PHYSICOCHEMICAL AND RESIDUE ANALYSIS OF HONEY FROM BLACK SEA REGION OF TURKEY

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ABSTRACT

Honey samples obtained from Black Sea Region of Turkey were screened for physicochemical quality as well as the presence of heavy metals and minerals, veterinary drugs (antibiotics), pesticide and naphthalene residues. Mean values obtained for physicochemical parameters were 5.42±0.02 pH; 16.66±0.12% moisture; 67.54±0.49% invert sugar; 3.62±0.13% sucrose; 0.2±0.01% ash; 0.48±0.03 mS/cm electrical conductivity; 24.97±0.27 meq/kg acidity; 8.86±0.38 mg/kg hydroxymethylfurfuraldehyde, and 10.45±0.26 diastase activity. In the analysed samples, potassium was the major element, and manganese the minor one. Mean values obtained were (mg/kg): Ca, 4.83±0.33; K, 195.29±5.28; Mg, 19.97±0.88; Na, 10.75±0.23; Mn, 0.34±0.05; B, 0.71±0.04. The contents of heavy metals (Cu, 0.18±0.01; Zn, 0.16±0.01; Cd, 0.07±0.00; Fe, 1.66±0.04; Ba, 1.02±0.03; Ni, 0.14±0.01; Al, 0.021±0.001 mg/kg; Cr, 6.58±0.28; Pb, 6.99±0.31 µg/kg) were within permitted levels. 32.5 % of the examined samples had drug residues at least one kind. Only the honey samples from Artvin had no residue of veterinary drugs (streptomycin, tetracyclines and sulphonamides). Amitraz and flumethrin were detected in 44 (21%) and 73 (34.9%) samples at concentration ranging from 57.9 to 167.4 ppb and 20.9 to 38.6 ppb, respectively. Although the mean value for naphthalene residue was found to be 4.04±0.48 µg/kg, it was understood that using of naphthalene had been progressing. The results obtained for physicochemical characteristics of Black Sea Region honeys indicate a good quality level, adequate processing, good maturity and freshness. But in terms of residues of antibiotics, pesticides and naphthalene, the beekeepers should be educated on the value and importance of the quality of honey, and further investigations are needed.

KEYWORDS: Heavy metal, mineral content, pesticide, physicochemical characteristics, naphthalene, veterinary drug

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1. INTRODUCTION

The composition of honey is variable, owing to the differences in plant types, climate, environmental conditions, and contribution of the beekeeper [1-3]. Generally, the pH of honey is approximately 4.0 [4]. The differences in the chemical composition of honey are reflected in many physicochemical properties [5], and the quality of honey is mainly determined by its sensorial, chemical, physical and microbiological characteristics. The major criteria of interest are moisture content, electrical conductivity, ash content, reducing and non-reducing sugars, free acidity, diastase activity, and 5- hydroxymethylfurfuraldehyde (HMF) content [6]. Among the honey producer countries in Turkey, thanks to geographical and climatic conditions that provide a suitable environment for apiculture in terms of the flowers, honey production has been well developed. The beekeeping that has been sustained in Turkey for thousands of years is an important agricultural activity [7].

Antibiotics and pesticides in agricultural foods such as honey are a recurrent problem [8]. According to the European Union (EU) regulations, honey must be free from organic and inorganic matter foreign to its composition [9], namely, the addition of additives and preservatives is not allowed.

The objective of the current study is characterization of 209 honey samples harvested in Black Sea Region of Turkey, with respect to physicochemical parameters (ash, electrical conductivity, moisture, pH, acidity, diastase activity, HMF content, invert sugar and sucrose), the content of mineral elements (Mn, B, Mg, Na, Ca and K) and heavy metals (Cu, Cd, Zn, Ni, Fe, Ba, Cr, Pb and Al), veterinary drugs (streptomycin (STR), tetracyclines (TCs) and sulphonamides), pesticide (amitraz and flumethrin) and naphthalene residues. Consequently, this part of Turkey was evaluated in terms of quality of the produced honeys, and the education level of region beekeepers.

2. MATERIALS AND METHODS

2.1 Honey samples

209 comb honey samples of different floral sources provided directly from beekeepers at eighteen cities of Black Sea Region were used as the most important material in this study. According to honey production data of these cities in the year 2005, the number of samples were determined. The harvest time of samples was in 2007. The comb honey samples were kept under suitable storage conditions until filtration. After the mechanical filtration, the samples were stored until analysis in a jar at room temperature. Honeycombs were also stored at the refrigerator for analysis.

Physicochemical (ash, moisture, HMF, sucrose, pH, acidity, invert sugar, diastase activity and electrical conductivity) as well as heavy metal and mineral analyses of samples were done. Furthermore, various kinds of veterinary drugs, pesticides and naphthalene residue analyses were performed for the samples.

2.2 Physicochemical analyses

The ash content was determined using the method adopted by the International Honey Commission [10]. 1 g of honey samples was placed in a furnace crucible at 550 °C to turn into white ash. The content of ash was expressed as g%.

Electrical conductivity was determined using a conductivity meter from a solution containing 10 g of honey in 50 ml of distilled water [10].

Moisture was determined by measuring the refractive indices at 20 °C with an Atago model refractometer, and the corresponding moisture contents (%) were calculated [11].

The pH of the honey was measured by a pH-meter (Sartorius) in solution of 10 g honey in 75 ml CO_2-free distilled water [12]. For determination of acidity, the solution was titrated with 0.05 N NaOH until pH 8.3, after adding phenolphthalein [11].

Diastase activity was measured using a buffered solution of soluble starch and honey which was incubated at 47 °C in a test tube. Results were expressed as ml of 1% of starch hydrolyzed by an enzyme in 1 g honey within 1 h [13].

For determination of HMF content, 10 g honey was solved in 50 ml oxygen-free water without heat. After adding p-toluidine and barbituric acid solutions, the resultant colour was measured against a blank in 1-cm cuvettes at 550 nm [14].

A modified method, involving the reduction of Soxhlet's modification of Fehling's solution by titration at boiling point against a solution of reducing sugars in honey using methylene blue as an internal indicator was used for determination of invert sugar [15]. The difference in concentrations of invert sugar was multiplied by 0.95 to give the apparent sucrose content.

Five ml of diluted nitric acid solution (6:1) were added to the ashes obtained by calcination at 550 °C, of approximately 5 g honey sample, until constant weight and the mixture was stirred on a heating plate to almost complete dryness. Then, the mixture was made up to 25 ml with distilled water and concentrations of Ba, B, Ca, Cd, Cr, Cu, Fe, Mg, Mn, Na, Ni, Pb, Zn, K and Al were determined directly in the ash solution by atomic absorption spectrometry at a suitable wavelength [16].

2.3 Residue Analysis

Screening of veterinary antibiotic residues (STR, TCs and sulphonamides) in honey was performed by using commercially available Charm II tests produced by Charm Sciences Inc. USA. It is a radio receptor assay based on the specific binding of antibiotics to receptors. Charm II principle was followed, and samples with high count results were considered to be negative while samples with low count were considered to be positive [8].

