# Levels of Chromium, Copper, Iron, Magnesium, Manganese, Selenium, Zinc, Cadmium, Lead and Aluminium of honey varieties produced in Turkey

Sema Sandıkcı ALTUNATMAZ<sup>1\*</sup>, Duygu TARHAN<sup>2</sup>, Filiz AKSU<sup>1</sup>, Nural Pastacı OZSOBACI<sup>2</sup>, Mehmet Erman OR<sup>3</sup>, Umit Bora BARUTÇU<sup>2</sup>

#### Abstract

(cc) BY

Honey is a natural animal product. The elemental composition of honey varies greatly depending on the source of nectar, honeydew, pollen and environmental conditions. The aim of the present study was to determine the levels of chromium (Cr), copper (Cu), iron (Fe), magnesium (Mg), manganese (Mn), selenium (Se), zinc (Zn), cadmium (Cd), lead (Pb) and aluminium (Al) in a total of 65 honey samples procured from different regions of Turkey. Element levels were measured by using an inductively coupled plasma optical emission spectrophotometer (ICP-OES). Minimum and maximum levels of the elements, indicated as µg/g wet weight were as follows: Cr: 0.126-7.964, Cu: 0.223-198.361, Fe: 3.506-1278.778, Mg: 5.830-309.783, Mn: 0.096-29.496, Se: 0.418-19.879, Zn: 1.734-245.205, Cd: 0.000-0.297, Pb: 0.000-3.035, Al: 0.775-155.585.

Keywords: trace element; toxic element; heavy metal; ICP-OES; honey.

**Practical Application:** Honey contains many elements necessary for life. Alongside its nutritional value, honey may also contain toxic elements.

#### 1 Introduction

Turkey ranks second among the other honey producing countries with an annual (2017) production of 114.471 tons (Turkish, 2017; Kaftanoglu, 2017). Botanical and geographical origin of honey is of great importance in terms of its chemical quality (Bargańska et al., 2016; Kılıç Altun et al., 2017). Turkey, owing to its rich plant diversity and favorable climate due to geographical features comes to the forefront in terms of production of high quality honey varieties with high mineral content (Kılıç Altun et al., 2017). Pine honey which is rich in mineral content is mainly produced in Turkey. Honeydew honey is known to have a higher mineral content (0.6-2%) in comparison with blossom honey (0.1-0.5%) (Mutlu et al., 2017). Honey contains high levels of potassium, sodium, calcium and magnesium, the sources of which were shown to be pollen and nectar (Kılıç Altun et al., 2017; Turhan, 2007; Batista et al., 2012; Fernández-Torres et al., 2005). Apart from the above mentioned minerals, honey may contain Fe, Cr, Se, Cu, Mn, Zn, Al, Pb, As, Cd and Hg, depending mainly on environmental factors (Kiliç Altun et al., 2017; Mutlu et al., 2017; Fernández-Torres et al., 2005). Environmental factors may be listed as soil, air, water and environmental pollution as well as the contaminated materials, tools and equipment used in beekeeping. Therefore, honey is recognized as one of the indicators of environmental pollution (Mutlu et al., 2017; Turhan, 2007).

Different descriptions have been defined for the metals currently known as heavy metals which have deleterious effects on human health. A heavy metal is essentially defined as a metal of relatively high density (above 5g/cm<sup>3</sup>). In medicine, the concept is used for metals with toxic properties regardless of their atomic weights. Although more than sixty elements were identified as heavy metals, the most frequently encountered and the most pronounced heavy metals are Hg, Mn, Fe, Co, Ni, Cu, Zn, Cd, As, Sn, Pb, Ag and Se. Heavy metals are accumulated particularly in certain organs, ultimately reaching up to toxic levels (Yılmaz-Aksu & Sandikci-Altunatmaz, 2017; Jaishankar et al., 2014). Some of them, for instance zinc, selenium and copper are essential for maintaining metabolic activities, however may elicit toxic effects at high concentrations. Elements which are required in minor quantities for physiology of organism are referred to as trace elements. In this regard, Fe, Cu, Se, Zn, Ni and Mn are recognized as trace elements which are needed by the organism for maintaining vital activities (Kılıç Altun et al., 2017). It has been indicated in research studies that honey may contain several minerals, trace elements and heavy metals at various concentrations, which differ according to regional and environmental conditions, seasonal and annual differences, as well as beekeeping and agricultural techniques applied (Batista et al., 2012; Fernández-Torres et al., 2005).

The objective of the present study was to determine the elemental composition of different honey samples consumed in Turkey by taking into account the regional differences. The results were considered to assist the establishment of legal permissible limits for certain minerals, trace elements and toxic heavy metals that may be found in honey and as well as to contribute to the assessment of the levels of trace elements from a nutritional perspective.

