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DETERMINATION OF HEAVY METALS CONTENT IN COTTON HONEY IN KAFR EL-SHIEKH PROVINCE, EGYPT

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ABSTRACT

Honey samples were collected from nine regions represented for Kafr El-Sheikh Province, Egypt during cotton season in 2011, to estimate certain heavy metals in cotton honey. The obtained results showed that, all honey samples contained four heavy metals (Fe, Mn, Zn and Pb) and their levels were found below acceptable limits The elements of Fe, Mn, Zn and Pb in cotton honey ranged between 20.00-44.50, 0.42-1.40, 1.20-3.28 and 0.50-2.37 mg/kg, respectively. The risk elements (Cu, Cd, Ni and Cr) were not detected in all samples. Significant ($P < 0.01$) positive correlations between Fe and Mn as well as between Zn and Pb were observed. It can be concluded that the heavy metals content of cotton honey in Kafr El-Sheikh Governorate significantly depended on the area from where it was harvested.

Keywords: Honey bee, honey, cotton, heavy metals, risk elements

INTRODUCTION

Honey and other bee products of honey bee have the image of being natural, healthy and clean. However, nowadays bee products are produced in an environment not free of contamination sources (Bogdanov, 2006). Any heavy metals present in honey above the permitted levels are threats to human body through the possible negative effect of the contaminants. Zugravu et al. (2009) reported that, heavy metals such as lead and cadmium can be harmful to the environment (pollution of soil and water, accumulation in plants) and can be serious health hazards for human (kidney diseases, provoking lung damage, nervous system failures, etc.).

Estimates showed that, most of tested honey samples were not free of heavy metals, but their levels were found well below the permitted limits (Nour,199 8;FakhimzadehandLodenius,1999;Sanna et al., 2000;ČelechovskáandVořlová, 2001; Devillers et al., 2002 a,b; Bogdanov et al., 2003; Taddia et al., 2004; Erbilir and Erdoqrul, 2005; Rateb, 2005; Bogdanov, 2006; Sheref, 2007; Zugravu et al., 2008; Zugravu et al., 2009; Taha et al., 2010).In Finland, the concentrations of heavy metals in honey were less than 10% of the acceptable level. Only in a few industrial sites, the concentrations of heavy metals slightly exceed the acceptable level (Fakhimzadeh and Lodenius, 1999).

Since honey is a nutritional resource that depends on biotic and abiotic factors around the beehives, the presence of heavy metals could be related to its botanical origin (Fredes and Montenegro, 2006). The heavy metals in honey may be variable according to botanical origin. Honey samples were collected from the same areas but for different types of honey: poly-flora and acacia were differed in heavy metals content (Bratu and Georgescu, 2005). In addition, the environmental pollution in the area where the apiaries and/or bee feed sources plays great role in contamination of bee products with certain risk elements.

Several authors have indicated that, bees and their products may be used as biological indicators of the environmental pollution present in the area where they fly (Sanna et al., 2000; Conti and Botre, 2001; Porrini et al., 2002; Celli and Maccagnani, 2003; Taddia et al., 2004; Herrera et al., 2005; Jin et al., 2006).

Environmental heavy metals pollution is a reality in our country and can lead to chemical contamination of products within the human food chain. The chosen research honey source (cotton) is already known for the beekeepers by frequent application of pesticides on cotton fields. Also, the cotton honey consists of flower nectar and nectar from extra-floral nectaries existed on leaves, which are more susceptible to atmospheric pollutant. There are three main purposes for monitoring bee products: consumer health protection, international commercial competition, and better product quality (Yakobson, 1996).

The present study aimed to investigate the relation between content of heavy metals in honey and geographical origin, and to verify honey use as an indicator of environmental pollution.

Materials and Methods

Honey samples

By the end of cotton season in 2011, 270 (5 samples × 6 apiaries × 9 regions) cotton honey samples were collected. The details of sampling as follows: nine regions (Mutubas, Fowah, Disuq, Qallin, Sidisalem, Kafrelsheikh, Riyadh, Al-Hamool and Beila) represented for Kafr El-Seikh Province, Egypt. Thirty honey samples were collected from 6 divergent apiaries (5 samples from each apiary) from each region. Each sample consists of 1.00 kg of capped honey collected from 10 colonies. All samples were taken directly from combs aged two years. The honey samples were packaged in small plastic jars, and clarified to determine the content of certain heavy metals at the Central Laboratory of Kafr El-Sheikh University, Egypt.