Honey samples were also analysed with an in-house developed method for determination of common acaricides, such as amitraz and flumethrin, by using gas chromatography–mass spectrometry (GC–MS) according to the procedure by Maver and Poklukar [17].

In honey samples, naphthalene residues were determined by using GC-MS system with solid-phase microextraction (SPME) method. 1 g honey and methanol-water mixture (1:1) were loaded into a vial. Commercially available fiber was employed for the extraction [18].

3. RESULTS AND DISCUSSION

3.1 Physicochemical analyses

All results of physicochemical analyses are summarized in Table 1. Statistically significant differences were observed for the tested parameters, such as ash, electrical conductivity, moisture, pH, diastase number and invert sugar of each group of honeys. But the mean total acidity, HMF and sucrose values obtained from the honey samples from all cities were not significantly different.

Ash content is a quality criterion used for the determination of the botanical (floral, mix or honeydew) and geographical origins of honey samples [7]. Harvesting processes, beekeeping techniques and the material collected by the bees foraging on the flora can affect the ash content of honeys [19]. The mean ash content of the honey samples provided from each city varied between 0.00 and 0.52%, and the mean was determined to be 0.20±0.01% for all regions. Generally, the ash content of blossom honey is ≤0.6% as compared to honeydew honey

TABLE 1- Results of physicochemical analyses of honey samples harvested in Black Sea Region of Turkey.

Geographical origin of honey (sample number)	Ash (%)	Electrical conductivity (mS/cm)	HMF (mg/kg)	Diastase number	Moisture (%)	Acidity (meq/kg)	Invert sugar (%)	Sucrose (%)	pH
Tokat (7)	0.11 ± 0.01 d	0.46 ± 0.19 c-f	4.94 ± 0.83	10.70 ± 1.85 a-c	16.68 ± 0.51 a-d	23.57 ± 1.33	60.41 ± 2.72 e	5.43 1.34	5.57 ± 0.13 a
Çorum (6)	0.17 ± 0.09 b-d	0.49 ± 0.13 c-f	8.19 ± 1.74	11.67 ± 1.20 a-c	15.69 ± 0.76 cd	26.17 ± 1.35	65.97 ± 3.11 b-e	3.14 ± 0.44	5.58 ± 0.15 a
Bolu (4)	0.23 ± 0.05 b	0.70 ± 0.25 b-f	7.72 ± 2.06	14.68 ± 2.86 ab	15.12 ± 0.27 d	28.25 ± 1.49	69.06 ± 2.87 a-e	6.15 ± 0.92	5.63 ± 0.13 a
Bartın (4)	0.16 ± 0.04 b-d	1.21 ± 0.48 ab	8.32 ± 3.77	12.30 ± 2.41 a-c	17.21 ± 0.59 a-d	21.50 ± 1.04	71.54 ± 1.50 a-d	3.49 ± 0.25	5.50 ± 0.20 a
Zonguldak (4)	0.12 ± 0.01 cd	1.45 ± 0.24 a	10.10 ± 1.14	10.65 ± 1.91 a-c	17.94 ± 0.32 a-c	27.50 ± 1.32	73.02 ± 3.09 a-c	2.91 ± 0.54	5.63 ± 0.13 a
Karabük (2)	0.17 ± 0.05 b-d	0.49 ± 0.19 c-f	12.94 ± 6.27	11.10 ± 2.80 a-c	17.12 ± 0.22 a-d	22.50 ± 1.50	64.34 ± 1.84 c-e	4.79 ± 1.30	5.50 ± 0.00 a
Amasya (2)	0.46 ± 0.03 a	0.43 ± 0.10 c-f	9.70 ± 2.98	15.90 ± 2.00 a	16.91 ± 0.36 a-d	24.00 ± 5.00	77.06 ± 0.53 a	4.38 ± 1.09	5.50 ± 0.00 a
Kastamonu (7)	0.11 ± 0.01 d	0.75 ± 0.16 b-e	9.54 ± 1.20	9.33 ± 1.02 bc	17.44 ± 0.61 a-d	25.86 ± 1.58	72.67 ± 3.07 a-c	3.26 ± 0.32	5.43 ± 0.13 ab
Düzce (4)	0.17 ± 0.09 b-d	0.82 ± 0.18 b-e	14.52 ± 3.86	10.25 ± 0.65 bc	19.02 ± 0.16 a	23.50 ± 1.94	71.78 ± 0.62 a-d	2.82 ± 0.34	5.50 ± 0.20 a
Sinop (4)	0.23 ± 0.05 b	0.90 ± 0.31 b-d	11.01 ± 3.08	10.55 ± 1.52 a-c	17.10 ± 0.23 a-d	24.75 ± 2.18	75.07 ± 2.39 ab	3.78 ± 0.16	5.25 ± 0.14 a-c
Gümüşhane (10)	0.16 ± 0.04 b-d	0.19 ± 0.01 f	8.88 ± 0.85	9.46 ± 1.51 bc	16.13 ± 0.45 b-d	24.80 ± 1.14	66.37 ± 2.39 b-e	3.98 ± 0.48	5.45 ± 0.12 ab
Giresun (14)	0.12 ± 0.01 cd	0.61 ± 0.15 c-f	8.23 ± 1.37	10.49 ± 0.83 a-c	16.50 ± 0.59 a-d	25.14 ± 1.14	63.11 ± 1.43 c-e	3.42 ± 0.20	5.64 ± 0.08 a
Rize (9)	0.17 ± 0.05 b-d	0.97 ± 0.16 a-c	9.19 ± 1.12	7.71 ± 0.69 c	18.57 ± 0.72 ab	25.33 ± 1.05	72.74 ± 2.12 a-c	3.25 ± 0.32	5.44 ± 0.10 ab
Ordu (86)	0.46 ± 0.03 a	0.41 ± 0.04 d-f	9.00 ± 0.64	9.55 ± 0.35 bc	16.60 ± 0.20 a-d	24.57 ± 0.41	68.25 ± 0.73 a-e	3.59 ± 0.23	5.38 ± 0.03 ab
Artvin (16)	0.20 ± 0.01 b-d	0.35 ± 0.06 d-f	8.45 ± 1.69	12.07 ± 1.15 a-c	16.03 ± 0.32 b-d	25.20 ± 0.87	63.43 ± 1.60 c-e	3.19 ± 0.29	5.53 ± 0.09 a
Samsun (17)	0.21 ± 0.01 b-d	0.28 ± 0.04 ef	9.41 ± 1.71	12.41 ± 1.10 a-c	16.29 ± 0.43 b-d	24.71 ± 1.18	65.99 ± 1.70 b-e	3.58 ± 0.36	5.44 ± 0.07 ab
Trabzon (12)	0.25 ± 0.03 b	0.26 ± 0.02 ef	7.36 ± 1.40	11.41 ± 1.04 a-c	16.12 ± 0.45 b-d	27.33 ± 1.29	66.16 ± 1.46 b-e	3.16 ± 0.30	4.92 ± 0.12 c
Bayburt (2)	0.46 ± 0.04 a	0.19 ± 0.02 f	6.50 ± 0.10	12.40 ± 1.50 a-c	18.29 ± 0.97 a-c	26.50 ± 1.50	62.42 ± 0.19 de	4.90 ± 2.69	5.00 ± 0.00 bc
Mean Value	0.20 ± 0.01	0.48 ± 0.03	8.86 ± 0.38	10.45 ± 0.26	16.66 ± 0.12	24.97 ± 0.27	67.54 ± 0.49	3.62 ± 0.13	5.42 ± 0.02
Min-Max	0.00-0.52	0.12-2.42	0.77-35.12	5.00-23.00	12.08-22.55	17.00-34.00	50.21-87.94	0.35-16.29	4.50-6.00
TFC, EU, CODEX standards	<0.6[1] <1.2[2]	<0.8[1] >0.8[2]	<40[1] <40[2]	>8[1] >8[2]	<20[1] <20[2]	<50[1] <50[2]	>60[1] >45[2]	<5[1] >10[2]	>4.2[1] >4.2[2]
Confidence level (P)	<0.01	<0.01	>0.05	<0.05	<0.01	>0.05	<0.01	>0.05	<0.01