Received 06 June, 2018

Accepted 02 Oct., 2018

<sup>&</sup>lt;sup>1</sup>Food Technology Programme, Food Processing Department, Veterinary Medicine Vocational High School, Istanbul University-Cerrahpasa, Avcilar, Istanbul, Turkey

<sup>&</sup>lt;sup>2</sup>Department of Biophysics, Cerrahpasa Medicine Faculty, Istanbul University-Cerrahpasa, Cerrahpasa, Istanbul, Turkey

<sup>&</sup>lt;sup>3</sup>Department of Internal Medicine, Faculty of Veterinary Medicine, Istanbul University-Cerrahpasa, Avcilar, Istanbul, Turkey

 $<sup>*</sup> Corresponding \ author: \ sandikci@istanbul.edu.tr$ 

#### 2 Materials and methods

#### 2.1 Sample collection

Honey samples were collected from areas of intensive beekeeping. Measurements for pre-determined minerals, trace elements and heavy metals were carried out in a total of 65 honeys. These samples were obtained from different regions of Turkey representing; Central Anatolia, the Black Sea, Eastern Anatolia, Aegean, Marmara, the Mediterranean, and Southeastern Anatolia (Table 1).

#### 2.2 Preparation of samples for measurements

Honey samples were placed into tared tubes, weighed using a precision balance and the results were recorded. 2 mL of nitric acid (HNO<sub>3</sub>) was added to honey samples placed into a heat-resistant graduated tubes and left to dissolve in a drying oven at 150 °C. The honey and nitric acid mixture was left to cool at room temperature and then 1 mL of perchloric acid (HClO<sub>4</sub>) was added into the mixture which was returned to oven and subjected to wet ashing. Finally, all samples were vortexed and distilled water was added into the tubes to a total volume of 12 mL and vortexed once again getting ready to conduct elemental analysis in an ICP-OES device.

## 2.3 Measurements of elements

Suitable wavelengths of 267.716, 324.754, 259.940, 285.213, 257.610, 196.090, 206.200, 228.802, 220.353 and 167.079 nm were used for the analysis of Cr, Cu, Fe, Mg, Mn, Se, Zn, Cd, Pb and Al respectively in an ICP-OES device (Thermo ICAP 6000 series) (Ali et al., 1988). Samples were subjected to wet ashing and prepared for measurements. Four samples were prepared from each honey type and mean values were calculated based on the results of elemental analysis.

## 3 Results and discussion

Levels of macroelement, Mg; microelements Cr, Cu, Fe, Mn, Se and Zn and toxic elements (heavy metals) Cd, Pb and Al were determined in a total of 65 honey samples. The results of the analyses were presented in Table 2 as  $\mu g/g$  wet weight. Minimum, maximum and mean values of the results were shown in Table 3 as  $\mu g/g$  honey.

On the basis of the findings, it may be assumed that lead and cadmium contamination was prevalent in honey samples. Out of 65 samples, only 5 and 2 honeys did not contain cadmium and lead, respectively. Cadmium free honeys were from the

Table 1. Regional distribution of honey samples.

Region	Honey Sample No
Central Anatolia	1-2-3-6-41-62-63-64
Black Sea	4-5-7-8-9-10-20-24-38-43-47-49-50-53-54-57-58
Eastern Anatolia	11-21-25-26-30-31-33-35-40-44-45-55-59-61-65
Aegean	13-17-34-39-42-46
Marmara	12-14-15-16-18-27-28-29-32-37-56
Mediterranean	19-22-23-36-51-52-60
Southeastern Anatolia	48

Black Sea (2 samples) and Thrace regions, the cities of Kars and Mus; whereas lead free honeys came from the Thrace region and Kars city. Cadmium concentration was detected to be below the level of 0.01  $\mu$ g/g in 7 samples while lead levels exceeded 0.01 µg/g in all samples that contained lead. Mean lead concentration was found to be  $0.349 \,\mu\text{g/g}$  and the highest lead level of 3.035 µg/g was detected only in one sample. Lead concentrations of other honey samples from the same city were 0.356; 1.586; 0.715 and 0.044  $\mu$ g/g, which was considered to be associated with subsequent contamination through water, tinware and etc. Lead is a naturally occurring element; however emerges as an environmental pollutant at excessive amounts by means of several factors such as mining, battery manufacturing, brazing, water pipe production, usage of paint and gasoline (Yılmaz-Aksu & Sandikci-Altunatmaz, 2017). It is worldwide proposed that all bee products, particularly honey must be free of any foreign substances in order to be considered valuable and beneficial (Cukur et al., 2016). Lead, cadmium, arsenic and mercury are involved in the list of toxic substances published by the Agency for Toxic Substances and Disease Registry (The Agency for Toxic Substances and Disease Registry, 2015). The Joint FAO/WHO Expert Committee on Food Additives (JECFA) withdrew the previously suggested provisional tolerable weekly intake (PTW1) level for lead in 2011 and indicated that there is no such threshold limit value that can be adopted as healthy (World Health Organization, 2011). Therefore, it is of utmost importance that honey is by no means contaminated with any heavy metals including lead. There are several studies available worldwide and also in Turkey with respect to the determination of lead and other elemental concentrations in honey (Kılıç Altun et al., 2017; Turhan, 2007; Batista et al., 2012; Fernández-Torres et al., 2005; Nascimento et al., 2015; Vanhanen et al., 2011; Doker et al., 2014; Tuzen et al., 2007; Kolayli et al., 2008; Sireli et al., 2015; Czipa et al., 2015; Chua et al., 2012; Liberato et al., 2013; Čelechovská & Vorlová, 2001; Caroli et al., 1999; Golob et al., 2005; Bontempo et al., 2017; Bilandžić et al., 2017; Formicki et al., 2013; Rashed & Soltan, 2004).

