Journal of Applied Pharmaceutical Science Vol. 5 (08), pp. 042-045, August, 2015
Available online at http://www.japsonline.com
DOI: 10.7324/JAPS.2015.50807
ISSN 2231-3354

Determination of Heavy Metals Contents of Some Monofloral Honey Produced in Turkey

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ARTICLE INFO

Article history:
Received on: 07/05/2015
Revised on: 24/05/2015
Accepted on: 21/06/2015
Available online: 28/08/2015

Key words:
Monofloral, Honey, Heavy metal, Turkey

ABSTRACT

In the present work, some element contents (Cd, Se, Cr, Pb, Cu, Mn, Ni, Zn, Fe) of 23 different monofloral honey samples from different regions of Turkey were assessed. Origins of monofloral honey research materials are as follows; *Pimpinella sp., Castanea sp., Astragalus sp., Onobrychis sp., Mentha sp., Coridothymus capitatus, Robinia pseudoacacia, Cedrus libani, Gossypium sp., Thymus sp., Euphorbia sp., Tilia sp., Eucalyptus sp., Ferula sp., Centaurea solstitialis, Petroselinum sp., Vitexagnus-castus, Helianthus annuus, Citrus sp., Pinus sp., Rhododendron sp., Arbutus sp., Ceratonia siliqua*. Metal contents of samples were determined by wet combustion method in ASS machine. As a result of the study, heavy metal contents of honey samples were as follows; Cd<Se<Cr<Pb<Cu<Mn<Ni<Zn<Fe. In addition, although none of the heavy metals were found in some honey samples, honey samples were found to be reliable in terms of heavy metal residues.

INTRODUCTION

Honey is used as food an important ingredient in different kinds of manufactured foods. It may be a significant source of vitamin and micro- and macro-elements essential for human health (Nanda *et al.*, 2003; Formicki *et al.*, 2013). The bee is active throughout the area surrounding the hive: for, although an opportunist in the sense that it prefers to gather pollen in the flowered fields nearby, the bee can range over long distances, even up to ten kilometers under exceptional circumstances: a hive can keep an area of seven square kilometers "under its control" (Crane, 1984; Celli and Maccagnani 2003).Bees are in close relationship with their environment, as here shown. Honey and bee products have the image of being natural, healthy and clean. However, today bee products are produced in an environment polluted by different sources of contamination (Bogdanov, 2006). Accumulation of metals may derive from external sources such as industrial pollution, incorrect treatment and agrochemicals (Bratu and Georgescu, 2005; Czipa *et al.*, 2015). Air and soil contain heavy metals, mainly from industry and traffic which can also contaminate the bee colony and its products (Bogdanov, 2006). Heavy metal present in atmosphere can deposit on the hairy bodies of bees and be brought back to the hive with pollen, or they may be absorbed together with the nectar of the flowers, or throught the water or the honeydew (Porrini *et al.*, 2003). Heavy metals are defined as metals with a density higher than 5 g cm^{-3}. 53 of the 90 naturally occurring elements are heavy metals (Weast, 1984). Human cells employ metals such as zinc, copper and iron to control significant metabolism and signaling functions making them essential for life (Sataray *et al.*, 2003; Haddad, 2012). Many heavy metals play an important biological role, the particular importance of iron and copper ions in biological processes (Halliwell and Gutteridge 1999; Haddad, 2012). On the other hand macro molecules in cell are damaged by metals induced production oxygen and nitrogen containing free radicals (oxidants) and/or metal induced depletion of the cells antioxidant defense programmed cell death or necrotic cell death are usual consequences (Pulido and Parrish, 2003).

MATERIAL AND METHODS

Total number of 23 different monofloral honey samples from different regions of Turkey were assessed in the present study and the samples were not subjected to any heating process.

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Table 1: Obtained honey samples in the study.

