RJC

RASĀYAN J. Chem.
Vol. 13 | No. 4 |2036-2044| October - December | 2020
ISSN: 0974-1496 | e-ISSN: 0976-0083 | CODEN: RJCABP
http://www.rasayanjournal.com
http://www.rasayanjournal.co.in

HEAVY METALS IN HONEY PRODUCED IN SOME LOCALITIES IN KOSOVO

M. Fadil[1], D. Krasniqi[2] and M. Ahmet[3,*]

[1]Faculty of Agriculture and Veterinary/University "Hasan Prishtina", Prishtina,10000, Kosovo
[2]Analytical, Food and Agriculture Laboratory/Phytosanitary Corporate,
Sara & Meti, 10000, Kosovo
[3]Faculty of Agribusiness/ University of Haxhi Zeka, Peja, 30000, Kosovo
*E-mail: ahmet_musa@live.com

ABSTRACT

Contamination of the honey with regards to heavy metals is important not just for quality control, but also for the assessment of environmental contamination. The goal of these investigations is to assess the levels of seven heavy metals in honey produced in Kosovo. In the territory of Kosovo in the year of 2018-2019, four types of honey (acacia, pine, flowers and meadows) were collected at four sites (Deçan, Mitrovicë, Podujevë and Prishtinë) to assess the concentration of the following metals: zinc (Zn), copper (Cu), iron (Fe), chromium (Cr), manganese (Mn), lead (Pb), as well cadmium (Cd). In total there were taken 80 samples (Deçan n=20, Podujevë n=20, Mitrovicë n=20 and Prishtinë n=20). Atomic absorption spectrophotometer was used to assess heavy metals. The results have indicated that the highest concentrations of zinc were recorded in pine honey (3.700 mg/kg), copper in meadow honey (0.935 mg/kg), iron in flower honey (5.610 mg/kg), chromium in meadow honey (0.199 mg/kg), manganese in flower honey (11.500 mg/kg), lead in meadow honey (6.100 mg/kg) and cadmium in flower honey (0.349 mg/kg). Kosovo honey is not free of heavy metals but the levels of them were found below the allowed levels, with exception to the Mitrovicë. The data obtained indicated that honey is a very suitable material that can be useful for the evaluation of the presence of the contaminant in the surrounding environment.

Keywords: Honey, Heavy Metals, Locality, Mitrovicë, Acacia.

INTRODUCTION

Kosovo is situated geographically in Balkan Peninsula. The country is bordered with Albania, Montenegro, Serbia and Macedonia and has 41º50'58'' and 43º15'42'' longitudes and 20º01'30'' and 21º48'02'' latitudes. Beekeeping as a branch of the economy recently has been widely spread in Kosovo. The country possesses good climate conditions to develop the beekeeping sector. Given the climate, terrain and numerous types of plants from different botanical origins Kosovo have great potential to achieve a good production of honey and other bees' products. In 2018, the number of beehives was 182,476 which in contrast to 2017 have increased by 11%. Kosovo's honey production ranges from about 12-15 kg per hive resulting in a total honey production of 2,120 tons in 2019. The import of honey in 2018 was 209 tons while the export of honey this year was only symbolic.[1] Domestic consumption per capita was 1.3 kg and domestic production met about 92% of consumption needs while the rest was met by imports. The average annual consumption per capita is constantly increasing due to the population's awareness of the many nutritional values of honey.

Honey is a very heterogenic sweet liquid that is naturally derived from pollen and nectar gathered from honey bees which are considered very suitable food for their survival. Some authors indicated that the plants have flowers that contain nectar and pollen needed by bees as a source of feed.[2]

The honey flavor, taste, color and texture depend on the floral nectar from which it is derived and is used in many types of food products as an ingredient.[3]

In daily diets metals in the trace are highly important, because of their essential nutritious value and of course their possible effects to harmful organisms. Several elements like Zn, Fe, Cu, Mn, etc. are essential because they play important role in biological systems while Pb and Cd are considered non-essential elements that even in trace amounts can be toxic.[4]

