have distinct flavor, color and texture.

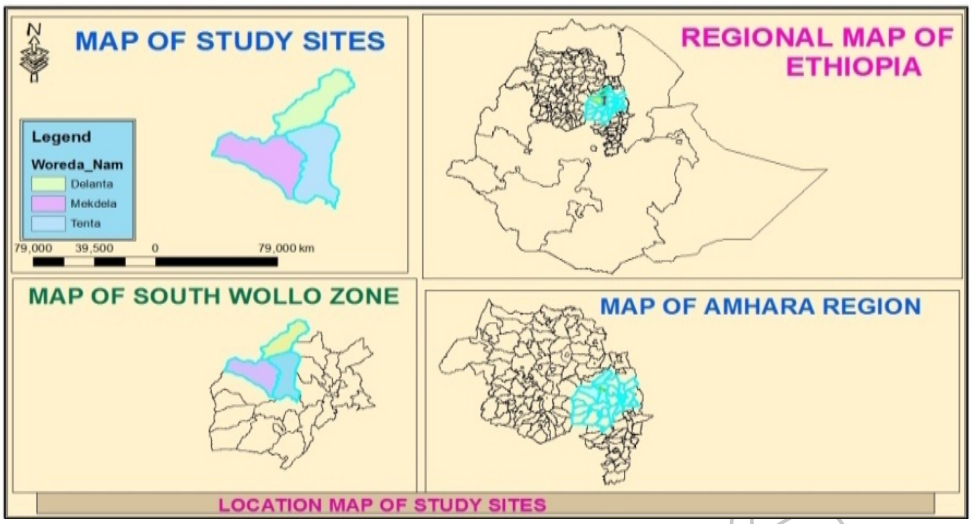

Figure 1. Map showing the sampling area

Collection of natural honey and propolis Samples

Seven natural honeys and three propolis samples (Figure 2) from traditional hives were randomly collected from different geographical areas of the South Wollo zone namely Ambamaryam district, Mekena district, Adjibar district, Tenta district, Wortej district, Kolo district and Chihna district. From one particular area, three to five hives were considered. From each hives about 300 grams of honey was collected and samples from similar area were mixed together to get a bulk sample. Table 1 shows details of the samples.

Figure 2 The three propolis and seven honey samples collected from study areas

Table 1 Description of sample type and sampling date

Sample code	Site of collection	Type of honey and color consistency	Harvest date (2016)	Production type

NH-1	Tenta district	Whitish yellow multi floral honey	October	Traditional
NH-2	Kolo district	Red multi floral	November	Traditional
NH-3	Kolo district	Medium yellow multi floral honey	December	Traditional
NH-4	Chihna district	Red multi floral	December	Traditional
NH-5	Wortej district	White multi floral	October	Traditional
NH-6	Mekena district	Whit multi floral honey	October	Traditional
NH-7	Ambamaryam district	Red multi floral honey	September	Traditional
Pro-1	Mekena district	Black multi floral propolis	March	Traditional
Pro-2	Mekena district	Medium black multi floral propolis	March	Traditional
Pro-3	Adjibar district	Light black multi floral propolis	March	Traditional

Sample preparation

The honey and propolis samples were processed for phenolic compounds analysis following the method reported by Sime et al., (2015). For honey samples, about 2.5 g of each sample was mixed with 50 mL distilled water. The filtrate was taken for analysis. Whereas, for the propolis samples, 5 g of each samples was mixed with 50 mL of 70% methanol in water and kept for one week by shaking intermittently. The filtrate was directly used for analysis. Total phenolics and total flavonoids contents were determined spectrophotometrically (Sime et al., 2015).

Determination of total phenolic compounds in honey and propolis samples

The total phenolic compounds of the seven honey and propolis samples were determined according to the Folin- Ciocalteau method (Sime et al., 2015). One milliliter of either honey solution (0.05 g/mL) or propolis (0.1 g/mL) was mixed with 2.5 mL of 7.5% sodium carbonate (Na_2CO_3) solution and 2.5 mL of 4 % sodium carbonate (Na_2CO_3) solution. To the mixture, 0.5 mL of Folin-Ciocalteau reagent which was prepared following the method reported by Bizuayehu et al., 2016 was added and the absorbance of the resulting solution was measured at 740 nm using double beam UV/ Vis spectrophotometer (lambda-35). All the measurements were done is triplicate and gallic acid was used as a reference standard. The results were expressed as mg gallic acid equivalents (GAE) per 100 g of samples.

