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Original Research

Determination of Heavy Metals in Honey Samples from Central Anatolia Using Plasma Optical Emission Spectrofotometry (ICP-OES)

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Abstract

In this study heavy metal concentrations in 34 different honey samples collected from different regions of Central Anatolia (and their environs) are determined using ICP-OES.

It is observed that the honey produced in Central Anatolia is good in quality, although they are not completely deprived of heavy metals. The contents of heavy metals in honey samples were found to be between 0.09-0.24 μgg^{-1} for Cd, 0.01-0.80 μgg^{-1} for Cu, 0.15-5.39 μgg^{-1} for Zn, 0.03-1.44 μgg^{-1} for Ni, 0.02-1.50 μgg^{-1} for Pb, 0.09 -1.89 μgg^{-1} for Cr, 0.02-1.56 μgg^{-1} for Mn, 0.57-8.74 μgg^{-1} for Fe and 0.00-0.58 μgg^{-1} for Se.

According to these results, it is concluded that the heavy metal concentrations in honey samples, except for the ones collected from the stations near settlement regions, are within acceptable parameters.

Keywords: ICP-OES, honey, heavy metal, pollution, Turkey

Introduction

Bee honey can be a good source of major and trace elements needed by humans. Their presence in human food is very important, but if they exceed safety levels, they can be toxic [1].

Besides the nutrient and medical characteristics of honey, it is used as a bio monitor to determine environmental quality in the environments which are polluted by heavy metals [2], radioactivity and pesticides [3, 4].

Jones reported that the honey bees could be a better indicator in monitoring environmental pollution because of the very low levels of trace elements in honey [5]. In addition, honey is a good indicator for the chemical constituents of the plants and their monitoring. Many researchers [6-11] have published studies about trace elements in honey.

Plasma Optical Emission Spectrometer (Inductively Coupled Plasma Optical Emmision Spectrofotometry = ICP-OES) is rather suitable for heavy metal determination and it is preferred by many research centres [12, 13].

According to different researchers, using the heavy metal contents of honey samples in determining the quality of the environment is accepted as a valid method. This study aims to determine the cadmium, nickel, zinc, copper, lead, chrome, manganese, iron and selenium contents of the honey samples which are produced in the different regions of central Anatolia province and to examine the determined results, whether they are in the acceptable borders or not from the point of view of human health.

Materials and Methods

Apparatus

A Varian Liberty Series II ICP-OES Spectrometer was used for metal determination. The instrument operating parameters for ICP-OES were: Rf power, 1.2 kW; Auxiliary flow 12 l/min; Nebulizer flow 0.75 l/min; The Standard

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one-piece torch; ultrasonic nebulizer type Glass concentric; and nebulization pressure, 160 kPa.

Reagents and Solutions

Standard stock solutions of different metal ions at a 1000 μg/ml concentration were prepared from atomic absorption spectroscopic grade chemicals and used to make working solutions by appropriate dilution. Reagent-grade nitric acid, double distilled water and the surfactant Merck were used.

Sample Preparation

Natural honey samples were collected from 34 different localities in Central Anatolia in 2004 (Table 1 and Fig. 1). Honey samples (1kg) were transferred to sterile jars. Ash concents were determined by heating 2.5 g of honey at 450ºC. The samples which had been converted to ash were dissolved in nitric acid (HNO_3). All samples were digested in duplicate, centrifuged and then made up to volume with 1% HNO_3 to 25 ml [14]. Determinations of the heavy metal concentrations in all samples were carried out by Inductively Coupled Plasma Optical Emission Spectroscopy (Varian Liberty II ICP-OES). The samples were analyzed in duplicate. An SPSS statistical program was used to calculate standard deviations and means.

Results and Discussion

Average heavy metal concentrations in the honey samples are given with their standard deviations in Tables 2, 3. It has been determined that the heavy metal contents of the samples decrease as the distance from settlement regions increases, as seen in Table 2. ANOVA test and Duncan Test as Post Hoc. are applied to the statistical analyses of the averages and it is found that our results are meaningful in the test according to P<0.05. The results of the test are given in Tables 2, 3.

When the results are examined, it is observed that the heavy metal contents of the honey samples, taken from the stations which are close to the settlement regions and pollution, are generally higher. As known, heavy metal pollution is in question at regions where human activity is present resulting from different origins, which include house waste, garbage and factors originating from traffic. Different ratios of heavy metals are seen in the plants which grow under these kinds of pollutants. High heavy metal concentrations in plant body can cause an increase in heavy metal concentrations in honey bees because bees collect pollens from different kinds of flowers.