Sample preparation

Wet digestion with nitric acid following the method of AOAC (2000) was used to prepare honey samples for determination of heavy metals content. One gram of each honey sample was digested in a Kjeldahl flask with 10 ml of 75% HNO_3 for oxidation of carbonaceous matter. The contents of the flask were heated 100-120°C, to evaporate the acid. Drops of perchloric acid were added until all the organic matter was oxidized. This point was reached when no further darkening of the solution occurred on continuous heating and a clear solution was obtained. It was cooled and gauged to 50 ml with distilled water. A blank experiment was carried out by adding same amount of nitric acid to 1 ml distilled water.

Atomic absorption spectroscopy

An Atomic Absorption GBC AVANTA Σ GF 3000 was used for detection of iron (Fe), zinc (Zn), copper (Cu), manganese (Mn), lead (Pb), cadmium (Cd), nickel (Ni) and chromium (Cr) concentrations. The limit of determination of the analysed elements is 0.01 mg/kg. The optimum working range for aforementioned elements is 2.00-2700.00, 1.00-27.00, 0.40-12000.00, 2.50-460.00, 1.00-730.00, 0.20-800.00, 0.18-80.00 and 0.20-80.00 mg/kg, respectively. The instrument was calibrated by using standard solutions of various metal salts. All determinations were made in triplicates.

Statistical analysis

Data were statistically analyzed by the analysis of variance (ANOVA).Significant differences among the means were detected using Duncan's Multiple Range Test (Duncan 1955). A Pearson's correlation was calculated to test for an association between the selected metals by using SAS Institute (2003).

RESULTS

Data summarized in (Table 1) revealed that the content of heavy metals in cotton honey was significantly affected by geographical origin. All honey samples collected from aforementioned regions of Kafr El-Sheikh Province, Egypt, were not free of heavy metals, but fortunately, their levels were found below the acceptable limits. Four heavy metals [iron (Fe), manganese (Mn), zinc (Zn) and lead (Pb)] were detected in all examined honey samples. The highest levels of Fe (38.82 mg/kg), Mn (1.17 mg/kg) were found in Disuq and Fowah regions, respectively, while highest levels of Zn (2.66 mg/kg) and Pb (2.11 mg/kg) were obtained from Riyadh region. Meanwhile, the lowest averages of Fe (21.50 mg/kg), Mn (0.46 mg/kg), Zn (1.63 mg/kg) and Pb (0.86 mg/kg) were found in honey samples from Beila, Riyadh, Mutubas and Kafrelsheikh regions, respectively. The risk elements [copper (Cu), cadmium (Cd), nickel (Ni) and chromium (Cr)] were not detected in all cotton honey samples collected from Kafr El-Sheikh Province.

Significant ($P < 0.01$) positive correlations between Fe and Mn as well as between Zn and Pb were observed, meanwhile, significant ($P < 0.05$) negative correlation between Mn and Zn was detected as shown in (Table 2).

DISCUSSION

The levels of found elements were varied and significantly ($P < 0.05$) affected by geographical origin. These results are in agreement with Fredes and Montenegro (2006). The levels of Fe, Mn, Zn and Pb in cotton honey collected from Kafr El-Sheikh Province ranged 20.00 - 44.50, 0.42 - 1.40, 1.20 - 3.28 and 0.50 - 2.37 mg/kg, respectively. These ranges are nearly similar to those obtained in other regions by Serra (1989), Terrab et al. (2004), Fernández-Torres et al. (2005), Hernández et al. (2005), Rateb (2005) and Taha et al. (2010). Significant ($P < 0.05$) differences among investigated regions were found. The total content of mineral substances depends on a location (Staniškienė et al., 2006).

Table (1). Heavy metals content in cotton honey collected from different regions in Kafr El-Sheikh, Egypt during 2011 season

