

ORIGINAL ARTICLE

Analysis of Heavy Metals in Honey from North-Western Regions of Iran

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KEYWORDS

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ABSTRACT: In the present study, the concentrations of three heavy metals including lead (Pb), zinc (Zn) and arsenic (As) in 72 different honey samples collected from North-Western regions of Iran were investigated using atomic absorption spectroscopy method. All metals were detected in 88.88% of the honey samples. The range of metals contamination in honey samples was 0.01 to 11.09 ppm. Zn was the most abundant metal, with an average concentration of 4.41 ± 2.4 ppm. The mean levels of Pb (0.12 ppm) and Zn (6.84 ppm) in honey samples of East Azerbaijan Province was significantly higher than average level of these metals in honeys from other provinces. Based on the results of this study and compared with recommended daily intakes, the heavy metal content of honey samples was not harm and is unlikely to cause any intoxication following honey consumption.

INTRODUCTION

Honey is one of the oldest natural food known to humans used as a whole food in all parts of the world, treatment of various ailments and diseases, and also as a biological monitor for the determination of heavy metals, radioactivity in the region and to determine environmental quality in polluted environments [1, 2]. Nutritional adverse effects of heavy metals, as well as some of their beneficial role in human life have been reported [3]. The major contaminants of food supply was

cadmium, lead and mercury, also these metals may be considered as the most important environmental contaminants, some metal such as iron, zinc and copper are need for human body metabolism [4]. The accumulation most of heavy metal in the different body organs cause the side effects because they are not biodegradable and long biological half-lives [3, 5]. There is a high correlation between nutrient and mineral

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composition of plant and animal tissues, as well as ultimately accumulate in human different tissues.

The quality of the soil and the plant's ability to absorb elements affect the essential elements content in plant tissue [3, 6]. Beekeeping as one of the most important branches of agriculture in the world was known. Nowadays it is estimated that there are 56 million bees worldwide, and an average of 1.2 million tons of honey is produced annually worldwide in one year [1]. Iran is one of the major countries in the world in production honey production. According to official statistics in 2007, 2% (36000 tons) of world's total honey production belonged to Iran. Approximately 10000 tons of honey was produced in northwest regions of Iran specially Urmia City yearly [1, 7].

Food safety is of great importance in human health. The food with high quality and safety will protect of public health. Many reports indicated that honey samples collected industrial regions had the higher heavy metal (Cd, Pb, Hg, Zn, Cu, Ni and Cr) than those from natural regions. The accumulation of the toxic metal in human body causing the side effects, so honey quality specific elemental content was the important factor for human nutrition and safety [8].

The present study aimed at evaluating the quality of Iranian honey (from northwest regions including West Azerbaijan, East Azerbaijan and Adabil provinces) in terms of some heavy metals (Pb, Zn and As) contamination.

MATERIALS AND METHODS

Sampling

All Seventy-two honey samples were prepared from Northwest regions of Iran in 2013-2014 (including: East Azerbaijan 19 samples, West Azerbaijan 31 samples and 22 samples from Ardabil). The honey samples were stored in plastic molds and kept at room temperature (25 °C) until they were analyzed.

Heavy metal analysis of honey samples

The analysis of heavy metal in honey samples was done using atomic absorption spectroscopy (AAS) methods according standard method [8]. Briefly, one gram of each honey sample from each region was mixed with magnesium acetate (1 mg/mL). The mixture was placed in a porcelain crucible. After drying at 100 °C for 2 h, the samples were ashed at 600 °C. Care was taken during heating so that no excess foaming took place. The ash was extracted with nitric acid (HNO₃) 2N and was diluted to 30 ml. The contents of lead (Pb), zinc (Zn) and arsenic (As) were determined directly in the ash solution using atomic absorption spectroscopy (GF 3000 model AAS, Graphite Furnace GF 3000, Auto Sampler GBC PAL 3000, GBC Scientific Equipment Pty Ltd, Australia). The results were read three times and the mean values and the relative standard deviations were computed. Limits of detection and quantification were 0.012 and 0.03 ppm for Pb, 2.16 and 0.023ppm for AS and 0.007 and 0.018 ppm for Zn.

STATISTICAL ANALYSIS

All experiments were conducted in triplicate, and statistical analysis was performed using SPSS 17.0 (Chicago, IL, USA). All results were computed as mean standard deviation and were subjected to one-way analysis of variance to establish whether the differences in experimental results were significant or not. The Statistical significance was determined at $P < 0.05$.