The contamination sources of bee products are separated as environmental and apicultural in the study which is carried out by S. Bogdanov et al. The principal environmental factor is heavy metals. Plants, pesticides and

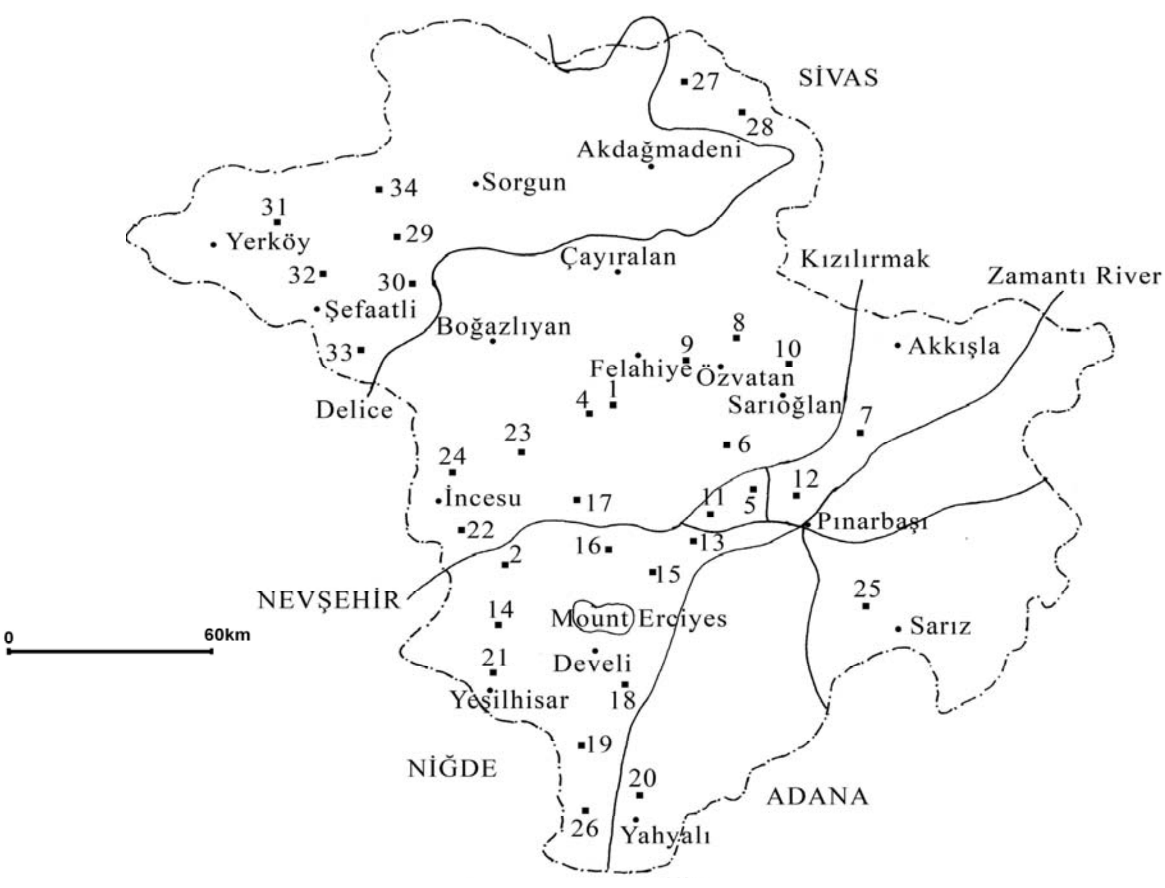

Fig. 1. Geographical distribution of the sampling points in Central Anatolia.

Table 1. The localities from which the honey samples are taken.