In the present study, minimum and maximum levels of 0.126  $\mu$ g/g and 7.964  $\mu$ g/g for chromium were found in Kars and Artvin honeys, respectively. In other studies carried out in Turkey, chromium levels in honey were indicated as <1 ppb, 88.1 ng/g (Doker et al. 2014) and 2.4-37.9 µg/g (Tuzen et al., 2007) and in one study chromium was below the detection limit (No Detection, ND) in selected honey samples (Kolayli et al., 2008). In worldwide studies, chromium levels were detected as 4.80-36.7 µg/g, 1.845-3.835 mg/kg, 0.023-0.170 µg/kg, 1.03-3.93 ng/g, 0.78-3.55 mg/kg, 0.0-0.0 mg/kg and 2.69-49.9 mg/kg in Hungary (Czipa et al., 2015), Malaysia (Chua et al., 2012), Brazil (Liberato et al., 2013), Czech (Čelechovská & Vorlová, 2001), Italy (Caroli et al., 1999), Slovenia (Golob et al., 2005), Italy (Bontempo et al., 2017) and Croatia (Bilandžić et al., 2017) respectively. The amount of chromium in honey is strongly affected by geographical, floral, environmental and geological factors. Furthermore, it was assumed that stainless steel surfaces might have been responsible for high levels of chromium contamination (Doker et al., 2014).

Table 2. Mean values of the elements analyzed in honey samples ( $\mu g$	/g honey).
--	------------

No	Cr	Cu	Fe	Mg	Mn	Se	Zn	Cd	Pb	Al
1	6.635	4.941	117.318	152.824	1.129	6.706	16.729	0.282	3.035	49.624
2	1.305	4.365	566.769	41.314	1.451	0.910	5.116	0.040	0.356	25.385
3	2.700	2.871	135.043	78.514	0.814	3.343	8.357	0.086	1.586	36.900
4	2.320	3.400	81.720	91.840	0.720	1.680	21.000	0.120	0.840	110.880
5	1.320	1.350	61.800	38.850	0.390	2.340	7.080	0.060	1.050	40.380
6	1.615	1.477	19.777	40.869	0.300	2.192	7.038	0.046	0.715	35.885
7	1.521	1.436	141.000	57.086	0.771	1.907	5.421	0.043	1.050	33.279
8	2.032	2.067	30.986	86.055	11.465	2.376	5.596	0.297	0.154	1.592
9	7.964	27.382	411.655	103.091	1.636	4.964	35.291	0.164	1.800	8.400
10	1.148	1.156	37.679	61.760	0.530	1.936	4.807	0.007	0.088	3.983
11	4.629	3.257	115.303	140.743	2.331	4.457	18.857	0.000	0.960	13.097
12	2.924	2.858	70.364	202.233	4.560	3.840	13.549	0.022	0.524	8.487
13	0.285	0.986	12.722	15.142	0.132	0.915	4.332	0.010	0.122	11.786
14	1.059	3.362	32.374	122.294	5.462	2.179	6.150	0.009	0.309	4.262
15	1.539	2.764	25.961	148.386	1.349	1.705	6.331	0.000	0.174	4.452
16	1.888	1.438	27.400	74.300	0.438	1.825	10.813	0.013	0.188	7.200
17	1.319	3.891	57.742	161.366	2.169	1.461	11.672	0.007	0.060	15.980
18	0.812	0.889	17.816	55.426	0.275	1.525	3.656	0.007	0.085	5.619
19	0.545	0.669	13.324	35,217	0.392	0.992	4.983	0.014	0.069	5.197
20	0.779	2.862	13.324	134,690	21.076	1.269		0.010	0.009	155.586
20	0.779	0.351	9.320	134,690	0.194		11.848 2.220	0.000		3.581
						0.678			0.104	
22	0.282	1.444	8.395	14.699	1.140	0.662	7.482	0.005	0.152	6.174
23	5.326	9.684	80.505	28.295	0.842	1.916	39.137	0.021	0.253	12.611
24	5.120	1.394	76.366	51.840	2.560	0.971	18.274	0.011	0.309	14.823
25	0.914	0.700	13.314	30.571	0.271	1.671	14.014	0.014	0.357	29.586
26	0.794	5.848	21.718	25.690	0.338	1.099	14.096	0.000	0.439	41.037
27	0.806	1.110	16.442	49.039	0.430	1.075	4.925	0.018	0.143	20.812
28	0.538	0.766	6.610	16.097	0.114	0.476	6.083	0.021	0.000	10.459
29	0.863	0.327	11.623	21,332	0.208	0.714	5.296	0.010	0.099	7.567
30	0.682	0.567	20.712	36.879	0.173	0.863	1.734	0.016	0.123	2.877
31	0.447	0.404	17.498	33.327	0.185	1.058	3.905	0.000	0.305	22.451
32	0.185	0.223	9.761	16.125	0.096	0.418	2.331	0.003	0.069	7.505
33	0.408	0.400	5.224	16.992	0.128	0.872	2.104	0.016	0.320	13.536
34	0.752	4.352	16.640	50.704	0.432	1.200	10.608	0.032	0.240	33.664
35	0.126	7.529	3.506	12.207	0.151	0.496	7.904	0.008	0.000	3.364
36	0.240	2.741	37.277	60.836	1.022	0.542	5.543	0.039	0.031	92.903
37	1.418	3.678	120.031	52.769	1.060	3.319	5.283	0.094	0.327	11.610
38	0.672	0.561	14.020	9.963	0.187	1.960	2.736	0.033	0.094	1.817
39	2.046	3.010	94.190	50.597	0.826	2.597	7.770	0.079	0.138	10.938
40	1.296	1.147	66.018	20.286	1.191	2.470	4.756	0.026	0.131	4.923
41	0.372	11.984	46.455	12.071	0.210	1.461	14.297	0.012	0.048	1.921
42	0.345	1.248	41.028	42.092	0.679	2.051	1.985	0.011	0.088	2.382
43	1.035	6.066	41.659	14.059	0.495	2.587	84.330	0.044	0.319	3.457
14	0.506	2.176	31.104	7.862	0.146	1.711	3.824	0.018	0.068	0.808
15	0.911	33.482	315.214	15.086	1.211	2.239	37.093	0.096	0.407	6.043
16	0.594	10.885	44.170	32.800	0.479	19.879	12.830	0.018	0.224	3.745
ŧ7	1.493	3.224	1278.779	50.913	29.496	3.151	6.357	0.027	0.330	5.405
19 18	0.689	2.074	18.222	17.902	0.203	2.123	6.382	0.018	0.283	2.332
19 19	0.931	20.620	39.714	12.824	0.203	2.125	30.702	0.010	0.212	2.694
50	0.670	7.886	25.645	51.605	18.696	2.294	9.321	0.041	0.064	1.784
50	1.333	1.600	159.911	18.800	0.498	2.996	5.324	0.021	0.004	1.784
51 52	0.497				0.498					1.449
14	0.49/	4.552	7.850	5.830	0.099	1.622	7.920	0.021	0.017	1.10/