Honey	Honey Origin	Localization	Honey	HoneyOrigin	Localization
Eucalyptus	*Eucalyptus sp.*	Adana/Center	Chestnut	*Castanea sp.*	Ordu/Fatsa
Sunflower	*H. annuus*	Adana/Ceyhan	Acacia	*R. pseudoacacia*	Ordu/Fatsa
Citrus	*Citrus sp.*	Adana/Yuregir	Rhododendron	*Rhododendron sp.*	Giresun/Bulancak
Strawberry tree	*Arbutus sp.*	Mersin/Silifke	Linden	*Tilia sp.*	Kastamonu/Azdavay
Carob bean	*C. siliqua*	Mersin/Silifke	Wild Mint	*Mentha sp.*	İstanbul/Catalca
Ferula	*Ferula sp.*	Mersin/Silifke	Yellowstar-thistle	*C. solstitialis*	Diyarbakır/Lice
Anise	*Pimpinella sp.*	Antalya/Korkuteli	Cotton	*Gossypium sp.*	Diyarbakır/Devegecidi
Cedrus	C. *libani*	Antalya/Elmalı	Sainfoin	*Onobrychis sp.*	Van/Baskale
Cesme thyme	*C. capitatus*	İzmir/Çesme	Thyme	*Thymus sp.*	Batman/Gercus
Chasteberry	*V. agnus-castus*	İzmir/Torbalı	Euphorbia	*Euphorbia sp.*	Mardin/Midyat
Parsley	*Petroselinum sp.*	Hatay/İskenderun	Pine	*Pinus sp.*	Mugla/Marmaris
Astragalus	*Astragalus sp.*	Konya/Hadim			

The honey samples were obtained from the manufacturers in 2012. The location of obtained honey samples were presented in Table 1. Honey samples were preserved in glass jars at room temperature. All samples were studied in triplicate.

Methods

Heavy metal analysis of monofloral honey samples

Heavy metal analysis of monofloral honey samples were evaluated according to wet combustion method of Dean *et al*. (2003). In the analysis of samples, lead (Pb), cadmium (Cd), zinc (Zn), iron (Fe), chromium (Cr), copper (Cu), manganese (Mn), nickel (Ni), selenium (Se) heavy metals were determined by Atomic Absorption Spectrophotometer (AAS) instrument. Results of the samples were determined by comparing the results with measurements of heavy metal standards.5 g of honey sample was mixed with 10 ml nitric oxide and heated at 95 ^{o}C for a period of 1 hour until the solution gets yellow color. After cooling the mixture, 5 ml concentrated nitric acid (HNO_3) was added and heated to 180^{o}C.1 mL of HNO3 were added if necessary until clear color or pale straw color. After cooling the sample, 1 ml of H_2O_2 (500 g/L) was added and heated to 200 ^{o}C. This step was repeated until malodorous brown fumes over. The samples cooled again and mixed with 10 ml distilled water and 0.5 ml concentrated HNO_3 and slowly heated to 200 ^{o}C until the white malodorous gases obtained. After that, the samples cooled again and mixed with 10 ml distilled water and 1 ml H_2O_2 (500 g/L) slowly heated to 240^{o}Cuntil white malodorous gases obtained. At the end, samples were cooled and transferred to 25 mL volumetric flask, and analyzed in AAS instrument (Dean, 2003). Heavy metal contents of samples were compared to standards and results were determined in ppm (mg/kg).

Analysis of sediment for the identification of honey samples

The monofloral honey samples were classified according to their botanical origin using the method of Maurizio, (1951). Pollen grains were microscopically observed and compared with there ferenceslides for identification. Botanical classification was achieved when the pollen spectrum contained >45 % of the corresponding dominant pollen. Floral origins of honey samples are; *Pimpinella sp., Castanea sp., Astragalus sp., Onobrychis sp., Mentha sp., Coridothymus capitatus*, *Robinia pseudoacacia*, *Cedrus libani*, *Gossypium* sp., *Thymus sp.*, *Euphorbia* sp., *Tilia* sp., *Eucalyptus* sp., *Ferula* sp., *Centaurea solstitialis*, *Petroselinum* sp., *Vitexagnus-castus*, *Helianthus annuus*, *Citrus* sp., *Pinus* sp., *Rhododendron* sp., *Arbutus* sp., *Ceratonia silique* (Table 2).

Table 2: Botanical origin of honey samples.

Monofloral honey	Pollen frequency (%)	Monofloral honey	Pollen frequency (%)
Pimpinella sp.	0.45	*Tilia sp.*	0.66
Castanea sp.	0.75	*Eucalyptus sp.*	0.73
Astragalus sp.	0.52	*Ferula sp.*	0.47
Onobrychis sp.	0.47	*Centaurea solstitialis*	0.46
Mentha sp.	0.51	*Petroselinum sp.*	0.79
Coridothymus capitatus	0.63	*Vitexagnus-castus*	0.86
Robinia pseudoacacia	0.54	*Helianthus annuus*	0.86
Cedrus libani	0.58	*Citrus sp.*	0.57
Gossypium sp.	0.53	*Rhododendron sp.*	0.56
Thymus sp.	0.48	*Arbutus sp.*	0.61
Euphorbia sp.	0.60	*Ceratonia siliqua*	0.46