Flower (1), Honeydew (2) honeys, Means with different letters in the same line are significantly different from another (P<0.05, P<0.01); No saturated values are determined for pH in EU and CODEX

or blends of honeydew and blossom honeys where this value is ≥1.2% [4]. We can conclude that the analysed samples could be similar to the blossom honeys, and were appropriate for the limit allowed in Turkish Food Codex (TFC). The result is good agreement with Sunay [20].

The electrical conductivity of honey is closely related to the concentration of mineral salts, organic acids and proteins. This parameter shows great variability according to the floral origin, and it is important for the differentiation of honeys of different floral origins [21]. Therefore, itis very often used in routine honey control. The electrical conductivity (mS/cm) in honey samples varied in the range of 0.12–2.42, and the mean value was 0.48±0.03 mS/cm.

These values are similar to those reported by Sahinler *et al.* [22], Gül [23], and another study on Saudi Arabian floral honeys [24]. The mean value obtained for the honey samples under study varied between below the maximum limit indicated by TFC and EU standards for blossom honeys (0.8 mS/cm). However, the electrical conductivity values of 31 samples provided from Zonguldak, Bartın, Rize, Sinop, Düzce and Rize were above the determined limit. This was an expected situation because there are honeydew honeys in honey samples of these cities, and a lower limit has been proposed for blossom than for honeydew honeys [5]. The lower value was seen for the samples from Gümüşhane and Bayburt.

The different moisture content of honey depends on harvest season, the degree of maturity reached in the hive, and moisture content of original plant [19]. Furthermore, the variations in this parameter have been attributed to the composition and floral origins of honey [25], and it can vary from year to year. In this study, only 2.39% of samples were in an unacceptable range according to moisture analyses. Moisture contents of almost all samples were below 20%, the maximum value allowed by TFC and European Communities (EC) and Codex standards, which means that the fermentation ability is low. Similar [22, 26, 27] and lower [28] results were detected in previous studies. Moisture content is practically the most important quality parameter, since it affects storage life and processing characteristics. The strong interaction of sugar in honey with water molecules may decrease the water available for microorganisms. The low moisture content of honey also forms an important part of the system which protects honey from attack by microorganisms [29], and lower moisture limits ensuring a better shelf-life of honey [29]; so, the honeys from Black Sea Region have good storage ability.

Honey pH is affected by the conditions during extraction and storage, which also influences texture, stability and shelf-life. The pH is indeed a useful index of possible microbial growth, since most bacteria grow in a neutral and mildly alkaline environment, while yeasts and moulds are capable of developing in an acidic environment (pH 4.0–4.5), and do not grow well in alkaline media [31]. The pH values of the analysed honey samples provided from Black Sea Region ranged from 4.50 to 6.00 (mean value 5.42±0.02). Except for the pH values obtained for Trabzon and Giresun honeys, differences in pH values of other cities were not statistically significant. All region honeys have required pH values being higher than 4.2 according to TFC. The pH values obtained in this study

were in accordance with pH value of 5.6 for sunflower honeys reported by Şahinler and Gül [28].

The mean value of acidity was found to be 24.97±0.27, with the range of 17.00 to 34.00 meq acid/kg. None of the samples exceeded the limit allowed by TFC and EU (<50 meq acid/kg), which may be taken as indicative of freshness of all honey samples. Similar results had been detected as 22.30, 28.19 and 25.0 meq/kg, respectively [23, 26, 32]. Obtained values were larger than those reported by Russo Almeida [33] of 16.50 meq acid/kg but smaller than the 31.62 meq acid/kg recorded by Sunay [20]. The variation in acidity among different honey samples reported in literature may be attributed to variation of harvest season [34], and the plants' floral types [35].

Diastase is a natural enzyme of honey [6]. Enzymes, like diastase, play an important role in the biological value of honey [27]. Variation of diastase activity is dependent on lots of factors including the amount of sucrose in food sources, rate of nectar flow, floral origins of the product, and even age of the bees [36]. Exposure to high temperatures and long storage periods inactivate diastase [27]. Herein, honey samples exhibited very different values, ranging between 5.00 and 23.00. The mean of diastase activity was 10.45±0.26 in all samples. This value was above the limit value of 8 determined by FAO/WHO Codex, TFC and EU. But, 50 of the samples investigated in this study presented an inappropriate diastase activity with values below 8, suggesting inadequate storage or processing. Sahinler *et al.* [22] as well as Ünal and Küplülü [27] reported that the diastase activity had values of 10.31 and 11.58, respectively. However, higher diastase numbers had been reported previously as 17.9, 21.43 and 22.81, respectively [20, 23, 28]. In contrary, lower values were found by Fallico *et al.* [32]. We found considerably lower diastase activity in honey from cities in which monofloral honey was mostly produced. Honey samples provided from Rize had the lowest diatase activity. Monofloral honeys, such as chestnut and clover, are produced in Rize.

The HMF content is widely recognized as a parameter of honey samples freshness, because it is absent in fresh honeys and tends to increase during processing and/or aging of the product. Several factors influence the levels of HMF, such as temperature and time of heating, storage conditions, pH and floral source; thus, it provides an indication of overheating and storage in unworthy conditions [37]. Before determining storage-dependent parameters like enzyme activity and colour, one should ensure that honeys are fresh and unheated. Before testing these parameters, it should be checked that the HMF content is below 15 mg/kg [38]. HMF values of all samples analysed fell within the international limit of 40 mg/kg according to WHO/FAO Codex, TFC and EU. The mean of HMF was found to be 8.86±0.38 mg/kg, with the range of 0.77 to 35.12 mg/kg. Significantly lower levels than the data presented here were reported as 3.3, 0.71 and 4.95 mg/kg, respectively [20, 22, 26]. Our results were in agreement with those reported by Gül [23], who indicated HMF content below 10 mg/kg. On the contrary, Ünal and Küplülü [27] had reported a high HMF value of 74.51 mg/kg for flower honeys consumed in Ankara, being remarkably above the international standard limit.