Table 2. Continued...

No	Cr	Cu	Fe	Mg	Mn	Se	Zn	Cd	Pb	Al
54	1.077	1.738	25.144	20.938	0.325	1.996	3.914	0.045	0.269	2.007
55	0.667	7.052	22.308	13.463	0.310	1.625	15.101	0.027	0.155	1.962
56	0.483	2.432	89.233	21.180	0.329	1.489	5.134	0.018	0.086	1.101
57	0.918	7.667	196.349	21.987	0.926	1.973	19.643	0.024	0.129	2.923
58	0.592	13.232	8.841	18.608	0.151	1.491	13.647	0.016	0.140	0.775
59	0.740	3.413	23.880	9.993	0.200	2.000	6.827	0.027	0.160	4.427
60	2.154	198.361	87.716	47.133	0.607	3.267	245.205	0.058	0.333	2.169
61	0.645	22.545	18.105	18.795	0.255	1.350	28.103	0.030	0.210	2.168
62	1.420	29.561	500.341	22.171	1.302	2.722	27.556	0.044	0.673	4.215
63	0.551	189.031	271.361	20.394	0.619	0.612	202.639	0.025	0.346	1.571
64	0.613	10.489	114.993	16.368	0.345	1.494	10.049	0.060	0.354	2.849
65	0.885	2.872	876.393	8.361	1.761	1.495	5.331	0.030	0.315	3.895

Table 3. Minimum, maximum and mean values for each element analyzed in honey samples ( $\mu g/g$  honey).

Elements	Cr	Cu	Fe	Mg	Mn	Se	Zn	Cd	Pb	Al
Min.	0.126	0.223	3.506	5.830	0.096	0.418	1.734	0.000	0.000	0.775
Max.	7.964	198.361	1278.779	309.783	29.496	19.879	245.205	0.297	3.035	155.586
Mean ± SE	$1.364 \pm 1.27$	$11.030\pm10.03$	$107.824\pm20.02$	$50.548\pm7.59$	$1.985\pm3.56$	$2.183 \pm 1.67$	$18.201\pm9.12$	$0.038 \pm 0.27$	$0.349 \pm 0.81$	$15.299\pm6.75$

Mean ± SE: Mean ± Standard Error.

In the present study, minimum 0.223  $\mu$ g/g and maximum 198.361 µg/g levels of copper were detected in Thrace honey and Muğla pine honey, respectively. Pine honey is known to have higher total mineral content in comparison with other types of honey. Copper levels in honeys were reported as <1-929 ppb (68.5 mean) (Kılıç Altun et al., 2017), 0.70-4.12 mg/kg (1.90 mean) (Turhan, 2007), 0.12-0.25 µg/g (0.17 mean) (Doker et al. 2014), 0.23-2.41 µg/g (Tuzen et al., 2007), 1.2-2.2 µg/g (Kolayli et al., 2008), 0.011-0.098 mg/kg (0.06 mean) (Sireli et al., 2015) in other studies conducted in Turkey. Levels of copper content were detected as 0.531-2.117 mg/kg, 0.046-0.236 mg/kg, 0.07-1.29 mg/kg, 0.057-1.55 mg/kg, 144-216 ng/g, 1.4-2.70 mg/kg, 0.1-0.6 mg/kg, 0.07-2.14 mg/kg and 1.0-1.75 mg/kg in Spain (Fernández-Torres et al., 2005), Malaysia (Chua et al., 2012), Brazil (Liberato et al., 2013), Czech (Čelechovská & Vorlová, 2001), Italy (Caroli et al., 1999), Slovenia (Golob et al., 2005), Italy (Bontempo et al., 2017), Croatia (Bilandžić et al., 2017) and Egypt (Rashed & Soltan, 2004), respectively.