Determination of total flavonoid compounds in honey and propolis samples

The total flavonoid contents of the honey samples were determined using aluminum chloride method and D-catechin as standard (Sime et al., 2015). Results were expressed as mg catechin equivalent /100 g honey or propolis samples.

Determination of metals content in honey

Honey samples were digested following the method reported by (Taddia et al. 2004). About 4.0 gram of honey sample was digested on a kjeldahl digestion apparatus using a mixture of HNO_3 and $HCLO_4$. The digested samples were diluted to 50 mL and filtered. The concentration of the elements (Cr, Co, Ni, Cu, Cd, Fe, Ca and Mg) in the digested solution of honey was determined using ICP-OES. Results were expressed as µg/100g sample and the data are presented in Table 4.

In order to evaluate the efficiency of the digestion procedure, 4.0 g of one of the honey sample (NH_1) was taken and spiked with 2.6 µg, 0.8 µg, 0.4 µg, 8.5 µg, 5.0 µg, 5.0 µg, 25.0 µg and 5.0 µg of Cr, Co, Cd, Ni, Cu, Mg, Ca, and Fe respectively. The spiked mixture was digested similar to the un-spiked sample. Results of the recovery experiment are indicated in Table 3. Following the same procedure, three blank samples were digested to calculate the detection limit of the method.

Statistical analysis

All the determinations were carried out in triplicate, and the data were expressed as mean ± standard deviation (SD). Significant differences of the data among the parameters were determined by analysis of variance (ANOVA) test with the help of SPSS version 20 software and means values were compared by tukey HSD (homogeneous subset difference) test. Difference at ($p \leq 0.05$) was considered significant.

Result and Discussion

Total phenolic content (TPC) in honey and propolis

The total phenolic content of the honey and propolis samples were estimated from the regression equation ($y=0.015x-0.053$; $R^2=0.9987$) of gallic acid which was derived from concentration ranging from 2.89 to 61.53 µg/mL. All data were acquired in triplicate and results are expressed in mean ± standard deviation (n=3, mean ± SD). Results are presented in Table 2

Table 2 The total phenol content (TPC) and total flavonoids content (TFC) of seven honey and three propolis samples, expressed as (mean ± SD, n = 3).

Types of sample	TPC (mg GAE/100) g samples.	TFC (mg CE/100g)
NH_1	51.7 ± 4.13^{a}	11.1 ± 2.3^{a}
NH_2	75.0 ± 3.08^{b}	10.9 ± 0.93^{a}
NH_3	58.4 ± 2.92^{c}	1.50 ± 0.12^{b}
NH_4	68.4 ± 5.08^{d}	41.7 ± 0.61^{c}
NH_5	45.4 ± 2.08^{e}	2.7 ± 0.10^{d}
NH_6	52.8 ± 1.08^{a}	ND
NH_7	73.5 ± 5.43^{b}	55.7 ± 4.23^{e}
PRO_1	262.5 ± 10.20^{f}	214.9 ± 13.86^{f}
PRO_2	204.4 ± 8.04^{g}	187.7 ± 11.61^{g}
PRO_3	236.2 ± 9.23^{h}	192.40 ± 0.87^{h}

Values in the same column that are followed by a different letters (a-f) are significantly different at $P \leq 0.05$ by tukey (homogeneous subset difference) test. ND = no detected.

In the studied honey samples, the total phenolic content was ranged from 45.4 ± 2.08 mg GAE/100 g (sample from Mekena district, NH-5) to 73.5 ± 5.43 mg GAE/100g (sample from Ambamaryam district, NH-7). In general, dark red colored honey samples (NH-7, NH-4 & NH-2) have shown the highest total phenolics content followed by light yellowish honey (NH-3 & NH-1) and whitish honey (NH-5 & NH-6). The same trend was obtained in other study (Sime et al., 2015). The data from this study was found to be about 5 to 10 times lower than the data obtained from other regions of Ethiopia (Sime et al., 2015). This is attributable to the differences in the floral origin of the northern part of the country as compared to the Easter, Southern and western parts of the country. However it was found to be comparable with Malaysian Tualang honey, Gelam honey and New Zealand Manuka honey (18.5-87.6 mg GAE/ 100g, 44.9-48.4 mg GAE/100g, and 43.5 mg GAE/100g respectively) (Khalil et al., 2011).