Botanical origin of honey is related to sugar composition because the carbohydrates are more than 95% of honey solids [39]. Reducing sugars (or invert sugar), mainly fructose and glucose, have been found to be the major constituent of honey [40]. A high sucrose concentration of honey, most of the time, means an early harvest of honey because sucrose has not been fully transformed to glucose and fructose by the action of invertase [35]. The mean percentage of sucrose (3.62±0.13 %) of all the honey samples was below the maximum allowable limit of 5% proposed by TFC, FAO/WHO Codex and EU. But 26 of all samples had sucrose levels above 5%. Our findings showed approx. similarity with the result of Sunay [20]. Higher and lower results were detected in previous studies [23, 27]. Moreover, the average invert sugar ratios of investigated samples were determined as 67.54±0.49. The values varied in the range of 50.21 and 86.53, and they were in association with the value of 70.30% [26]. The results of this study indicate that honeys contain sugar, bees were fed with sugar solution instead of nectars, and early harvesting before honey ripened in the honeycomb honeys have been remixed [27].

The mineral content is an important index of possible environmental pollution and a potential indicator of geographical origin of honey [2]. Air and soil contain heavy metals, mainly from industry and traffic, which can also contaminate the bee colony and its products [41]. The amounts of the 9 heavy metals and 6 mineral elements determined in honey samples are summarized in Tables 2 and 3. Contents of heavy metals in Black Sea Region honeys are well below the permitted limits of Codex Standards, and the determined values were generally comparable to those reported by other authors. But worldwide, there are no specific Maximum Residue Limit (MRL) levels for heavy metals, such as Cd, Ni, Ba, Cr and Al in honey.

Honey is an important source of trace elements that play fundamental negative and positive roles in human life. Many factors are effective in element contents of honey, such as geographical and botanical origin, soil, atmosphere, beekeeping equipments, element content of nectar, season of the year, and rainfall among the others [42-44]. The honey samples from West Black Sea Region cities have higher quantities of mineral elements (Mn, B, Mg, Na, Ca and K) than other city honeys investigated in this study because in this region chestnut honeys are produced mostly in ratio to the other cities. In terms of 6 of the investigated heavy metals (Cd, Ni, Al, Cr, Ba and Pb), Bayburt honeys have the lowest values. In our study, it

TABLE 2 - Results of heavy metal analyses of honey samples harvested in Black Sea Region of Turkey.

Geographical origin of honey (sample number)	Ca (mg kg^{-1})	Mg (mg kg^{-1})	Na (mg kg^{-1})	Fe (mg kg^{-1})	Ba (mg kg^{-1})	Ni (mg kg^{-1})	Al (mg kg^{-1})	Cr (ng g^{-1})	Pb (ng g^{-1})
Tokat (7)	0.17 ± 0.03 ab	0.16 ± 0.03 ab	0.07 ± 0.02	1.50 ± 0.23 bc	1.20 ± 0.12	0.16 ± 0.03	0.026 ± 0.004	5.63 1.34	6.80 ± 1.89
Çorum (6)	0.20 ± 0.04 ab	0.17 ± 0.03 ab	0.07 ± 0.02	1.28 ± 0.21 c	1.45 ± 0.19	0.12 ± 0.03	0.020 ± 0.004	5.60 ± 0.44	7.53 ± 1.65
Bolu (4)	0.10 ± 0.00 b	0.13 ± 0.03 ab	0.08 ± 0.03	1.78 ± 0.06 bc	1.28 ± 0.17	0.25 ± 0.03	0.018 ± 0.003	5.78 ± 0.92	6.23 ± 2.51
Bartın (4)	0.23 ± 0.03 a	0.08 ± 0.05 ab	0.13 ± 0.03	1.37 ± 0.11 bc	1.10 ± 0.07	0.13 ± 0.03	0.018 ± 0.005	3.95 ± 0.25	4.08 ± 0.38
Zonguldak (4)	0.18 ± 0.05 ab	0.10 ± 0.41 ab	0.08 ± 0.03	2.13 ± 0.21 ab	1.05 ± 0.17	0.13 ± 0.03	0.015 ± 0.003	7.05 ± 0.54	3.58 ± 0.77
Karabük (2)	0.15 ± 0.05 ab	0.00 ± 0.00 b	0.00 ± 0.00	2.74 ± 0.32 a	1.75 ± 0.25	0.20 ± 0.10	0.015 ± 0.005	4.20 ± 1.30	14.65 ± 0.05
Amasya (2)	0.25 ± 0.05 a	0.25 ± 0.05 a	0.00 ± 0.00	1.37 ± 0.40 bc	1.35 ± 0.25	0.10 ± 0.00	0.015 ± 0.005	2.50 ± 1.09	8.05 ± 3.45
Kastamonu (7)	0.24 ± 0.02 a	0.09 ± 0.03 ab	0.06 ± 0.02	1.33 ± 0.05 c	1.27 ± 0.14	0.17 ± 0.03	0.027 ± 0.003	5.74 ± 0.32	6.77 ± 1.73
Düzce (4)	0.15 ± 0.03 ab	0.15 ± 0.07 ab	0.02 ± 0.03	1.46 ± 0.09 bc	1.23 ± 0.26	0.15 ± 0.03	0.020 ± 0.004	5.88 ± 0.34	5.95 ± 1.82
Sinop (4)	0.10 ± 0.00 ab	0.03 ± 0.03 b	0.13 ± 0.03	1.60 ± 0.12 bc	0.98 ± 0.03	0.15 ± 0.03	0.025 ± 0.003	4.80 ± 0.16	3.95 ± 1.34
Gümüşhane (10)	0.15 ± 0.02 ab	0.12 ± 0.03 ab	0.09 ± 0.01	1.72 ± 0.03 bc	1.14 ± 0.15	0.19 ± 0.03	0.019 ± 0.003	8.31 ± 0.48	6.81 ± 1.58
Giresun (14)	0.20 ± 0.02 ab	0.18 ± 0.07 ab	0.06 ± 0.01	1.78 ± 0.04 bc	1.06 ± 0.13	0.10 ± 0.02	0.021 ± 0.003	7.21 ± 0.20	7.44 ± 1.13
Rize (9)	0.22 ± 0.02 a	0.16 ± 0.03 ab	0.04 ± 0.02	1.83 ± 0.17 bc	0.92 ± 0.18	0.16 ± 0.03	0.020 ± 0.003	8.91 ± 0.32	7.47 ± 1.78
Ordu (86)	0.18 ± 0.01 ab	0.21 ± 0.02 ab	0.07 ± 0.01	1.64 ± 0.07 bc	1.03 ± 0.05	0.14 ± 0.01	0.020 ± 0.001	6.66 ± 0.23	7.40 ± 0.51
Artvin (16)	0.17 ± 0.02 ab	0.12 ± 0.02 ab	0.06 ± 0.01	1.53 ± 0.14 bc	0.87 ± 0.12	0.14 ± 0.02	0.021 ± 0.002	5.96 ± 0.29	7.63 ± 0.99
Samsun (17)	0.20 ± 0.01 ab	0.17 ± 0.03 ab	0.08 ± 0.02	1.56 ± 0.08 bc	0.68 ± 0.14	0.10 ± 0.02	0.021 ± 0.002	7.83 ± 0.36	6.52 ± 0.76
Trabzon (12)	0.14 ± 0.02 ab	0.10 ± 0.03 ab	0.08 ± 0.02	1.95 ± 0.17 bc	0.88 ± 0.18	0.11 ± 0.02	0.022 ± 0.003	6.22 ± 0.30	6.23 ± 1.24
Bayburt (2)	0.15 ± 0.05 ab	0.10 ± 0.00 ab	0.00 ± 0.00	2.05 ± 0.29 bc	0.50 ± 0.50	0.00 ± 0.00	0.015 ± 0.005	2.15 ± 2.69	2.00 ± 0.10
Mean Value	0.18 ± 0.01	0.16 ± 0.01	0.07 ± 0.00	1.66 ± 0.04	1.02 ± 0.03	0.14 ± 0.01	0.021 ± 0.001	6.58 ± 0.13	6.99 ± 0.31
Min-Max	0.10-0.30	0.00-1.00	0.00-0.30	0.48-3.30	0.00-2.00	0.00-0.30	0.00-0.04	1.10-18.90	1.10-18.90
CODEX standards	0.1-5.0	-	-	1.5-15	-	-	-	<5	0.1-2.0
Confidence level (P)	<0.05	<0.05	>0.05	<0.05	>0.05	>0.05	>0.05	>0.05	>0.05