RESULTS

Heavy metal content in all honey samples (from tree province of northwest region of Iran) is shown in Table 1 as mean±SD. All of the metals were detected in 88.88 % of the honey samples (Figure 1).

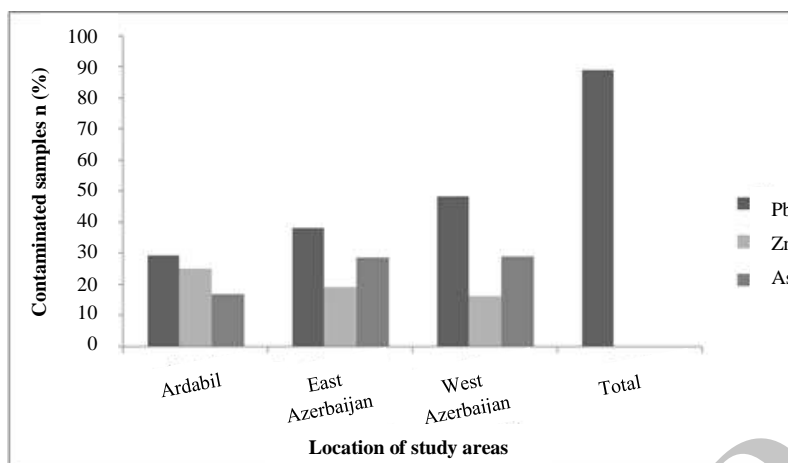


Figure 1. Comparison the percentage metal contamination of honey samples from different areas

The range of variation of each element was very wide in the samples from the same region, and between different provinces as well. Zn was the most abundant metal, with an average concentration of 4.41 ± 2.4 ppm. The highest concentration of metal detected in honey samples was the Zn (11.09 ppm), followed by As (0.22 ppm). The lowest level was found for Pb, at a concentration of 0.01

ppm. Metal levels were significantly differed in honey samples belonged to the different regions. The mean level of Pb (0.12 ppm) and Zn (6.84 ppm) in honey samples of East Azerbaijan Province were higher than concentrations of these metals in the honey samples from other ($P < 0.05$). Based on our results the lowest mean values for all metals belonged to honey samples from Ardabil.

Table 1. Metal contamination of 72 honey samples collected from North-Western regions of Iran

Regions	Number of Samples	Mean \pm SD (ppm)		
		Pb	Zn	As
Ardabil	24	0.06 ± 0.01^a	2.03 ± 0.92^a	0.08 ± 0.02^a
East Azerbaijan	21	0.12 ± 0.06^b	6.84 ± 1.74^b	0.11 ± 0.04^a
West Azerbaijan	31	0.08 ± 0.04^a	4.37 ± 1.01^c	0.16 ± 0.13^b
Total	72	0.08 ± 0.04	4.4 ± 3.40	0.11 ± 0.04

Means \pm SD in the same column with different letters are significantly different ($P < 0.05$).

DISCUSSION

The term heavy metal refers to high-density metallic chemical element, toxic or poisonous at low concentrations and cannot be degraded or destroyed. Heavy metal poisoning could result from drinking water contamination, breathing air with high concentrations of metals and contaminated food [1]. Studies on honey have been carried out with emphasis on organic, but

little has been done on inorganic aspect. The amount of minerals in honey is not only effective on honey quality, but is also useful indicators of environmental pollution [9].

Honey acidic nature can lead to enter heavy metal from containers and processing equipment [10]. There are various reviews about heavy metal contamination of honey samples from different parts of the world, some mentioned in Table 2.

Table 2. Heavy metal contamination of honey samples from different parts of the world