In the present study, minimum and maximum iron levels of  $3.506 \ \mu g/g$  and  $1278.779 \ \mu g/g$  were detected in Kars and Bolu honeys, respectively. In other studies carried out in Turkey, iron levels in honey samples were found to be <1-7254.62 ppb (268 mean) (Kiliç Altun et al., 2017), 0.84-18.21 mg/kg (7.95 mean) (Turhan, 2007), 9.70-11.60  $\ \mu g/g$  (10.5 mean) [15], 1.1-12.7  $\ \mu g/g$  (Tuzen et al., 2007), 3.2-6.7  $\ \mu g/g$  (Kolayli et al., 2008), and 13.45-97.30 mg/kg (41.13 mean) (Sireli et al., 2015). Iron levels in honeys in other countries such as Hungary (Czipa et al., 2015), Malaysia (Chua et al., 2012), Brazil (Liberato et al., 2013), Italy (Caroli et al., 1999), Italy (Bontempo et al., 2017), Croatia (Bilandžić et al., 2004) were detected as <0.005-2.86 mg/kg, ND-32.480 ppm, 0.12-8.76 mg/kg, 191-651 ng/g, 0.5-3.1 mg/kg, 1.03-2.4 mg/kg, 0.08-0.24 \ \mu g/g x 10<sup>2</sup> and 58-202 mg/kg, respectively.

In the study, minimum 5.830  $\mu$ g/g and maximum 309.783  $\mu$ g/g levels of magnesium were found in honey samples collected from Rize and Muğla cities, respectively. Other studies from Turkey revealed levels of 23-64 mg/kg (39.10 mean) (Turhan, 2007) and 10.90-93.90  $\mu$ g/g (26.7 mean) (Doker et al., 2014) for magnesium in honeys analyzed. Magnesium levels in honey samples from Spain (Fernández-Torres et al., 2005), Hungary (Czipa et al., 2015), Malaysia (Chua et al., 2012), Brazil (Liberato et al., 2013), Italy (Bontempo et al., 2017), Croatia (Bilandžić et al., 2017), Poland (Formicki et al., 2013) and Egypt (Rashed & Soltan, 2004) were detected to be 13.26-74.38 mg/kg, <0.104-35.1 mg/kg, 5.209-89.502 ppm, 2.48-28.33 mg/kg, 50-79.0 ppm, 11.0-195 mg/kg, 0.42-0.86  $\mu$ g/g × 10<sup>2</sup> and 102-300  $\mu$ g/g, respectively.

In the present study, minimum and maximum manganese levels of 0.096  $\mu$ g/g and 29.496  $\mu$ g/g were detected in Bolu and Thrace honeys, respectively. In other studies carried out in Turkey, manganese levels in honey samples were found to be <1-274 mg/kg (45.6 mean) (Kılıç Altun et al., 2017), 0.47-2.60 mg/kg (1.13 mean) (Turhan, 2007), 0.04-0.25 µg/g (0.13 mean) (Doker et al., 2014), 0.32-4.56 µg/g (Tuzen et al., 2007) and 1.2-17.20 µg/g (Kolayli et al., 2008). Manganese levels in honey samples from Spain (Fernández-Torres et al., 2005), Hungary (Czipa et al., 2015), Malaysia (Chua et al., 2012), Brazil (Liberato et al., 2013), Italy (Caroli et al., 1999), Slovenia (Golob et al., 2005), Italy (Bontempo et al., 2017), Croatia (Bilandžić et al., 2017) and Egypt (Rashed & Soltan, 2004) were detected as 0.133-9.471 mg/kg, 0.026-4.23 mg/kg, 0.455-6.859 ppm, 0.06-1.96 mg/kg, 223-580 ng/g, 0.3-2.30 mg/kg, 0.2-8.3 mg/kg, 0.19-3.77 mg/kg and 0.50-1.70 mg/kg, respectively.

In the study, minimum 0.418  $\mu$ g/g and maximum 19.879  $\mu$ g/g selenium levels were obtained from Thrace honey and Muğla pine honey, respectively. Other studies in Turkey revealed levels of <1-65.9  $\mu$ g/g (54.1 mean) (Kılıç Altun et al., 2017), 54.1-67.50 ng/g

(60.7 mean) (Doker et al., 2014) and 38-113  $\mu$ g/g (Tuzen et al., 2007) for selenium content in selected honey samples. Honeys from Hungary (Czipa et al., 2015), Malaysia (Chua et al., 2012), Brazil (Liberato et al., 2013) and Croatia (Bilandžić et al., 2017) contained selenium at the levels of 2.66-36.4  $\mu$ g/kg, ND-17.202 ppm, 0.0036-0.062  $\mu$ g/kg and 0.16-36.3  $\mu$ g/kg, respectively.