The total phenolic content of the propolis samples in this study ranged from 204.44 ± 8.04 mg GAE/100 g propolis sample (sample from adjibar district) to 262.51 ± 10.20 mg GAE/100g propolis (sample from mekena district). Propolis sample (Pro-1) has higher TPC, which was very black in color followed by the medium-blacked (pro-3) and light black (pro-2). This trend is in agreement with the result reported by Sime et al (2015) but the data found in this study are 1.5 to 4 times lower than the reported data from the other regions of the country.

Total flavonoid content (TFC) in honey and propolis samples.

The content of total flavonoid in the honey and propolis samples were derived from standard curve of catechin ranged from 5.0 to 320 µg in 5 mL ($y=0.005x + 0.068$; $R^2=0.9959$). The total flavonoid contents of the seven honey samples and three propolis samples expressed as mg catechin/100g of samples are given in Table 2. The total flavonoid content of the tested honey samples ranged from 1.5 ± 0.12 mg CE/100g (sample from Kolo district) to 55.7 ± 4.23 mg CE/100g (sample from Ambamaryam district). The red honey samples (NH-7 & NH-4) collected from Ambamaryam and Chihna district were found to contain significantly higher flavonoids content as compared to white honey (NH-6) and slightly yellowish honey (NH-3). Among the studied samples, flavonoids were not detected in white honey (NH-6). This confirms the dependence of flavonoids on the color of the honey samples. Comparing with the earlier study on Ethiopian honey, the honey sample (NH-7) was found to

contain slightly higher concentration than one of the honey sample reported by Sime et al., (2015) while the TFC in the other honey samples were lower than the data reported by Sime et al., (2015). One potential source of variation in TFC is because these honey samples were obtained from different beekeepers in various geographical regions and/or at different harvesting area. Even within honeys from a particular floral source, the composition can vary depending on climate and environmental stress factors, such as humidity, temperature, and soil composition (Khalil et al., 2011, Perna et al., 2012; Wieczorek et al. 2014).

Looking at the TFC in propolis samples (table 2), the black propolis (pro-1: 262.5 ± 10.20 mg CE/100g sample) contained considerable amounts of flavonoids than the medium (pro-3: 192.40 ± 0.87 mg CE/100 g sample) and light black propolis (pro-2: 187.7 ± 11.61 mg CE/100g samples). The observed variation in the flavonoid concentration of the investigated propolis is mainly accounted to the difference in the preferred regional plants (flora) collected by bees.

Elemental analysis

Method evaluation

The average recovery of each of the metals from the spiked sample was tabulated and shown in Table 3. The recoveries of the metals in the spiked honey sample were ranged between 88% to 103%. This indicates that the performance of the digestion method was within the acceptable range (80%-120%).

Table 3: Recovery test of honey sample.

Element	Concentration before spiked (mg/L)	Spiked concentration (mg/L)	% recovery (mean ± std)	Limit of detection (LOD)	Limit of quantification (LOQ)
Cr	0.052	0.052	94.9 ±4.45	0.0004	0.0015
Co	0.016	0.016	90.9±3.47	0.0046	0.016
Cd	0.008	0.008	91.6±11.4	0.0037	0.0123
Cu	0	0.10	103±8.98	0.0844	0.2816
Ni	0	0.10	91.7±3.07	0.0067	0.0223

Mg	0.17	0.17	88.2±5.89	0.00462	0.0154
Ca	0.496	0.50	89.5±9.83	0.0216	0.072
Fe	5.488	0.10	91.4±10.6	0.2633	0.8788

Level of metals in honey samples

Table 4 shows the concentration of eight elements in 7 honey samples collected from the Northern part of Ethiopia, specifically from North Wolo zone districts. It was observed that the concentration of the 8 elements varied widely within the different honey samples. Last column of Table 4 shows the total concentration of the element calculated by summing up the mean concentration of each of the eight elements investigated in a particular honey sample.

It was found that the yellowish honey sample (NH_1) from Tenta district had the highest mineral content (7788 µg/100 g) followed by medium yellowish honey sample (NH_3) from Kolo district (6233 µg/100 g) and Reddish honey (NH_2) from Kolo district (6003 µg/100 g). It has to be noted that the highest concentration of total element in samples (NH_1, NH_2 and NH_3) was mainly due to the presence of Fe in high concentration as compared to the other elements.

Table 4. Total mean concentration (µg/100 g honey) of each metal in seven different honey samples.