RESULTS AND DISCUSSION

As a result of the study obtained results of the heavy metal contents of samples were presented in Table 3. İn this study, the average cadmium (Cd) contents of honey samples were determined to be 0.0002±0.0 mg/kg (ppm). Generally, Cd is toxic heavy metal and should not be present in food samples. World-wide there are no specific MRL (Maximum Residue Limit) levels for these heavy metals in honey. The average recommended daily intake in foods is reported to be 60 mg/day for cadmium (Silici et al. 2008). Average lead (Pb) contents of samples were determined as 0.04±0.09mg/kg. The highest lead content in honey samples were determined in Cesme thyme and Wild mint honey samples which are Wild mint obtained from Istanbul and Cesme thyme obtained from İzmir, respectively. TheMRL proposal for EU is at 1 mg/kg for lead (Byrne, 2000, Bogdanov, 2006). Lead contents of all honey samples are appropriate with the standards of EU. On average, the Pb concentration was low and unproblematic in honey. Bee products are less suitable to serve as indicators for the measurement of Pb and Cd pollution due to considerable natural variation (Porrini *et al.*, 2002; Bogdanov 2006). Average iron (Fe), copper (Cu) and zinc (Zn) contents of samples in our study was 2.85±0.5, 0.15±0.1 and0.48±0.3 mg/kg. Cu contents of honey samples in our study was determined to below the limit standards. The average recommended daily intake in foods is reported to be 30 mg/day for copper and 12–15 mg/day for zinc (Silici et al. 2008).

On the other hand, The Food and Agriculture Organization (FAO) and the World Health Organization (WHO) have set a limit for heavy metal intake based on body weight (b.w.). For an average adult (60 kg b.w.), the provisional tolerable daily intake for Pb, Fe, Cu, and Znare 214 mg, 48 mg, 3 mg, and 60 mg, respectively (Joint FAO/WHO Expert Committee on Food Additives 1999) (Silici et al., 2014). Additionally, average contents of other heavy metals in honey samples were as follows; chromium (Cr) 0.0086 ± 0.01 ppm, manganese (Mn) 0.187 ± 0.18 ppm,nickel (Ni) 0.26 ± 0.23 ppm and selenium 0.0003 ± 0.0. Type and quality of equipment used to store honey after harvesting, processing and/ or preparation of honey can generate high Cr content. Likewise, storing honey in galvanized containers can be source of Zn contamination (Silici *et al.*, 2008). The MRL levels in Switzerland for Nivaries is between 0.1 mg/kg (beer) to 0.2 mg/kg (edible fat). Following levels have been found in honey: Ni: 0.004–3.23 mg/kg (Porrini *et al.*, 2002; Bogdanov, 2006). As a result of the heavy metal analysis, although heavy metal contents of wild mint honey which is obtained from Istanbul and Cesmethyme honey which is obtained Izmir is appropriate according to TGK and Codex standards, both have the highest heavy metal content among other honey samples in the study. Because of their production in close proximity to industrial and residential areas, this honey samples are thought to be exposed to heavy metal contamination.

Some studies related to heavy metal content in honey samples were presented in Table 4. As indicated in the table some of the values are in parallel with our findings. Generally, findings of the present study suggest that heavy metal contents of the honey samples assessed in this study are not high compared with the literature (Table 4). Although honey is and important food and consumed for the health, heavy metal contamination in honey and toxic levels of these heavy metals in honey samples can lead and to serious health problems.

Table 3: Heavy metal contents of honey samples (mg/kg)