In the present study, minimum and maximum levels of  $1.734 \,\mu$ g/g and  $245.205 \,\mu$ g/g for zinc were found in Muğla pine honey and Kars honey, respectively. In other studies carried out in Turkey, zinc levels in honey were indicated as <1-237 ppb (49.9 mean) (Kılıç Altun et al., 2017), 0.98-4.35 mg/kg (2.69 mean) (Turhan, 2007), 0.43-0.96 µg/g (0.67 mean) [15], 1.1-12.7 µg/g (Tuzen et al., 2007), 1.2-17.2 µg/g (Kolayli et al., 2008) and 6.76 mg/kg (Sireli et al., 2015). Zinc levels were detected to be 0.132-7.825 mg/kg, 0.185-7.20 mg/kg, 2.353-18.112 ppm, 0.07-1.85 mg/kg, 0.190-22.9 mg/kg, 565-1144 ng/g, 2.92-4.73 mg/kg, 0.5-1.5 mg/kg, 0.46-7.19 mg/kg, 1.66-5.97 µg/g and 5.0-9.3 µg/g in honey samples collected in Spain (Fernández-Torres et al., 2005), Hungary (Czipa et al., 2015), Malaysia (Chua et al., 2012), Brazil (Liberato et al., 2013), Czech (Čelechovská & Vorlová, 2001), Italy (Caroli et al., 1999), Slovenia (Golob et al., 2005), Italy (Bontempo et al., 2017), Croatia (Bilandžić et al., 2017), Poland (Formicki et al., 2013) and Egypt (Rashed & Soltan, 2004), respectively.

Minimum and maximum cadmium levels in honeys were found to be 0.000 µg/g and 0.297 µg/g, respectively in the present study. Other studies from Turkey revealed different results regarding cadmium content in honey samples such as <1 ppb (Kılıç Altun et al., 2017), below the detection limit-2.20 µg/g (<1 mean) (Doker et al., 2014), 0.9-17.9 µg/kg (Tuzen et al., 2007) and 0.343 mg/kg (Sireli et al., 2015). Levels of cadmium content in honey samples were detected as <0.003-3.31 µg/g, ND-4.502 ppm, 0.5-77.4 g/kg, <0.50-0.74 ng/g, 0.12-16.4 µg/kg, 1.0-6.5 µg/g × 10<sup>-3</sup> and 0.01-05 µg/g in Hungary (Czipa et al., 2015), Malaysia (Chua et al., 2012), Czech (Čelechovská & Vorlová, 2001), Italy (Caroli et al., 1999), Croatia (Bilandžić et al., 2017), Poland (Formicki et al., 2013) and Egypt (Rashed & Soltan, 2004), respectively.

Minimum and maximum levels of  $0.000 \ \mu g/g$  and  $3.035 \ \mu g/g$ were detected for lead concentration in honeys analyzed in the present study. According to other studies carried out in Turkey with different honey samples revealed levels of <1 ppb (Kılıç Altun et al., 2017), < below the detection limit-59.40 mg/kg (12.1 mean) (Doker et al., 2014), 8.4-105.8  $\mu g/kg$  (Tuzen et al., 2007), ND (Kolayli et al., 2008) and 1.101  $\mu g/g$  (Sireli et al., 2015) for lead. Lead content in honeys from Hungary (Czipa et al., 2015), Malaysia (Chua et al., 2012), Czech (Čelechovská & Vorlová, 2001), Italy (Caroli et al., 1999), Slovenia (Golob et al., 2005), Italy (Bontempo et al., 2017), Croatia (Bilandžić et al., 2017), Poland (Formicki et al., 2013) and Egypt (Rashed & Soltan, 2004) were detected to be 11.2-133  $\mu g/kg$ , ND-4.502 ppm, 18.4-1000.3  $\mu g/kg$ , 3.20-186 ng/g, 1.86-4.3mg/kg, 0.0-0.1 ppm, 3.28-50.0  $\mu g/kg$ , 0.06-0.13  $\mu g/g$  and 4.2-6.3  $\mu g/kg$ , respectively.

In the present study, minimum and maximum aluminium levels of  $0.775 \,\mu$ g/g and  $155.586 \,\mu$ g/g were detected in Kastamonu mad honey and Rize honey, respectively. In other studies carried out in Turkey, aluminium levels in honey samples were found to be <1-960 ppb (69.7 mean) (Kılıç Altun et al., 2017),

0.74-1.35 µg/g (1.06 mean) (Doker et al., 2014) and 83-325 µg/kg (Tuzen et al., 2007). Aluminium levels in honey samples from Hungary (Czipa et al., 2015), Malaysia (Chua et al., 2012), Italy (Bontempo et al., 2017) and Croatia (Bilandžić et al., 2017) were reported as <0.004-4.39 mg/kg, 0.708-1.872 ppm, 0.6-3.8 mg/kg and 0.46-28.8 mg/kg, respectively.