Region	Mean (mg/kg) Range (mg/kg)							
	Iron Fe	Manganese Mn	Zinc Zn	Lead Pb	Cadmium Cd	Copper Cu	Nickel Ni	Chromium Cr
Mutubas	32.72^{ab} (29.5-37.00)	1.08^{a} (0.83-1.40)	1.63^{b} (1.20-2.10)	1.23^{bc} (0.87-1.83)	ND	ND	ND	ND
Fowah	33.28^{ab} (31.00-36.50)	1.17^{a} (1.11-1.37)	2.33^{ab} (2.00-2.50)	1.88^{ab} (1.42-2.33)	ND	ND	ND	ND
Disuq	38.82^{a} (36.50-42.00)	1.11^{a} (0.93-1.37)	2.03^{ab} (1.80-2.30)	1.50^{abc} (1.33-1.79)	ND	ND	ND	ND
Qallin	30.82^{b} (30.00-32.00)	1.11^{a} (0.77-1.28)	2.27^{ab} (2.30-2.40)	1.34^{abc} (1.04-1.66)	ND	ND	ND	ND
Sidi Salem	35.56^{ab} (36.00-38.00)	1.09^{a} (0.86-1.40)	2.57^{a} (2.40-2.80)	1.67^{abc} (1.59-1.75)	ND	ND	ND	ND
Kafrelsheikh	22.44^{c} (20.00-25.00)	0.96^{a} (0.64-1.40)	2.07^{ab} (1.60-2.60)	0.86^{c} (0.50-1.29)	ND	ND	ND	ND
Riyadh	24.83^{c} (20.00-30.00)	0.46^{b} (0.42-0.54)	2.66^{a} (2.10-3.28)	2.11^{a} (1.96-2.37)	ND	ND	ND	ND
Al-Hamool	34.00^{ab} (29.00-44.50)	0.83^{ab} (0.64-1.09)	2.03^{ab} (2.00-2.10)	1.68^{abc} (1.29-2.37)	ND	ND	ND	ND
Beila	21.50^{c} (21.00-22.00)	0.73^{ab} (0.68-0.77)	2.40^{a} (2.10-2.80)	1.48^{abc} (0.75-2.25)	ND	ND	ND	ND
Significant Level	**	*	*	*	NS	NS	NS	NS
Grand mean	30.44	0.95	2.22	1.53	ND	ND	ND	ND

- ND, below detection limit (not detected).
- Different letters a,b,c in the columns indicate the significance of differences at different levels.
- **, * and NS indicate $P < 0.01$, $P < 0.05$ and non-significant, respectively.

Frequent adding of the micronutrient Znas to the field crops in Riyadh region may be lead to increase Zn element in honey collected from this area. Honey contact with galvanized containers surfaces during extracting, and/or preparation for the market, can be a source of Zn contamination in honey (González et al., 2000; Bogdanov et al., 2003).

The high level of Pb in honey collected from bee colonies of Riyadh region may be due to vicinity of some apiaries to the traffic roads, where large quantities of exhausts from cars. Atmospheric pollutant deposition on plants and may be absorb together with flower nectar and pollute combs through bees' contaminated bodies, or contaminate water and pollen which collected by bees to the hive (Porrini et al, 2003). One easy way of Pb to enter in our food chain is throughout their spread over flowering plants (Fakhimzadeh and Lodenius, 2000). Polluted pollen will raise the level of metals in honey and other hive products (Zugravu et al., 2009).

Although, the observed values in the present study was less than the acceptable limits. They were higher than those reported in other geographical regions by different authors. This may be due to the presence of some apiaries near the traffic roads and the variations in fuel quality used in Egypt.

Generally, the grand averages of Fe, Mn, Zn and Pb in all tested cotton honey samples were 30.44, 0.95, 2.22 and 1.53 mg/kg, respectively. These values except of Fe are nearly similar to those found in Brazil which were 2.79, 3.88 and 2.34 mg/kg for Fe, Mn and Zn, respectively (Mattoset al., 1998), Turkey which were 0.36 and 0.32 mg/kg for Fe and Mn, respectively (Erbilir and Erdoqrul, 2005) and Egyptian clover honey that were 25.75, 0.41, 1.78 and 0.90 mg/kg for Fe, Mn, Zn and Pb, respectively (Taha et al., 2010).

Lead content (mg/kg) in honey was stated to be 0.2 in Switzerland (Bogdanov et al., 1986), 0.06 in Turkish honey (Uren et al., 1997), 0.00 - 0.27 in Spain (Vinas et al., 1997), 0.03 - 0.38 in Egyptian honey (Nour, 1998), 0.37 in Finland (Fakhimzadeh and Lodenius, 1999), 0.02 - 1.00 in Poland (Belechovski and Vorlovl, 2001), 0.01–1.80 in Switzerland (Bogdanov, 2006). Studies from Switzerland have shown that heavy metals presence in hive products was high in industrial and heavy traffic area (Bogdanov et al., 2003).