Means with different letters in a same line are significantly different from one another (P<0.05, P<0.01)

TABLE 3 - Results of mineral analyses of honey samples harvested in Black Sea Region of Turkey.

Geographical origin of honey (sample number)	Ca (mg kg^{-1})	Mg (mg kg^{-1})	Na (mg kg^{-1})	Mn (mg kg^{-1})	K (mg kg^{-1})	B (ng g^{-1})
Tokat (7)	4.60 ± 0.87 ab	26.40 ± 5.04 a-e	13.46 ± 0.97 ab	0.03 ± 0.02 de	154.90 ± 23.94 cd	0.69 ± 0.16
Çorum (6)	3.98 ± 0.87 ab	31.70 ± 4.37 a-c	16.03 ± 0.74 a	0.03 ± 0.02 de	190.18 ± 23.17 a-d	0.68 ± 0.25
Bolu (4)	3.13 ± 0.98 ab	33.05 ± 5.94 a-c	13.75 ± 0.68 ab	0.38 ± 0.28 de	259.73 ± 20.15 abc	0.08 ± 0.25
Bartın (4)	3.60 ± 0.51 ab	16.23 ± 2.60 d-g	13.43 ± 1.46 ab	0.65 ± 0.24 b-e	226.98 ± 42.19 a-d	0.75 ± 0.25
Zonguldak (4)	7.10 ± 0.30 ab	30.00 ± 4.50 a-d	14.15 ± 1.46 ab	3.13 ± 1.15 a	294.16 ± 2.78 a	1.05 ± 0.25
Karabük (2)	3.10 ± 0.50 ab	18.65 ± 7.25 c-f	13.50 ± 0.80 ab	0.10 ± 0.10 de	240.80 ± 7.70 a-d	0.90 ± 0.25
Amasya (2)	7.45 ± 0.15 ab	27.95 ± 0.25 a-e	12.75 ± 1.45 ab	0.10 ± 0.00 de	205.40 ± 20.00 a-d	0.00 ± 0.25
Kastamonu (7)	4.17 ± 0.95 ab	34.17 ± 5.85 ab	11.43 ± 0.27 b	0.37 ± 0.17 de	250.59 ± 19.70 a-c	0.29 ± 0.25
Düzce (4)	6.88 ± 1.01 ab	34.03 ± 2.30 ab	12.38 ± 0.36 ab	1.38 ± 0.85 b	270.58 ± 10.78 ab	0.18 ± 0.25
Sinop (4)	6.08 ± 1.83 ab	35.65 ± 4.72 a	11.38 ± 0.84 b	1.23 ± 0.39 bc	219.98 ± 27.38 a-d	0.98 ± 0.25
Gümüşhane (10)	2.99 ± 0.35 ab	11.51 ± 1.15 fg	11.83 ± 0.34 ab	0.02 ± 0.01 de	201.97 ± 22.40 a-d	0.66 ± 0.19
Giresun (14)	4.71 ± 0.70 ab	19.12 ± 2.59 b-f	12.34 ± 0.67 ab	0.91 ± 0.33 b-d	183.55 ± 24.18 b-d	0.88 ± 0.14
Rize (9)	4.89 ± 1.28 ab	9.36 ± 2.10 fg	11.86 ± 1.49 ab	0.43 ± 0.11 c-e	263.29 ± 16.06 ab	0.48 ± 0.17
Ordu (86)	5.64 ± 0.71 ab	22.39 ± 1.26 a-f	9.90 ± 0.35 bc	0.22 ± 0.04 de	197.32 ± 7.65 a-d	0.80 ± 0.06
Artvin (16)	0.98 ± 0.26 b	2.73 ± 0.58 g	9.81 ± 0.55 bc	0.16 ± 0.03 de	134.93 ± 23.26 d	0.74 ± 0.14
Samsun (17)	4.38 ± 0.73 ab	8.76 ± 1.51 fg	7.17 ± 0.74 c	0.17 ± 0.08 de	137.11 ± 19.66 d	0.65 ± 0.13
Trabzon (12)	5.46 ± 0.69 ab	21.73 ± 2.80 a-f	10.24 ± 0.93 c	0.02 ± 0.01 de	183.78 ± 9.93 b-d	0.73 ± 0.15
Bayburt (2)	8.85 ± 1.25 a	13.95 ± 2.35 c-g	11.00 ± 2.20 bc	0.00 ± 0.00 e	185.55 ± 54.15 b-d	0.00 ± 0.00
Mean Value	4.83 ± 0.33 ab	19.97 ± 0.88	10.75 ± 0.23	0.34 ± 0.05	195.29 ± 5.28	0.71 ± 0.04
Min-Max	0.00-41.50	0.10-54.70	2.10-18.40	0.00-5.30	10.50-311.60	0.00-1.90
Confidence level (P)	<0.01	<0.01	<0.01	<0.01	<0.01	>0.05