Industrial applications and agricultural chemicals including pesticides pollute the soil, water resources and plants. These chemicals may contain various elements in their compositions. Bees and bee products including honey are exposed to the contaminants via polluted pollen, water and air. Furthermore, poor beekeeping practices may be the source of elemental and heavy metal residues in honey. Honeys can be contaminated with miscellaneous elements through many factors such as containers used in beekeeping, packaging materials (paints and finish-coat materials used in the internal surfaces of the containers, uncoated tins), metal lids, caring chemicals, tranquilizer bee smoke, cleaning materials (residues on contact surfaces) and feeding syrups given to bees. The mineral composition of honey differs tremendously based on its geographic, geologic and floral origins as well as the diversity of elemental resources. Therefore, despite the concordant results recorded in some studies, different mineral compositions may be obtained even from the honey samples collected from the same region in the same study. Determination of the mineral and heavy metal contents of honeys is a useful tool for monitoring environmental pollution; therefore it is of utmost importance that the consumers are informed about the geographic origin (including the altitude and source of honey) of the products in details by proper labeling.

## **4** Conclusion

The aim of the present study was to investigate the levels of certain heavy metals and trace elements that may be found in honeys. It is considered that the findings of the study will be a contribution to assess whether or not honey varieties sold in Turkey pose a risk for public health in terms of heavy metal content and also it is assumed that demonstration of trace element composition of honeys will elucidate the nutritional value of these products. Moreover, it is anticipated that the findings will serve as a tool for estimating the permissible limit of elements in honey, which are yet neither nationally nor internationally established.

## Acknowledgements

We are grateful to Dr. Deniz Aktaran Bala for her contributions to this research and her assistance in providing the honey samples and also we would like to thank Dr. Damla Haktanir for the translation of the manuscript. This work was supported by Scientific Research Projects Coordination Unit of Istanbul University. Project number: BYP-2018-28916.

## References

Ali, M. W., Zoltai, S. C., & Radford, F. G. (1988). A comparison of dry and wet ashing methods for the elemental analysis of peat. *Canadian Journal of Soil Science*, 68(2), 43-447. http://dx.doi.org/10.4141/ cjss88-041.

- Bargańska, Ż., Ślebioda, M., & Namieśnik, J. (2016). Honey bees and their products: bioindicators of environmental contamination. Publication Cover. *Critical Reviews in Environmental Science and Technology*, 46(3), 235-248. http://dx.doi.org/10.1080/10643389.2 015.1078220.
- Batista, B. L., Silva, L. R. S., Rocha, B. A., Rodrigues, J. L., Berretta-Silva, A. A., Bonates, T. O., Gomes, V. S. D., Barbosa, R. M., & Barbosa, F. (2012). Multi-element determination in Brazilian honey samples by inductively coupled plasma mass spectrometry and estimation of geographic origin with data mining techniques. *Food Research International*, 49(1), 209-215. http://dx.doi.org/10.1016/j. foodres.2012.07.015.
- Bilandžić, N., Tlak Gajger, I., Kosanović, M., Čalopek, B., Sedak, M., Solomun Kolanović, B., Varenina, I., Luburić, Đ. B., Varga, I., & Đokić, M. (2017). Essential and toxic element concentrations in monofloral honeys from southern Croatia. *Food Chemistry*, 234, 245-253. http:// dx.doi.org/10.1016/j.foodchem.2017.04.180. PMid:28551232.
- Bontempo, L., Camin, F., Ziller, L., Perini, M., Nicolini, G., & Larcher, R. (2017). Isotopic and elemental composition of selected types of Italian honey. *Measurement*, 98, 283-289. http://dx.doi.org/10.1016/j. measurement.2015.11.022.
- Caroli, S., Forte, G., Iamiceli, A. L., & Galoppi, B. (1999). Determination of essential and potentially toxic trace elements in honey by inductively coupled plasma-based techniques. *Talanta*, 50(2), 327-336. http://dx.doi.org/10.1016/S0039-9140(99)00025-9. PMid:18967723.
- Čelechovská, O., & Vorlová, L. (2001). Groups of honey-physicochemical properties and heavy metals. *Acta Veterinaria Brno*, 70(1), 91-95. http://dx.doi.org/10.2754/avb200170010091.
- Chua, L. S., Abdul-Rahaman, N. L., Sarmidi, M. R., & Aziz, R. (2012). Multi-elemental composition and physical properties of honey samples from Malaysia. *Food Chemistry*, 135(3), 880-887. http:// dx.doi.org/10.1016/j.foodchem.2012.05.106. PMid:22953800.
- Cukur, F., Yucel, B., & Demirbas, N. (2016). Food safety applications towards beekeeping activities in the EU and Turkey: problems and suggestions. *Journal of Agricultural Economics*, 22(1), 87-95.
- Czipa, N., Andrasi, D., & Kovacs, B. (2015). Determination of essential and toxic elements in Hungarian honeys. *Food Chemistry*, 175, 536-542. http://dx.doi.org/10.1016/j.foodchem.2014.12.018. PMid:25577117.
- Doker, S., Aydemir, O., & Uslu, M. (2014). Evaluation of digestion procedures for trace element analysis of Cankiri, Turkey honey by inductively coupled plasma mass spectrometry. *Analytical Letters*, 47(12), 2080-2094. http://dx.doi.org/10.1080/00032719.2014.895908.
- Fernández-Torres, R., Pérez-Bernal, J. L., Bello-López, M. A., Callejón-Mochón, M., Jiménez-Sánchez, J. C., & Guiraúm-Pérez, A. (2005). Mineral content and botanical origin of Spanish honeys. *Talanta*, 65(3), 686-691. http://dx.doi.org/10.1016/j.talanta.2004.07.030. PMid:18969853.
- Formicki, G., Gren, A., Stawarz, R., Zysk, B., & Gal, A. (2013). Metal content in honey, propolis, wax and bee pollen and implications for metal pollution monitoring. *Polish Journal of Environmental Studies*, 22(1), 99-106.
- Golob, T., Doberšek, U., Kump, P., & Nečemer, M. (2005). Determination of trace and minor elements in Slovenian honey by total reflection X-ray fluorescence spectroscopy. *Food Chemistry*, 91(4), 593-600. http://dx.doi.org/10.1016/j.foodchem.2004.04.043.