	Elements with total mean ± SD of 7 honey samples in µg/100 g honey								Total
Types of honey	Cr	Co	Cd	Ni	Cu	Mg	Ca	Fe	
NH_1	65.0 ± 1.3^{A}	20.0 ± 1.7^{A}	10.0 ± 0.63^{A}	Nd	Nd	212.5 ± 10.75^{A}	620 ± 29.1^{A}	6860 ± 192^{A}	7788
NH_2	57.5 ± 2.5^{B}	12.5 ± 2.13^{B}	10.9 ± 0.75^{A}	Nd	Nd	162.5 ± 3.50^{B}	250 ± 27.5^{B}	5510 ± 226^{B}	6003
NH_3	53.8 ± 1.63^{C}	114.0 ± 7.5^{C}	17.5 ± 0.5^{B}	Nd	Nd	172.5 ± 1.25^{C}	425 ± 23.7^{C}	5450 ± 142^{C}	6233
NH_4	60 ± 1.75^{B}	21.3 ± 1.63^{AD}	12.5 ± 4.3^{C}	Nd	Nd	147.5 ± 2.75^{D}	870 ±67.0^{D}	3738 ± 113^{D}	4849
NH_5	33.8 ± 0.63^{D}	23.8 ± 3.87^{D}	7.5 ± 1.13^{D}	Nd	Nd	132.5 ± 3.25^{E}	790±32.9^{E}	3172 ± 98.8^{E}	4160
NH_6	25.0 ± 1.37^{E}	31.3 ± 3.75^{E}	12.5 ± 0.25^{C}	Nd	Nd	127.5 ± 3.25^{E}	624±22.1^{A}	807 ± 133^{F}	1627
NH_7	63.8 ± 1.25^{A}	10.0 ± 1.63^{F}	3.75 ± 1.50^{D}	Nd	Nd	296 ± 2.50^{F}	910 ± 80.0^{F}	4234 ± 40.8^{G}	5518

Values in the same column that are followed by a different letters (A-F) are significantly different at $P \leq 0.05$ by tukey (homogeneous subset difference) test. The mean difference is significant at 0.05 levels. ND = no detected.

Iron (Fe) is one of the critical elements for humans due to its role in the production of red blood cells and its association with hemoglobin and the transfer of oxygen from lungs to the tissue cells. Fe was the most abundant among the tested metals in all the samples ranged from 807-6880 µg/100 g. The high concentration of Iron in the studied samples might be due to the high iron concentration in the pollen and in the environment as a whole. The Fe concentration obtained from this study is higher than Turkish honey (0.1-700 µg/100 g), Brazilian honey (178-3828 µg/100 g), Rumanian honey (2.2 µg/100 g), Egyptian honey (277 µg/100 g), Greece honey (239 µg/100 g) (Kılıç Altun et al., 2017; Santos et al., 2008; Mendes et al., 2006). While a relatively higher concentration of Fe was reported in honey samples from Saudi Arabia (6960-9813 µg/100 g (Alqarni et al., 2014) and from some other countries (Table 5). This variation can be ascribed to variation in the floral origin as well as soil composition of the regions. On the other hand, some of the honey samples from Malaysia contained an iron concentration closer to our finding (Chua et al., 2012).

Although most of the reported papers indicated that the relative concentration of Fe was lower than those of Ca and Mg (Rashed et al, 2004; Pohl, 2009; others) . However, In accordance with our data, a relatively higher concentration of Fe followed by Ca and Mg was reported in most of Malaysian honey except Manuka and few other honey types (Moniruzzaman et al., 2014).

Calcium (Ca) is the major abundant mineral in the body. Calcium in the body is mainly found in the bones & teeth (Saadiyah et al. 2015). The Ca level in the studied honey samples varies over a range of 250-910 µg/100 g. The Ca concentration obtained from this study is higher than those reported from Malaysia Turkey, Egyptian and Kenyan (Table 5) while a lower concentration of Ca was obtained in this study as compared to honey samples from (Brazil, Zech, France, India, Ireland, Itali, Macedonia, Spain and Turkey) (Table 5).

In this study the concentration of Mg was ranging from 127-296 µg/100 g. This concentration range is lower as compared to some known honey samples (Table 5). Karabagias et al., (2017), reported higher mean values of magnesium ranging from 810-1320 µg/100 in a study carried out on 37 honey samples collected from Egypt, Spain, Greece and Morocco. The same concentration range (600-3300 µg/100 g) was reported by Santos et al., (2008) on 52 honey samples produced in three different regional climates in the southwest Bahia, Brazil (semi-arid, Atlantic and Transitional Forest Zones). In

a study carried out by Chua et al (2012), much higher magnesium content was reported (mean values ranging from 5200-8950 µg/100 g).