Means with different letters in a same line are significantly different from one another (P<0.05, P<0.01)

was determined that K had been quantitatively the most important mineral with an average of 195.29 ppm. Studies from other geographical locations also revealed potassium to be the most abundant element [45, 46]. Our results showed statistically significant differences ($P< 0.01$) for Cu, Zn, Fe, Mn, Mg, Na, Ca and K in the individual groups (cities) of honey. In a similar study, the mean concentrations of elements were as follows: K 328, Na 19.52, Ca 42.1, Mg 33.12, Fe 2.9, Cu 0.34, and Mn 0.32 ppm for clover honey from Egypt [47], and the mean values for Na 98, K 653, Ca 88, Mg 38, Cu 0.62, Fe 5.3 and Mn 1.92 mg/kg were recorded in Spanish commercial honeys [46]; these levels of elements were higher than those from our study. Furthermore, in a previous study for honey from southeastern Ana-

tolia, concentrations for Na, K, Ca, Mg, Fe, Zn, Cu, Mn and Zn had been determined to be 118, 296, 51, 33, 6.6, 2.7, 1.8, 1.0 and 1.0 ppm, respectively [48].

3.2 Residue Analysis

The antibiotic residues in honey are sulphonamides, aminoglycosides (STR), TCs, amphenicols, macrolides, beta-lactams and nitrofuran metabolites. In this study, we only investigated the degree of sulphonamides, TCs and STR in honey samples according to CHARM II screening method. The results of this drug residues in locally produced honey are summarised in Table 4. Treatments with antibiotics are not allowed in the EU, while in many other countries they are widely used. Thus, in most EU countries, there are no MRL levels for antibiotics, which means that honey containing antibiotic residues are not permitted to be sold. As no residues are permitted, no MRL values are established [41].

TABLE 4 - The number and ratio of honey samples with antibiotic residues from Black Sea Region of Turkey.

Geographical origin of honey (sample number)	HONEY				
	ST+SL+TC (negative)	ST+SL+TC (only one kind of antibiotics)	ST	SL	TC
Tokat (7)	4	3	0	2	1
Çorum (6)	2	4	4	2	0
Bolu (4)	1	3	0	3	0
Bartın (4)	3	1	1	1	0
Zonguldak (4)	1	3	3	2	1
Karabük (2)	1	1	0	1	0
Amasya (2)	1	1	0	1	0
Kastamonu (7)	6	1	0	1	0
Düzce (4)	2	2	0	2	0
Sinop (4)	3	1	1	1	0
Gümüşhane (10)	7	3	0	3	0
Giresun (14)	8	6	1	5	1
Rize (9)	4	5	0	5	0
Ordu (86)	67	19	2	16	2
Artvin (16)	15	0	0	0	0
Samsun (17)	8	9	0	9	2
Trabzon (12)	7	5	1	4	0
Bayburt (2)	1	1	0	1	0
ALL (209)	141	68	13	59	7
The Ratio (%)	67.5	32.5	6.2	28.2	3.4

ST: Streptomycin; TC: Tetracyclines; SL: Sulphonamides

In the 209 honey samples, 13 of the samples have STR, 59 sulphonamide and 7 TC contents, and these samples were not appropriate according to TFC, EU, FAO/WHO CODEX standards. STR and TC residues are most abundant in Zonguldak honeys, but the sulphonamides are most in Bolu honeys. None of these drugs was found in honey samples produced in Artvin. Furthermore, 67% of the examined samples had no drug residues.

Honey must remain free of any chemical or biological contaminant to be safe for human consumption. However, some studies have reported the presence of pesticide residues in honey samples [49, 50]. The most commonly used pesticides are amitraz, cymiazole, bromopropylate, coumafos, flumethrin, fluvalinate, imidacloprid and fipronil [51, 52]. Although the regulatory agencies of several countries have established the MRL for some of these pesticides in honey samples, these limits are not included in the Codex Alimentarius [53]. The determination of contaminants and residues in honey and other bee products has become a growing concern in recent years, especially as these compounds may diminish the beneficial properties of honey, and if present in significant amounts, may pose a serious threat to human health [54]. Amitraz and flumethrin were detected in 44 (21%) and 73 (34.9%) samples at concentrations ranging from 57.9 to 167.4 ppb, and 20.9 to 38.6 ppb, respectively. The value obtained for amitraz is below the MRL standard value (200 ppb) determined by CODEX, TFC, and EU. Different national regulations have established MRLs permitted in honey, but the lack of homogeneity causes problems in international marketing and trade. As an example, Germany, Italy, and Switzerland have set MRLs for amitraz, bromopropylate, coumaphos, cyamizole, flumetrine, and fluvalinate, which oscillate between 10-100 µg/kg, (ppb) in Germany, between 5 and 500 µg/kg in Switzerland, and are of 10 µg/kg in Italy [5]. There is not an accepted regulation about flumetrine residue in ABD and EU Union [55]. However, the countries exporting honey to the European Union should be following the accepted limit (10 ppm) for drug residues when having not an MRL value [56]. Thirteen of all samples were contaminated by both of the acaricides. As a result of this study, we can conclude that most of the honey samples analysed are safe for human consumption. To avoid residues, pesticides should be used outside the bloom period or, at least, not during the foraging time of bees. Beekeepers can also avoid residues by placing their hives more than 3 km from agricultural plants treated with pesticides [41].

TABLE 5 - The number of honey samples with pesticide residues from Black Sea Region of Turkey.

Geographical origin of honey (sample number)	Amitraz		Flumethrin		A + F (both)	
	P	N	P	N	P	N
Tokat (7)	3	4	4	3	1	6
Çorum (6)	0	6	3	3	0	6
Bolu (4)	0	4	2	2	0	4
Bartın (4)	2	2	1	3	1	3
Zonguldak (4)	0	4	2	2	0	4
Karabük (2)	1	1	1	1	0	2
Amasya (2)	0	2	1	1	0	2
Kastamonu (7)	1	6	3	4	0	7
Düzce (4)	0	4	2	2	0	4
Sinop (4)	1	3	1	3	0	4
Gümüşhane (10)	0	10	4	6	0	10
Giresun (14)	5	9	6	8	1	13
Rize (9)	3	6	4	5	2	7
Ordu (86)	21	65	24	62	6	80
Artvin (16)	2	14	6	10	1	15
Samsun (17)	5	12	6	11	1	16
Trabzon (12)	0	12	3	9	0	12
Bayburt (2)	0	2	0	2	0	2
ALL (209)	44	165	73	136	13	196
The Ratio (%)	21.0	79.0	34.9	65.1	6.2	83.8

The results of naphthalene residues in samples are summarised in Table 6. It can be seen from the table, that the mean value for naphthalene residue for region honeys is 4.04±0.48 ppb. Twenty of all samples have naphthalene residues being higher than the determined values (10 ppb) in standards.