- Jaishankar, M., Tseten, T., Anbalagan, N., Mathew, B. B., & Beeregowda, K. N. (2014). Toxicity, mechanism and health effects of some heavy metals. *Interdisciplinary Toxicology*, 7(2), 60-72. http://dx.doi. org/10.2478/intox-2014-0009. PMid:26109881.
- Kaftanoglu, O. (2017, September-October 29-04). Future of beekeeping. In *Proceedings of the 45th Apimondia International Apicultural Congress* (pp. 9-10). Turkey: Apimondia/Turkish Association of Beekeepers.
- Kılıç Altun, S., Dinç, H., Paksoy, N., Temamoğulları, F. K., & Savrunlu, M. (2017). Analyses of mineral content and heavy metal of honey sample from South and East Region of Turkey by using ICP-MS. *International Journal of Analytical Chemistry*, 2017, 6391454. http:// dx.doi.org/10.1155/2017/6391454. PMid:28584526.
- Kolayli, S., Kongur, N., Gundogdu, A., Kemer, B., Duran, C., & Aliyazicioglu, R. (2008). Mineral composition of selected honeys from Turkey. *Asian Journal of Chemistry*, 20, 2421-2425.
- Liberato, M. C. T. C., Morais, S. M., Magalhães, C. E. C., Magalhães, I. L., Cavalcanti, D. B., & Silva, M. M. O. (2013). Physicochemical properties and mineral and protein content of honey samples from Ceará state, Northeastern Brazil. *Food Science and Technology*, 33(1), 38-46. http://dx.doi.org/10.1590/S0101-20612013005000028.
- Mutlu, C., Erbas, M., & Tontul, A. S. (2017). Some characteristics of honey and other bee products and their effects on human health. *Academic Food*, 15(1), 75-83.
- Nascimento, A. S., Marchini, L. C., Carvalho, C. A. L., Araújo, D. F. D., Silveira, T. A., & Olinda, R. A. (2015). Determining the Levels of Trace Elements Cd, Cu, Pb and Zn in Honey of Stingless Bee (Hymenoptera: Apidae) Using Voltammetry. *Food and Nutrition Sciences*, 6(07), 591-596. http://dx.doi.org/10.4236/fns.2015.67062.
- Rashed, M. N., & Soltan, M. E. (2004). Major and trace elements in different types of Egyptian mono-floral and non-floral bee honeys. *Journal of Food Composition and Analysis*, 17(6), 725-773. http:// dx.doi.org/10.1016/j.jfca.2003.10.004.
- Sireli, U. T., Iplikcioglu Cil, G., Yurdakok Dikmen, B., Filazi, A., & Ulker, H. (2015). Detection of Metals in Different Honey Brands. *Kafkas Üniversitesi Veteriner Fakültesi Dergisi*, 21(6), 915-918.
- The Agency for Toxic Substances and Disease Registry ATSDR. (2015). *Priority list of hazardous substances*. Atlanta: ATSDR. Retrieved from https://www.atsdr.cdc.gov/spl/resources
- Turhan, K. (2007). Chemical contents and some trace metals of honeys produced in The Middle Anatolia Region of Turkey. *Fresenius Environmental Bulletin*, 16(5), 460-465.
- Turkish. (2017). *Turkish statistical institute*. Turkish: Tuik. Retrieved from http://www.tuik.gov.tr/PreHaberBultenleri.do?id=27704
- Tuzen, M., Silici, S., Mendil, D., & Soylak, M. (2007). Trace element levels in honeys from different regions of Turkey. *Food Chemistry*, 103(2), 325-330. http://dx.doi.org/10.1016/j.foodchem.2006.07.053.
- Vanhanen, L. P., Emmertz, A., & Savage, G. P. (2011). Mineral analysis of mono-floral New Zealand honey. *Food Chemistry*, 128(1), 236-240. http://dx.doi.org/10.1016/j.foodchem.2011.02.064. PMid:25214355.
- World Health Organization WHO. (2011). Evaluations of the joint FAO/WHO Expert Committee on Food Additives (JECFA): lead general information. Geneva. Retrieved from http://apps.who.int/foodadditives-contaminants-jecfa-database/chemical.aspx?chemID=3511
- Yılmaz Aksu, F., & Sandikci Altunatmaz, S. (2017). Risks related to heavy metal contamination in foods. J Food Hyg Technol-Special Topics, 3(3), 218-230.