Region No.	Region that the honey is taken from	Distance to highway
1	Ebiç Village home garden (Erkilet)	2km
2	Sakar farm, mountain foot (Hacılar)	10km
3	Pınarbaşı-Pazarören	6km
4	Mahzemin City home garden (Erkilet)	3km
5	Entrance of Bünyan	to road 1.5k
6	Mountain foot of Kardeşler Village, near water(Bünyan)	4km
7	Özvatan 2 km to center, rocky field	2km
8	Taşlık village near highway 1km distance to Özvatan	50m
9	Büyüktoraman mountain foot (Felahiye)	2.5km
10	Küpeli road side Kabaktepe site (Özvatan)	500m
11	Gesi home garden away from highway(Melikgazi)	3km
12	Entrance of Pınarbaşı, near highway (Pınarbaşı)	500m
13	Erciyes Municipality mountain foot (Talas)	1km
14	Şeyhşaban Plateaus (İncesu)	6km
15	Erciyes Turkish World Forest	100m
16	Near roadside to Erciyes	10m
17	Kızılören Village Sarıgöl Plateau (İncesu)	20km
18	Develi road side on the top of hill	1km
19	Close to the roadside of Sındelhöyük (Develi)	100m
20	Entrance of Yahyalı, near highway	50m
21	Yeşilhisar home garden	3km
22	Roadside between Incesu- Yeşilhisar	100m
23	Yemliha Village mountain foot (Himmetdede)	3km
24	Yuvalı Village mountain foot (Himmetdede)	2km
25	Sarız Kıskaçlı Village Plateaues	6km
26	Yahyalı home garden, close to the roadside	2km
27	Yozgat- Sivas Overland route	200m
28	Yozgat- Sivas Overland route	50m
29	Yozgat centre	Distance to road 2 km
30	Yozgat Cehirlik Zone	9 km
31	Yozgat- Under the New İndustries	40m
32	Yozgat centre	Distance to road 10m
33	Yozgat Gülpınar Village, Şefaatli	20 km
34	Yozgat centre	150m

pathogens follow. The principal apicultural origin is varroacides and bee disease, wax moth, beehive materials and infections while harvesting follows [15].

In Table 2, when we examine the statistical results obtained for Zn, we see that there are differences between the stations. The highest concentration of Zn is measured at the value of 5,3906 μgg^{-1} at station 10. The most important reason for high pollution here is that it is close to the roadside and it is an agricultural area. It is reported that the most important sources that cause Zn pollution are fossil fuels, fertilizers and metal alloy [16]. According to the standards determined by the Codex Alimentarius Commission, the maximum Zn value that must be found in sweet nutrients such as sugar and honey is 5μgg^{-1} [17]. It is seen that the obtained values are within the given borders except for station 10.

Zinc values in honey samples have been reported in the range of 0.18-19.1 μgg^{-1}[11], 1.15-4.95 μgg^{-1} [9], 1.6-22.5 μgg^{-1} [18], 4.17-22.3 μgg^{-1} [8]. The range of zinc for the central Anatolia region is similar to that reported by Tuzen [9], for honey samples from the Black Sea Region of Turkey. It is seen that the values determined by the others for zinc is higher than the values obtained in this study.

The higher Cd concentrations were found as 0,24 μgg^{-1} in station 19. Cadmium is a non-essential toxic heavy metal that seriously threatens human health. The Turkish Food Codex determined the maximum Cd values that must be found in nutrients such as fruit juices and nectars as 0.03 μgg^{-1} [19]. It is seen that the values that are obtained from the stations for this element exceeds the given limits, and the samples suffer Cd pollution. The most important reason for this is that the stations are close to the roadside. The most important sources that cause cadmium pollution are fossil fuels from vehicles, metal business, plastics, house construction tools and sewers [16]. Cadmium contents of honey samples in the literature have been reported as 0.008-0.027 μgg^{-1} [8], 0,078-0,222μgg^{-1} [10], 0.005-0.009 μgg^{-1} [9], 0.008 μgg^{-1} [20], and <0.002-0.06 μgg^{-1} [21]. The range of cadmium for the Central Anatolia Region is similar to that reported by M.D. Ioannıdou et al. [10]. The level of cadmium of our samples was higher than some of the previous data [8, 9, 20, 21].

When Table 2 is examined for Pb, the highest value is seen in station 20 as 1,5062 μgg^{-1}. The most important reason for the high Pb concentration here can be considered vehicles because they are close to the roadside. It is reported that the most important sources of Pb pollution are vehicles, fossil fuels, metal business and refinery [16].

The maximum Pb value that must be found in sweet substances such as sugar and honey is determined as 0.3 μgg^{-1} by Codex Alimentarius Commission [17]. It is seen that the values obtained for this element from various stations exceeds given limits, and the samples suffer from Pb pollution. Lead data of honey samples around the world have been reported as 0.71-1.52 μgg^{-1} [14], 0.025-0.071μgg^{-1} [8], 0.03-0.05 μgg^{-1} [9], 0.03-0.24 μgg^{-1} [20] and 0.003-0.04 μgg^{-1} [21]. The range of lead in Central Anatolia Region is similar to that reported by Cerutti et al. [14]. The level of lead in our samples was higher than some of the previous data [8, 9, 21].