TABLE 6 - The naphthalene concentration (ng g^{-1}) in honey samples from Black Sea Region of Turkey.

Geographical origin of honey (sample number)	The mean concentration	Minimum	Maximum
Tokat (7)	4.26 ± 1.99	0.49	14.11
Çorum (6)	1.42 ± 1.99	0.58	1.89
Bolu (4)	3.30 ± 1.99	0.00	9.71
Bartın (4)	5.88 ± 1.99	0.00	21.90
Zonguldak (4)	12.30 ± 1.99	4.85	27.95
Karabük (2)	4.92 ± 1.99	0.58	9.25
Amasya (2)	2.98 ± 1.99	0.08	5.87
Kastamonu (7)	6.374 ± 1.99	0.14	17.64
Düzce (4)	3.37 ± 1.99	0.11	7.43
Sinop (4)	2.40 ± 1.99	0.00	5.71
Gümüşhane (10)	1.84 ± 1.99	0.00	9.34
Giresun (14)	9.44 ± 1.99	0.00	72.93
Rize (9)	3.13 ± 1.99	0.17	9.14
Ordu (86)	3.39 ± 1.99	0.00	22.36
Artvin (16)	3.85 ± 1.99	0.00	12.34
Samsun (17)	3.74 ± 1.99	0.00	12.64
Trabzon (12)	2.90 ± 1.99	0.00	16.86
Bayburt (2)	5.02 ± 1.99	0.62	9.41
Mean value	4.04 ± 1.99	0.00	72.93

Confidence level P>0.05

In this work, the principal physico-chemical parameters, such as ash, moisture, pH, acidity, diastase activity, HMF, electrical conductivity, invert sugar and sucrose, have been determined in 209 honey samples from the Black Sea Region of Turkey. Although the mean values for these parameters are appropriate according to standards, 24%, 8% and 12% of the samples are not suitable in terms of diastase activity, invert sugar and sucrose, respectively. The region honeys were evaluated as safe with respect to content of heavy metals. Furthermore, while the honey samples are appropriate for human consumption in terms of pesticide resdiues, 33 % and 10% of them are not suitable in terms of antibiotic and naphthalene residues, respectively, according to FAO/WHO CODEX, TFC and EU.

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REFERENCES

[1] Blasa, M., Candiracci, M., Accorsi, A., Piacentini, M. P., Albertini, M. C. and Piatti, E. (2006). Raw Millefiori honey is packed full of antioxidants. Food Chemistry, 97, 217–222.

[2] Anklam, E. (1998). A review of the analytical methods to determine the geographical and botanical origin of honey. Food Chemistry, 63, 549–562.

[3] Azeredo, L. da. C., Azeredo, M. A. A., De Souza, S. R. and Dutra, V. M. L. (2003). Protein contents and physicochemical properties in honey samples of Apis mellifera of different floral origins. Food Chemistry, 80, 249–254.

[4] Ouchemoukh, S., Louaileche, H. and Schweitzer, P. (2007). Physicochemical characteristics and pollen spectrum of some Algerian honeys. Food Control, 18, 52–58.

[5] Bogdanov, S., Lüllmann, C., Martin, P., von der Ohe, W., Russmann, H., Vorwohl, G., Oddo, L.P., Sabatini, A.G., Marcazzan, G.L., Piro, R., Flamini, C., Morlot, M., Lheretier, J., Borneck, R., Marioleas, P., Tsigouri, A., Kerkvliet, J., Ortiz, A., Ivanov, T., D'arcy, B., Mossel, B. and Vit, P. (1999). Honey quality, methods of analysis and international regulatory standards: review of the work of the International honey comission. Mitteilungen aus Lebensmitteluntersuchung und Hygiene, 90, 108–125.

[6] Gomes, S., Dias, L.G., Moreira, L.L. Rodrigues, P. and Estevinho, L. (2010). Physicochemical, microbiological and antimicrobial properties of commercial honeys from Portugal. Food and Chemical Toxicology, 48, 544–548.

[7] Kahraman, T., Buyukunal, S.K., Vural, A., Sandıkcı Altunatmaz, S., 2010. Physico-chemical properties in honey from different regions of Turkey. Food Chemistry 123, 41–44.

[8] Salter, R. (2003). Charm II System - Comprehensive Residue Analysis System For Honey. Apiacta, 38, 198–206.

[9] EU (2001). Council Directive 2001/110 relating to honey. Official Journal of the European Communities.

[10] Bogdanov, S., Martin, P. and Lullmann, C. (1997). Harmonized methods of the international honey commission [Extra issue] Apidologie, 1–59.

[11] AOAC, (1990). Official methods of analysis, 15th ed. In: K. Helrich (Ed.), Arlington, VA. USA.

[12] NP 1309:1976 (Ed. 1) – Mel – Determinação da acidez.

[13] AOAC, (1995). Official methods of analysis, 16th ed. Washington, DC.

[14] Winkler, O. (1955). Beitrag zum Nachweis und zur Bestimmung von Oxymethylfurfurol in Honig und Kunsthonig. Zeitschrift für Lebensmitteluntersuchung und -Forschung A102, 161–167.

[15] Lane, J.H. and Eynon, L. (1923). Determination of reducing sugars by means of Fehling's solution with methylene blue as internal indicator. Journal of the Indian Chemical Society, 42, 32–36.

[16] Silva, L.R., Videira, R., Monteiro, A.P., Valentão, P. and Andrade, P.B. (2009). Honey from Luso region (Portugal): Physicochemical characteristics and mineral contents. Microchemical Journal, 93, 73–77.

[17] Maver, L. and Poklukar, J. (2003). Coumaphos and amitraz residues in Slovenian honey. Apiacta, 38, 54–57.

[18] Tsimeli, K., Triantis, T.M., Dimotikali, D. and Hiskia, A. (2008). Development of a rapid and sensitive method for the simultaneous determination of 1,2-dibromoethane, 1,4-dichlorobenzene and naphthalene residues in honey using HS-SPME coupled with GC–MS. Analytica Chimica Acta, 617, 64–71.

[19] Finola, M. S., Lasagno, M. C. and Marioli, J. M. (2007). Microbiological and chemical characterisation of honeys from central Argentina. Food Chemistry, 100, 1649–1653.

[20] Sunay, A.E. (2006). Problem of Antibiotic Residues in Honey, Uludag Bee Journal, 4, 143–148.

[21] Terrab, A., Díez, M.J. and Heredia, F.J. (2002). Characterization of Moroccan Monofloral honeys by their physicochemical characteristics, Food Chemistry, 79, 373–379.

[22] Sahinler, N., Sahinler, S. and Gül., A. (2001). Hatay Yöresi Ballarının Bileşimi ve Biyokimyasal Analizi. Mustafa Kemal Üniversitesi Ziraat Fakültesi Dergisi, 6, 93–108.