Significant positive correlations at probably ($P < 0.01$) were observed between Fe and Mn as well as between Zn and Pb (Table 2). Similar results were reported by Taha et al. (2010) who found highly significant positive correlations between Fe, Mn, Zn and Pb in clover honey apart from that between Mn and Pb which was significant only at ($P < 0.05$). Besides, Rateb (2005) and Bogdanov et al. (2007) observed significant positive correlations between Fe, Zn, Mn and Pb in honey. Significant negative correlation between Mn and Zn was detected at ($P < 0.05$) (Table 2). This result was disagree with the findings of Tahaet al. (2010) who found highly significant positive correlations between Mn and Zn in clover honey.

The elements (Cu, Cd, Ni and Cr) were not detected in all examined honey samples. These results are in agreement with those obtained by Badei and Shawer (1986) for Cu, Nour (1998), Rateb (2005) and Sheref (2007) for Cd and Taha et al. (2010) for Cu, Cd, Ni and Cr in honey. The absence of Cd in Egyptian honey may be due to the decrease of the element in alkaline soil (the soil of Kafr El-Sheikh Province). Egypt is one of the lowest "international Cd field", the Egyptian national mean and median Cd values are only about half of the respective international parameters (Sillapââ and Jansson, 1992). On the other hand, Cu (mg/kg) content in honey was stated to be 0.54 in Brazil (Mattoset al., 1998), 0.06-1.55 in Czech (Čelechovská and Vorlová, 2001) and 0.01 in Turkey (Erbilir and Erdoqrul, 2005).Following Ni (mg/kg) levels have been found in honey: 0.004-3.23 in Italy (Porriniet al., 2002), < 0.001in France (Devillers et al., 2002a) and 0.09-0.34 (Devillerset al., 2002b). Also, Cd(mg/kg) content in honey was varied according to geographical origin, the recorded values were 4.53 in Turkey (Uren et al.,1997), 0.00-5.4 in Spain (Vinas et al.,1997), 0.15 in Finland (Fakhimzadeh and Lodenius, 1999),0.00-0.08 in Poland (Belechovski andVorlovl, 2001), 0.03 in Turkey (Erbilir and Erdoqrul, 2005) and 0.5-77.4 μ g/kg in Czech (Čelechovská and Vorlová, 2001).

Table (2). Pearson correlation coefficients for heavy metals of cotton honey collected from different regions in Kafr El-Sheikh Province

	Fe	Mn	Zn	Pb
Iron (Fe)				
Manganese (Mn)	0.50**			
Zinc (Zn)	-0.19	-0.44*		
Lead (Pb)	0.12	-0.32	0.55**	

- * and ** Correlation is significant at the 0.05 and 0.01 level (2-tailed), respectively

CONCLUSION

From this study, it can be concluded that the heavy metals content of honey significantly depended on the area from where it was harvested. Honey samples collected from the same botanical origin but from different geographical locations were differed in heavy metals content. The highest levels of Fe (38.82 mg/kg), Mn (1.17 mg/kg) were found in Disuq and Fowah regions, respectively, while the highest levels of Zn (2.66 mg/kg) and Pb (2.11 mg/kg) were obtained from Riyadh region.

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تقدير محتوى المعادن الثقيلة في عسل القطن في محافظة كفر الشيخ– مصر

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تم جمع عينات من عسل القطن من تسع مناطق ممثلة لمحافظة كفر الشيخ، مصر خلال موسم قطن ٢٠١١، لتقدير بعض العناصر الثقيلة في عسل القطن. لظهرت النتائج المتحصل عليها أن كل عينات العسل تحتوي على أربعة عناصر معدنية ثقيلة هي (الحديد، المنجنيز، الزنك والرصاص) ومستوياتها بالعسل كانت أقل من الحدود المسموح بها وقد تراوحت قيم هذه العناصر بين ٢٠,٠٠– ٤٤,٥٠، ٠,٤٢– ١,٤٠، ١,٢٠–٣,٢٨ و ٠,٥٠–٢,٣٧ مجم/كجم، على الترتيب. ولم تكتشف أي من العناصر المعدنية الخطرة (النحاس، الكادميوم، النيكل والكروم) في كل العينات. ولوحظت ارتباطات موجبة ذات معنوية إحصائية عالية على مستوى إحتمالية ١% بين عنصري الحديد والمنجنيز وكذلك بين عنصري الزنك والرصاص. وقد أستنتج أن محتوى المعادن الثقيلة في عسل القطن في محافظة كفر الشيخ تعتمد بشكل ملحوظ على المنطقة التي جُمع منها.

قام بتحكيم البحث

أ.د / حسن محمد فتحى	**كلية الزراعة – جامعة المنصورة**
أ.د / محمد السعيد الشريف	**كلية الزراعة – جامعة عين شمس**