Table 2. Average Cd, Pb, Fe, Cu and Zn concentrations in the honey samples (μgg^{-1}) and their standard deviations.

Sample	Cd	Pb	Fe	Cu	Zn
1	0.18±0.01^{cd}	0.29±0.05^{bc}	1.40±0.02^{ab}	0.11±0.01^{b}	3.63±0.07^{bc}
2	0.19±0.01^{cd}	0.30±0.05^{bc}	1.24±0.02^{ab}	0.15±0.01^{b}	1.78±0.06^{ab}
3	0.19±0.01^{cd}	0.19±0.02^{b}	3.57±0.04^{ab}	0.19±0.04^{b}	4.64±0.22^{cd}
4	0.20±0.01^{cd}	0.49±0.01^{bcd}	2.52±0.02^{ab}	0.17±0.06^{b}	3.01±0.05^{b}
5	0.14±0.04^{b}	0.78±0.01^{cd}	3.74±0.01^{ab}	0.21±0.06^{b}	3.51±0.03^{bc}
6	0.19±0.02^{cd}	0.89±0.04^{cd}	2.03±0.06^{ab}	0.21±0.04^{b}	2.78±0.01^{b}
7	0.16±0.03^{bcd}	0.96±0.02^{cd}	5.46±0.02^{ab}	0.22±0.05^{b}	2.54±0.05^{ab}
8	0.16±0.04^{bcd}	0.42±0.08^{bc}	1.24±0.07^{ab}	0.72±0.08^{c}	4.53±0.01^{cd}
9	0.15±0.05^{bc}	0.46±0.04^{bcd}	5.30±0.04^{ab}	0.13±0.05^{b}	3.00±0.01^{b}
10	0.15±0.05^{bc}	0.16±0.05^{b}	3.65±0.04^{ab}	0.18±0.05^{b}	5.39±0.03^{d}
11	0.16±0.05^{bcd}	0.71±0.08^{cd}	5.56±0.01^{ab}	0.19±0.01^{b}	2.95±0.03^{b}
12	0.15±0.04^{bc}	0.12±0.05^{ab}	2.19±0.09^{ab}	0.37±0.01^{b}	3.93±0.01^{c}
13	0.17±0.02^{bcd}	0.26±0.05^{b}	3.24±0.05^{ab}	0.16±0.05^{b}	1.53±0.07^{ab}
14	0.16±0.02^{bcd}	0.39±0.01^{bc}	3.65±0.01^{ab}	0.16±0.07^{b}	2.36±0.04^{ab}
15	0.16±0.05^{bcd}	0.68±0.01^{bcd}	8.74±0.09^{b}	0.18±0.04^{b}	2.25±0.08^{ab}
16	0.14±0.04^{b}	0.58±0.04^{bcd}	2.65±0.08^{ab}	0.14±0.07^{b}	4.81±0.02^{cd}
17	0.09±0.01^{a}	0.02±0.01^{a}	0.57±0.01^{a}	0.01±0.01^{a}	1.29±0.01^{ab}
18	0.13±0.03^{b}	0.74±0.07^{cd}	5.02±0.09^{ab}	0.11±0.02^{b}	2.07±0.02^{ab}
19	0.24±0.06^{d}	0.62±0.03^{bcd}	5.90±0.05^{ab}	0.23±0.02^{b}	2.37±0.08^{ab}
20	0.17±0.08^{bcd}	1.50±0.10^{e}	5.96±0.08^{ab}	0.18±0.01^{b}	3.09±0.05^{b}
21	0.15±0.05^{bc}	0.64±0.02^{bcd}	3.49±0.01^{ab}	0.17±0.06^{b}	2.37±0.06^{ab}
22	0.14±0.04^{b}	0.33±0.06^{bc}	5.19±0.06^{ab}	0.21±0.05^{b}	2.19±0.02^{ab}
23	0.14±0.04^{b}	0.54±0.04^{bcd}	4.11±0.07^{ab}	0.19±0.03^{b}	4.93±0.07^{cd}
24	0.15±0.04^{bc}	0.60±0.07^{bcd}	5.21±0.08^{ab}	0.15±0.02^{b}	3.51±0.09^{bc}
25	0.17±0.09^{bcd}	0.99±0.03^{d}	5.10±0.05^{ab}	0.19±0.01^{b}	4.17±0.05^{c}
26	0.14±0.04^{b}	0.65±0.01^{bcd}	6.06±0.09^{ab}	0.16±0.05^{b}	2.37±0.04^{ab}
27	0.12±0.01^{ab}	0.27±0.02^{bc}	6.10±0.67^{ab}	0.51±0.03^{bc}	1.93d±0.06^{ab}
28	0.12±0.01^{ab}	0.34±0.02^{bc}	8.66±0.76^{b}	0.80±0.05^{c}	3.23±0.29^{b}
29	0.21±0.01^{cd}	0.25±0.03^{b}	4.55±0.39^{ab}	0.46±0.01^{bc}	1.15 ±0.03^{ab}
30	0.10±0,01^{ab}	0.23±0.01^{b}	3.96±0.24^{ab}	0.45±0.01^{bc}	0.71±0.02^{ab}
31	0,13±0.01^{ab}	0.38±0.09^{bc}	5.36±0.16^{ab}	0.67±0.01^{bc}	1.47 ±0.02^{ab}
32	0.11±0.01^{ab}	0.22±0.01^{b}	4.51±0.41^{ab}	0.49±0.01^{bc}	0.97 ±0.01^{ab}
33	0.11 ±0.01^{ab}	0.22±0.03^{b}	3.02 ±0.47^{ab}	0.45±0.01^{bc}	0.50 ±0.01^{ab}
34	0.12±0.01^{ab}	0.39 ±0.01^{bc}	6.32±0.52^{ab}	0.48±0.01^{bc}	0.15±0.02^{a}