HONEY	**Pb**	**Zn**	**Fe**	**Cd**	**Cu**	**Cr**	**Mn**	**Ni**	**Se**
Pimpinella sp.	0.08	0.84	2.81	0.0001	0.29	0.024	0.35	0.53	0.0001
Castanea sp.	<0.001	0.34	3.28	<0.0001	0.16	0.01	0.18	0.06	<0.0001
Astragalus sp.	<0.001	0.75	3.12	<0.0001	0.36	0.003	0.32	0.41	0.0001
Onobrychis sp.	<0.001	0.57	2.98	<0.0001	0.28	0.005	0.17	0.36	0.0001
*Mentha*sp.	0.33	1.16	3.61	0.0023	0.32	0.036	0.63	0.87	0.0028
C. capitatus	0.25	0.98	3.54	0.0015	0.35	0.026	0.67	0.85	0.0032
R. pseudoacacia	<0.001	0.61	3.18	<0.0001	0.18	0.001	0.34	0.23	<0.0001
C. *libani*	<0.001	0.097	1.94	<0.0001	0.04	0.003	0.01	0.08	<0.0001
Gossypium sp.	0.02	0.53	3.17	0.0012	0.03	0.004	0.07	0.16	<0.0001
*Thymus*sp.	<0.001	0.56	2.87	<0.0001	0.05	0.003	0.04	0.12	<0.0001
Euphorbia sp.	<0.001	0.5	2.75	<0.0001	0.03	0.007	0.07	0.15	<0.0001
*Tilia*sp.	0.12	0.48	2.97	<0.0001	0.14	0.004	0.14	0.22	<0.0001
*Eucalyptus*sp.	0.09	0.085	2.81	<0.0001	0.03	0.014	0.12	0.13	<0.0001
*Ferula*sp.	<0.001	0.62	3.12	<0.0001	0.17	0.004	0.21	0.25	0.0001
C. solstitialis	<0.001	0.81	3.25	0.0001	0.32	0.003	0.36	0.47	0.0002
Petroselinum sp.	<0.001	0.08	2.2	<0.0001	0.02	0.002	0.08	0.09	<0.0001
V. agnus-castus	<0.001	0.05	1.74	<0.0001	0.01	0.001	0.01	0.02	<0.0001
H. annuus	<0.001	0.39	1.71	<0.0001	0.02	0.01	0.02	0.04	<0.0001
*Citrus*sp.	<0.001	0.32	2.1	<0.0001	0.03	0.02	0.05	0.17	<0.0001
Rhododendron sp.	<0.001	0.1	3.27	<0.0001	0.08	0.006	0.11	0.18	<0.0001
Arbutus sp.	<0.001	0.24	2.77	<0.0001	0.05	0.004	0.06	0.12	<0.0001
C. siliqua	<0.001	0.31	2.95	<0.0001	0.11	0.003	0.13	0.21	0.0001
Pinus sp.	<0.001	0.52	3.46	<0.0001	0.18	0.005	0.16	0.25	0.0001
Min	<0.001	0.05	1.71	<0.0001	0.01	0.001	0.01	0.02	<0.0001
Max	0.33	1.16	3.61	0.00023	0.36	0.036	0.67	0.87	0.0032
Mean	0.04±0.09	0.48±0.3	2.85±0.5	0.0002±0	0.15±0.1	0.0086±0.01	0.187±0.18	0.26±0.2	0.0003±0

Table 4: Some studies related to heavy metal content in honey samples (ppm).

HEAVY METALS	**This Study**	**Bilandžićet al. (2014)**	**Ru et al. (2013)**	**Chudzinska and Baralkiewicz (2011)**	**Gül (2008)**	**Silici et al. (2008)**	**Pisani et al. (2008)**	**Terrab et al. (2005)**	**Tüzen (2002)**
Pb	0.04	0.81	0.03*	0.59	0.29	0.00151-0.0553*	0.08	0.08	0.0303-0.0586
Cu	0.15	20.6	0.05	1.31	0.24*	0.009-0.035*	0.91	4.18	0.25-1.30
Cr	0.0086	-	-	0.036	0.03	*0.00124-0.013	-	0.1	-
Zn	0.48	6.78	0.01	3.27	1.53	*0.47-6.57	1.82	5.65	1.15-4.95
Fe	2.85	0.003	-	-	3*	*1.21-12.9	3.07*	9.19	3.45-8.94
Mn	0.187	-	-	6.5	1.27	1.11-61	1.54	1.14	0.32-0.70
Ni	0.26	-	-	0.84	0.32*	0.00121-0.131	0.31*	0.33*	-
Cd	0.0002	0.002	0.001	0.02	0.00	*0.00028-0.0023	0.003	0.04	0.00523-0.00982
Se	0.0003	-	-	-	-	0.014-0.323	-	0.05	-

CONCLUSION

In conclusion, heavy metal contents of honey samples were0.0002, 0.04, 0.48, 2.85, 1.15, 0.187, 0.059, 0.26, 0.0086, and 0.0003 ppm for cadmium (Cd), lead (Pb), zinc (Zn), iron (Fe), copper (Cu), manganese (Mn), nickel (Ni), chromium (Cr), selenium (Se). Although heavy metals were determined in some honey samples, honey samples in this study was found to be reliable in terms of heavy metal residues. Additionally, consumption of the food samples with high heavy metal residues can lead to serious health manifestations.

Acknowledgement

Pollen analyses in the this study was supported by the Comission of Scientific Researches Projects (Mustafa Kemal University), Proje number: BAP-409.

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How to cite this article:

Tuba Pehlivan and Aziz Gül. Determination of Heavy Metals Contents of Some Monofloral Honey Produced in Turkey. J App Pharm Sci, 2015; 5 (08): 042-045.