No	Region	Heavy metal	Level contamination	References No
1	Kahramanmaras, City, Turkey	Cd, Mn, Fe and Mg	0.32, 0.03, 0.36 and 10.45 (ppm)	[2]
2	Middle Anatolia, Turkey	Mn, Pb and Cd	0.32-4.56 ppm, 8.4-105 ppb and 0.9-17.9 ppb	[8]
3	southeastern Anatolia, Turkey	Mn	1.0 ppm	[11]
4	different regions in Turkey	Mn	0.49 ppm	[12]
5	different regions in Turkey	Pb, Cd and Mn	55.2 ppb, 4.53 ppb and 0.31 ppm	[13]
6	Kayseri	cadmium, copper, zinc, nickel, lead	0.11-0.18 ppm, for 0.15-0.66 ppm, 2.2-11 ppm, 0.2-0.8 ppm and 0.1-0.85 ppm	[14]
7	-	Pb, Cd, Cu, Fe and Mn	0.02±0.03, 0.01±0.01, 0.91±0.66, 3.13±1.44, 1.26±1.29 ppm,	[15]
8	Argentina	Cd and Pb	0.2 to 1.37 ng/g and from 4.6 to 30.5 ng/g ⁻¹	[16]
9	Italia	Pb, Cd, Cu, Fe and Zn	0.10005, 0.25005, 5.00005 and 3.105 ppm	[17]
10	France	Pb, Cd, Cu, Fe, Mn and Zn	were 0.793 ppm, 0.152 ppm, 0.305 ppm, 11.03 ppm, 3.685 ppm and 1.343 ppm	[18]
11	Nigerian	Cu, Fe, Mn and Zn	21 ppm, 220,6 ppm, 3 ppm and 63.4 ppm	[19]

In Iran, Saghaei et al. investigated the level of some heavy metals in different Urmai honey samples. Accordingly, the mean contaminations of Pb, Cr, Zn, as and Ni were 0.04±0.1 ppm, 7.09±9.4 ppm, 9.99±26.5 ppm, 0.0008±0.0011 ppm, 0.003±0.005 ppm, respectively. Based on their results, the Pb level was lower than the maximum residue limits (EU ML). Other metal levels were within the acceptable levels [1].

Akbaria et al. assessment of metal levels of 10 different honey brands and results showed that the average amounts of metals (including: Se, Cu, Cd, Pb, As and Mn) were measured less than 0.5 mg/kg, lead content had the lowest level concentration (0.11 mg/kg) [20].

In the current study, Pb and Zn levels were 0.08±0.04 and 4.41±3.40 ppm respectively (Table 1). Determined

levels of Pb and Zn in the present study were lower than the levels of these metals in honey samples collected from different regions of the world specially Turkey and Iran.

Among the examined elements, Pb residues in honey are of great concern. According to WHO reports, the average recommended daily intake of Pb is 210 µg/d for a 60 kg adult person. In a theoretical food basket, an ordinary person should take 20 gram honey daily [21]. Based on this study results the mean Pb content of honey samples was 0.08 ppm. Therefore, consuming 20 g per day of honey provides 1.6 µg Pb in a day. The results showed that the Iranian people with an average weight of 60 kg for lead intake levels lower than is recommended limit.

The average recommended daily intake of As with FAO/WHO is 130 µg/day for a 60-kg adult person [1], and although the average level of As in our honey samples (0.11 ppm) was lower than that in other reports and it was still less than the recommended dose [21]. The amount of Zn residues in honey samples examined in the present study was higher than that of was reported by other researchers [14, 22, 18, 19].

The mean value of the Zn intake from honey consumption was 4.41 ppm, which is less than the amount recommended by PMTDI as 60 mg/day [1]. Contamination of plant tissues with heavy metal is hazardous for the food chain and these accumulations causing dangerous effects on humans or animals health [22]. The main sources of pollution of heavy metals in plants and lands are including atmospheric precipitation, contaminated water and the use of insecticides and pesticides and fertilizers [23]. Presence of lead and arsenic in food is strongly banned because was the toxic heavy metals, although other elements may be found in human diet (like Cu, Fe, Zn, Al, Mg and Cr) [24].

Estimated amounts of metals in honey samples produced from the northwest of Iran was lower than permitted limit. The low levels of toxic metals in honey might be an indication of a clean environment as some studies suggested that honey could be used as an indicator of metal environmental metal contamination.

CONCLUSIONS

Based on the results of this study and compared with recommended daily intakes, the heavy metal content of honey samples was not harm and is unlikely to cause any intoxication following honey consumption. However, it is recommended to beekeeping activities to be done away from industrial areas with high pollution of heavy metals.