For a given metal, mean concentrations followed by the same letter are not significantly different ($p<0.05$).

Table 3. Average Mn, Cr, Ni, and Se concentrations in the honey samples (μgg⁻¹) and their standard deviations.

Sample	Mn	Cr	Ni	Se
1	0.93±0.03 b	1.07±0.02 bc	0.99±0.05bc	0.06±0.01 c
2	1.05±0.01 bc	0.94±0.01 bc	0.94±0.01b	0.05±0.01 c
3	1.11±0.02 bc	1.10±0.01 bc	0.95±0.01 b	0.03±0.01 bc
4	1.56±0.09 c	0.89±0.05 b	1.19±0.05 bc	0.06±0.01 c
5	1.07±0.05 bc	0.89±0.06 b	0.99±0.06 bc	0.04±0.01 c
6	1.15±0.06 bc	0.85±0.08 b	0.88±0.01 b	0.09±0.01 cd
7	1.20±0.09 bc	0.76±0.03 b	0.94±0.06 b	0.06±0.02 c
8	1.03±0.03 bc	1.89±0.06 c	1.29±0.05 bc	0.03±0.01 bc
9	1.50±0.01 c	0.76±0.06 b	0.83±0.05 b	0.02±0.01 bc
10	1.33±0.07 bc	0.77±0.07 b	0.87±0.01 b	0.04±0.02 c
11	0.79±0.04 b	0.82±0.05 b	1.42±0.09 c	0.09±0.03 cd
12	1.27±0.02 bc	1.04±0.03 bc	1.05±0.09 bc	0.45±0.01 e
13	1.08±0.04 bc	0.82±0.08 b	0.86±0.06 b	0.08±0.01 cd
14	0.84±0.02 b	0.72±0.04 b	0.82±0.04 b	0.58±0.08 e
15	0.95±0.01 b	0.80±0.04 b	0.82±0.09 b	0.08±0.01 cd
16	0.86±0.02 b	0.79±0.01 b	0.83±0.04 b	0.01±0.01 b
17	0.02±0.01 a	0.09±0.01 a	0.03±0.01 a	0.006±0.01 a
18	1.13±0.03 bc	0.95±0.04 bc	0.98±0.01 bc	0.01±0.01 b
19	0.83±0.01 b	0.89±0.06 b	0.77±0.07 b	0.04±0.01 c
20	0.78±0.08 b	0.78±0.06 b	0.93±0.03 b	0.02±0.01 b
21	0.91±0.03 b	0.77±0.02 b	0.81±0.02 b	0.05±0.01 c
22	0.99±0.05 b	0.99±0.05 bc	1.44±0.07c	0.07±0.01 c
23	1.41±0.01 bc	0.77±0.02 b	0.79±0.03 b	0.05±0.01 c
24	0.95±0.02 b	0.83±0.09 b	0.89±0.07 b	0.05±0.01 c
25	0.58±0.02 b	0.81±0.03 b	0.94±0.01 b	0.08±0.01 cd
26	1.23±0.07 bc	0.89±0.07 b	0.91±0.09 b	0.06±0.01 c
27	0.74±0.21 b	0.16±0.01 ab	0.76±0.01 b	0.09±0.08 cd
28	0.82±0.01 b	0.16±0.01 ab	0.79±0.01 b	0.03±0.01 bc
29	0.74±0.10 b	0.16±0.01 ab	0.79±0.01 b	0.05±0.01 c
30	0.67±0.15 b	0.15±0.01 ab	0.76±0.01 b	0.02±0.01 b
31	1.01±0.15 bc	0.16±0.01 ab	1.01±0.03 bc	0.11±0.01 d
32	0.94±0.07 b	0.16±0.01 ab	0.79±0.01 b	0.10±0.01 d
33	0.24±0.39 b	0.15±0.01 ab	0.70±0.01 b	0.01±0.01 b
34	0.87±0.09 b	0.16±0.01 ab	0.76±0.01 b	0.09±0.04 cd