[23] Gül, A. (2009). Türkiye'de Üretilen Bazı Balların Yapısal Özelliklerinin Gıda Güvenliği Bakımından Araştırılması. Doktora Tezi, M. K. Ü. Fen Bilimleri Enstitisü.

[24] Ashraf, M.W. and Akram, S. (2008) Physicochemical characteristics and heavy metal contents of Saudi Arabian floral honeys. Fresenius Environmental Bulletin, 17, 877-881.

[25] Malika, N., Mohamed, F. and Chakib, E. (2005). Microbiological and physico - chemical properties of Moroccan honey. International Journal of Agriculture & Biology, 7, 773–776.

[26] Yılmaz, H. and Küfrevioğlu, I. (2001). Composition of honeys collected from eastern and south-eastern Anatolia and effect of storage on hydroxymethylfurfural content and diastase activity. Turkish Journal of Agriculture and Forestry, 25, 347–349.

[27] Ünal, C. and Küplülü, Ö. (2006). Chemical quality of strained honey consumed in Ankara. Veterinary Journal of Ankara University, 53, 1–4.

[28] Şahinler, N. and Gül, A. (2004). Yayla ve Ayçiçek Ballarının Biyokimyasal Analizi. 4. Ulusal Zootekni Bilim Kongresi

[29] Omafuvbe, B. O. and Akanbi, O.O. (2009). Microbiological and physico-chemical properties of some commercial Nigerian honey. African Journal of Microbiol Research, 3, 891–896.

[30] Bogdanov, S. (2002). Harmonised Methods of the International Honey Commission, Swiss Bee Research Centre FAM, Liebefeld, CH-3003 Bern, Switzerland.

[31] Conti, M.E. (2000). Lazio region (central Italy) honeys: a survey of mineral content and typical quality parameters, Food Control, 11, 459–463.

[32] Fallico, B., Zappalà, M., Arena, E. and Verzera, A. (2004). Effect of conditioning on HMF content in unifloral honeys. Food Chemistry, 85, 305–313.

[33] Russo Almeida, P.A. (1997). Honey of transmontane Terra Quente. Some chemical parameters of honey from transmontane Terra Quente. Apicultor, 5, 29–35.

[34] Singh, N. and Bath, P. K. (1997). Quality evaluation of different types of Indian honey. Food Chemistry 58, 129–133.

[35] Küçük, M., Kolaylı, S., Karaoğlu, Ş., Ulusoy, E., Baltacı, C. and Candan, F. (2007). Biological activities and chemical composition of three honeys of different types from Anatolia. Food Chemistry, 100, 526–534.

[36] Özcan, M., Arslan, D. and Ceylan, D.A. (2006). Effect of inverted saccharose on some properties of honey. Food Chemistry, 99, 24-29.

[37] Fallico, B., Arena, E., Verzera, A. and Zappalà, M. (2006). The European Food Legislation and its impact on honey sector. Accreditation and Quality Assurance, 11, 49–54.

[38] Bogdanov, S., Ruoff, K. and Persano Oddo, L. (2004). Physicochemical methods for the characterisation of unifloral honeys: a review, Apidologie, 35, 4–17.

[39] Dag, A., Afik, O., Yeselson, Y., Schaffer, A. and Shafir, S. (2006). Physical, chemical and palynological characterization of avocado (Persea americana Mill.) honey in Israel. International Journal of Food and Science Technolgy, 41, 387–394.

[40] Mendes, E., Brojo, P. E., Ferreira, I. M. P. L. V. O. and Ferreira, M. A. (1998). Quality evaluation of Portuguese honey. Carbohydrate Polymers, 37, 219–223.

[41] Bogdanov, S. (2006). Contaminants of bee products. Apidologie, 37, 1–18.

[42] Silici, S., Uluozlu, Ö.D., Tuzen, M. and Soylak, M. (2008). Assessment of trace element levels in Rhododendron honeys of Black Sea Region, Turkey. Journal of Hazardous Materials, 156, 612–618.

[43] Turhan, K. (2007). Chemical contents and some trace metals of honeys produced in the middle Anatolia region of Turkey. Fresenius Environmental Bulletin, 16, 459-464.

[44] Tüzen, M. (2002). Determination of some metals in honey samples for monitoring environmental pollution. Fresenius Environmental Bulletin, 11, 366-370.

[45] Downey, G., Hussey, K., Kelly, J., Walshe, T. and Martin, P. (2005). Preliminary contribution to the characterisation of artisanal honey produced on the island of Ireland by palynological and physico-chemical data. Food Chemistry, 91, 347–354.

[46] Rodriguez-Otero, J.L., Paseiro, P., Simal, J. and Cepeda, A. (1994). Mineral content of the honeys produced in Galicia (North-west Spain). Food Chemistry, 49, 169–171.

[47] El-Sherbiny, G.A. and Rizk, S.S. (1979). Chemical composition of both clover and cotton honey produced in A.R.E. Egypt. Journal of Food Science, 7, 69–75.

[48] Yılmaz, H. and Yavuz, Ö. (1999). Content of some trace metals in honey from southeastern Anatolia. Food Chemistry, 65, 475–476.

[49] Kadar, A. and Faucon, J. (2006). Determination of traces of fipronil and its metabolites in pollen by liquid chromatography with electrospray ionization–tandem mass spectrometry. Journal of Agricultural and Food Chemistry, 54, 9741–9746.

[50] Pirard, C., Widart, J., Nguyen, B. K., Deleuze, C., Heudt, L., Haubruge, E., et al. (2007). Development and validation of a multi-residue method for pesticide determination in honey using on-column liquid–liquid extraction and liquid chromatography–tandem mass spectrometry. Journal of Chromatography A, 1152, 116–123.

[51] Korta, E., Bakkali, A., Berrueta, L. A., Gallo, B. and Vicente, F. (2001). Study of semiautomated solid-phase extraction for the determination of acaricide residues in honey by liquid chromatography. Journal of Chromatography A, 930, 21–29.

[52] Rial-Otero, R., Gaspar, E.M., Moura, I. and Capelo J.L. (2007). Chromatographic-based methods for pesticide determination in honey: An overview, Talanta, 71, 503–514.

[53] Codex Alimentarius, (1998). Draft revised for honey at step 6 of the Codex Procedure. CX 5/10.2, CL 1998/12-S.

[54] Kujawski, M.W. and Namieśnik, J. (2011). Levels of 13 multiclass pesticide residues in Polish honeys determined by LC-ESI-MS/MS. Food Control, 22, 914–919.

[55] Daş, Y.K., Kaya, S. (2004). The research of some synthetic pyrethroids insecticide residues in honey produced in Turkey, Journal of Vet. Microbiology, . Etlik, 2004,15(1-2):15-38

[56] Martin, P. Chem,C., Chem,M.A. (2002). Imports into the EU from third countries, veterinary and other requirements", European Federation of Honey Packers and Distributors. Third Caribbean Beekeeping Congress.

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