Besides the major minerals (Ca, Mg and Fe), minor elements such as Co, Cr and Cd were also detected in the studied honey samples while Ni and Cu were below the detection limit of the instrument. The three elements are present in less than 1 µg/ g in all the studied samples. The concentration range of Cr, Co and Cd was 25-65 µg/100 g, 10-114 µg/100 g, 4.0-17.0 µg/100 g respectively. Comparing with literature values, a significantly higher concentration of Cr, Co, Cd, Ni and Cu were reported in most of honey samples collected from Africa, Asia and Europe (Table 5).

The low concentrations of Cr, Ni, Cd, and Cu are attributed to the uncontaminated environment of the sampling area. The districts where these samples were taken are less industrialized area and farming is the only source of the livelihood.

Table 5 Comparison of the concentration (µg/100 g) of metals in the studied samples with literature data.

Origin	Cr	Co	Cd	Ni	Cu	Mg	Ca	Fe	Ref.
Mexico	–			–	-	13.1-24.6	38.6-127.3	0.82-4.72	Mondragón-Cortez et al., 2013
Brazil	–	–	–	–	ND	ND–373	7.0–237	ND–15.0	Mendes et al., 2006; Santos et al., 2008
Chile	0.03-1.98	0.03-0.6	0.01-0.05	0.01-1.48	0.06–4.32	–	–	0.10–7.66	Fredes et al., 2006
Czech	–	–	–	0.06-1.53	0.12–0.89	18.5–89.1	11.3–142	–	Lachman et al., 2007
Egypt	–	1.75-3.20	0.010-0.5	1.25-4.10	1.00–1.75	103–1322	–	58–3691	Rashed et al., 2004
France			0.08-0.25	0.09-0.34	0.03–2.30	1.43–110	2.98–108	0.1–87.0	Devillers et al., 2002
Malaysia	3.08-3.68	ND-3.97	ND	ND-0.084	0.046-0.19	3.6-87.1	ND	11.0-31.1	Chua et al., 2012
Hungary	0.0018-0.109		0.0008-0.0033	–	0.04–0.44	–	–	–	Ajtony et al., 2007
India	–	0.05-0.25	0.3-0.5	0.37-0.4	1.06–2.91	–	32.6–84.6	3.60–28.4	Nanda et al., 2003; Buldini et al., 2001
Ireland	–	ND	–	–	1.00–2.30	18.9–53.3	74.9–175	1.70–36.3	Downey et al., 2005
Italy	ND-0.089	0.002-0.057	ND	ND-2.76	0.14–5.90	3.90–159	9.10–409	0.30–35.1	Caroli et al., 1999; Pisani et al., 2008

Macedonia	–	–	0.001-0.27	–	0.02–5.90	4.40–182	4.10–170	0.03–7.00	Stankovska et al., 2008
Poland	–	–	–	–	ND–1.82	1.10–19.8	3.30–159	ND–16.1	Przybyłowski et al., 2001; Madejczyk et al., 2008
Spain	0.006-0.041	–	0.0008-0.006	0.012-0.17	0.04–7.80	18.0-308	41-385	ND–21.0	Rodríguez García et al., 2006
Turkey	0.002-0.54	–	< 0.001	0.00-0.42	ND–3.50	0.001–111	0.001–900	0.04–19.7	Uren et al., 1998; Kılıç Altun et al., 2017
Ethiopia	0.25-0.65	0.1-1.14	0.038-0.18	ND	ND	1.28-2.96	2.50-9.10	8.07-68.6	This study

Conclusion

In this report, we have presented the total phenolic content, total flavonoids and some selected macro and micro elements in seven honey and three propolis samples collected from the Northern part Ethiopia, South Wolo zone districts. The composition of the honey and the propolis samples vary with the color of the honey and geographical origin. This study confirmed that, colored honey and propolis samples were rich in phenolic compounds than the white or light colored samples, which signifies that the former samples are medicinally more important in terms of phenolic content. The mineral data suggested that honey samples of South Wolo zone districts of the Amhara region of Ethiopia were of good quality because the concentration of some of the toxic heavy metals were below the maximum permissible limit.

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Competing interest

The authors declare no competing interest.

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