For a given metal, mean concentrations followed by the same letter are not significantly different ($p<0.05$).

The lower and higher copper concentrations were found as 0.018 μgg^{-1} in the honey sample from Kayseri, Kızılören and 0.82 μgg^{-1} in the honey sample from Yozgat-Sivas overland route, respectively. The most important reasons for the high Cu concentration in this station can be that this station is a home garden, apiculture, and animal breeding are done together and also it is very close to the highway. In addition to these results, the most important sources of Cu pollution are indicated as animal fertilizers, pesticides, sewage, ash, metal business and iron and steal industry [16]. According to the standard values determined by the Codex Alimentarius Commission; the maximum Cu value that must be found in sweet nutrients such as sugar and honey is reported as 5 μgg^{-1} [17]. The values that are obtained in this study do not exceed these limits. Copper values in the literature have been reported as 0.25-1.30 μgg^{-1} [9] for honey samples from the Black Sea (Turkey), 1.8μgg^{-1} [7] from southeastern Anatolıa of Turkey, 0.31 μgg^{-1} [22] and for Lazio region (central Italy) honeys. The copper levels of our samples are lower than those reported by Yılmaz for honey samples from southeastern Anatolia [7].

When Table 2 is examined for Fe, the highest value is found at station 15 as 8,7431μgg^{-1}. The reason for high Fe values here can result from soil and plants. The most important sources of Fe pollution are indicated as metal corrosion, digging and drilling [16]. According to the standard values determined by Codex Alimentarius Commission; the maximum Fe value that must be found in sweet nutrients such as sugar and honey is reported as 15μgg^{-1} [17].

The values that are obtained in this study do not exceed these limits. Iron values in honey samples have been reported in the range of 0.40-52.51 μgg^{-1} [11], 3.45-8.94 μgg^{-1} [9], 0.97-1.91 μgg^{-1} [20]. The values for the iron contents in our samples are generally at the same level as in the values cited in literature [9, 11, 20].

The higher Mn concentrations were 1.56 μgg $^{-1}$ found in station 4. Some reported manganese values in the literature for honey were 0.32-1.70 μgg^{-1} [9], 0.11-7.22 μgg^{-1} [10]. Manganese values found in the present study are in agreement with the manganese levels of honey samples from the Black Sea Region in Turkey [9], and Greece [10].

When Table 3 is examined for Cr, the highest value is seen in station 8 as 1.89 μgg^{-1}. Reported chromium values in the literature for honey were 0.010–0.10 μgg^{-1} [23], 0.043–1.07 μgg^{-1} [6]. The values for the Cr contents in our samples are generally at the same level as in the literature values [6, 23].

The lower and higher nickel concentrations were found as 0.038 μgg^{-1} in the honey sample from Kayseri, Kızılören and 1.43 μgg^{-1} in the honey sample from Kayseri, Yeşilhisar. Nickel values in the literature have been reported as 0.23–0.27 μgg^{-1} [24], for the honey samples from Italy. The nickel levels in our samples are higher than that reported by Caroli [24].

The higher Se concentrations were found as 0.58 μgg^{-1} in station 14. The reason for high Se values here can result from soil and plants (for example like *Astragalus sp*. L.).

Conclusions

The honeys in the beehives located close to the settlement regions can be exposed to home, industrial and traffic-originated pollutants. Therefore, making the apiculture activities away from the pollution threats is necessary. In conclusion, it is determined that the honey samples produced in Central Anatolia do not completely lack heavy metals, but they are at acceptable limits for some elements.

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