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REFERENCES

1. Saghaei S., Ekici H., Demirbas M., Yarsan E., Tumer I., 2012. Determination of the Metal Contents of Honey Samples from Orumieh in Iran. *Kafkas Univ Vet Fak Derg.* 18(2), 281-284.
2. Erbilir F., Erdogru O., 2005. Determination of heavy metals in honey in Kahramanmaraş, City, Turkey. *Environ Monitor Ass.* 109, 181-187.
3. Sobukola O.P., Adeniran O.M., Odedairo A.A.; Kajihausa O.E., 2010. Heavy metal levels of some fruits and leafy vegetables from selected markets in Lagos, Nigeria. *Afr J Food Sci.* 4(2), 389 – 393.
4. Zaidi M.I., Asrar A., Mansoor A., Farooqui M.A., 2005. The heavy metal concentrations along roadsides trees of Quetta and its effects on public health. *J Appl Sci.* 5(4), 708-711.
5. Sathawara N.G., Parikish D.J., Agrwal Y.K., 2004. Essentials heavy metals in environmental samples from western Indian. *Bull Environ Cont Toxicol.* 73, 756-761.
6. Divrikli U., Saracoglu S., Soylak M., Elci L., 2003. Determination of trace heavy metal contents of green vegetables samples from Kayseri- Turkey by flame atomic absorption spectrometry. *Fresenius Environ Bull.* 12, 1123-1125.
7. Anonymous: <http://www.farmna.ir/Pages/News-7496.html>. Accessed: 24 May 2011.
8. Tuzen M., Soylak M., Trace heavy metal levels in microwave digested honey samples from Middle Anatolia, Turkey. *J Food Drug Anal.* 13(4), 343-347(2005).
9. Alissandrakis E., Tarantilis P.A., Harizanis P.C., Polissiou M., 2007. Aroma investigation of unifloral Greek citrus honey using solid-phase microextraction

coupled to gas chromatographic- mass spectrometric analysis. Food Chem. 100, 396–404.

10. Pisani A., Protano G., Riccobono F., 2008. Minor and trace elements in different honey types produced in Siena County (Italy). Food Chem. 107, 1553–1560.

11. Yilmaz H., Yavuz O., 1999. Content of some trace metals in honey from south-eastern Anatolia. Food Chem. 65, 475-476.

12. Yarsan E., Karacal F., Ibrahim I.G., Dikmen B., Koksak A., Das Y.K., 2007. Contents of some metals in honeys from different regions in Turkey. Bull Environ Contam Toxicol. 79, 255-258.

13. Uren A., Serifoglu A., Sariyaha Y., 1998. Distribution of elements in honeys and effects of a thermoelectric power plant on the element contents. Food Chem. 61, 185-190.

14. Demirezen D., Aksoy A., 2005. Determination of heavy metals in bee honey using by inductively coupled plasma optical emission spectrometry (ICP-OES). GU J Sci. 18(4), 569-575.

15. Fredes C., Montenegro G., 2006. Heavy metals and other trace elements contents in Chilean honey. Cien Inv Agr. 33 (1), 50-58.

16. Frazzoli C., D'Ilio S., Bocca B., 2007. Determination of Cd and Pb in honey by SF-ICP-MS: Validation figures and uncertainty of results. Analytical Letters. 40, 1992-2004.

17. Antonescu C., Mateescu C., 2001. Environmental pollution and its effects on honey quality. Roum Biotechnol Lett. 6 (5), 371-379.

18. Devillers J., Dore, J.C., Marengo M., Poirier-Duchene F., Galand N., Viel C., 2002. Chemometrical analysis of 18 metallic and nonmetallic elements found in honeys sold in France. J Agric Food Chem. 50, 5998-6007.

19. Adebisi F.M., Akpan I., Obiajunwa E.I., Olaniyi H.B., 2004. Chemical/physical characterization of Nigerian honey. Pak J Nutr. 3(5), 278-281.

20. Akbaria B., Gharanfobid F., Hassanzadeh Khayyat M., Khashyarmansheh Z., Rezaee R., Karimi G., 2012. Determination of heavy metals in different honey brands from Iranian markets. Food Additives Contam. Part B, 1–7.

21. World Health Organization. 1993. Evaluation of certain food additives and contaminants. WHO Technical Report Series, Number 837. Geneva: World Health Organization.

22. Mantovi P., Bonazzi G., Maestri E., Marmiroli N., 2003. Accumulation of copper and zinc from liquid manure in agricultural soils and crop plants. Plant Soil. 250(2), 249–257.

23. Marcovecchio J.E., Botte S.E., Freije R.H., 2007. Heavy metals, major metals, trace elements. In: Nollet LML, editor. Handbook of water analysis. 2nd ed. Boca Raton: CRC Press. Pp. 275–311.

24. Ahmed F.E., 1999. Trace metal contaminants in food. In: Moffat CF, Whittle KJ, editors. Environmental contaminants in food. Sheffield: CRC Press. Pp.146–214.