Because of the fact they are difficult degradable and toxic, these metals are extremely serious pollutants. Heavy metal is defined as any metal that has specific gravity above 4.00 g/cm^3 and is toxic even at low concentration.[5]

Environmental factors, botanical as well as geographical have a great impact on the concentrations of the metals in honey, and it is better to consider all of these factors when studying honey metal composition.[6]

These contaminants may come from different external sources like mines, industrial and urban areas, which are close to the bees' forage area, or maybe introduced by inappropriate procedures during processing and storage of the honey[7]. Because of the acidic reaction of honey, the tools and containers of metallic origin may release elements such as Cr, Pb.[7]

The level of the metals in different types of the honey is closely related to its botanical as the well geographical area in which it originated, but also it can be influenced by activities of the anthropogenic nature.[8,9] Heavy metals are present in the soil, and air, mainly from industrial activities as well the traffic which might contaminate the bee and its products.[10] The heavy metal present in the air might be deposited on the bee bodies and after to be taken back at the beehive with pollen, or maybe absorbed via the nectar from plant flowers, or through the drinking water.[11]

Dark color honey (chestnut, heather, avocado, etc.) contain higher quantities of certain metals compared to pale honeys.[12]

To our knowledge, heavy metal concentration in honey produced in Kosovo was not investigated previously. Humans are in turn exposed to heavy metals via consuming contaminated honey, fish, fruits, vegetables, and this might result in various biochemical disorders.[5,13-15] These researches aim to assess the level of heavy metals of four types of honey from four localities of Kosovo which are indicators for environmental pollution.

EXPERIMENTAL

Material and Methods

Sample Collection

Eighty (80) honey samples in total from different botanical origins and sampling sites were collected in the year of 2018 and 2019 from four areas of Kosovo, Deçan, Podujevë, Mitrovicë and Prishtinë to study their levels of heavy metals. Honey samples directly were taken from the beekeepers at the farm level. The honey types are acacia (n = 20), pine (n = 20), floral (n = 20) and meadow (n = 20). Samples in the quantity each of 500 g were taken in jars and were put in a dark, at the temperature 4-8 °C until they were analyzed.

Through wet digestion procedures, the samples of 5 g each of all honey were treated with a 4 ml of the mixture of concentrated HNO_3 and concentrated $HClO_4$ at a temperature of 240°C for over 3 hour.[16] The samples after mineralization were mixed with distilled water and later on filtered with filter paper 0.45 µm. The filtrate was set at 50 cm^3 in a volumetric balloon and leveled up to the mark with double distilled water. Concentrations of different metals in honey samples were assessed through Atomic Absorption Spectrophotometer, model 1100 from Perkin-Elmer (MA, Boston from the USA).

Reagents

Pure analytical grade reagents from well-known company Sigma Aldrich, German, were used during this research. In all solutions, we have utilized distilled water that was produced through a system suitable for water purification (Thermofisher, Germany).

Calibration of the Equipment

The AAS was calibrated and later on standardized using different working standards. After we have confirmed that AAS has been correctly calibrated, we started to measure concentrations of the heavy metals in each honey sample. The standard solutions concerning each metal were obtained through dilution of stock solution that contained 1000 mg/L of Zn, Cu, Fe, Cr, Mn, Pb, Cd. To prepare element standard solutions of respective metals used for calibration, stock solutions of 1,000 mg/liter Zn, Cu, Fe, Cr, Mn, Pb as well Cd (Sigma Aldrich) were diluted in HNO_3. These serial dilutions for each metal were prepared fresh during the day we have performed the analysis.

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Recovery Rate

Since we did not have appropriate reference materials certified for honey, the rate of the recovery was obtained using a known spiked concentration of all the studied metals. The recoveries in percentages of the analyzed metals were satisfactory and were 99.4 to 103 % (Table-1), signifying good precision, validity and accuracy of the method used.

Table-1: Recovery Rate of the Analyzed Heavy Metals in the Spiked Samples

Metal	Spiked (mg/kg)	Measured (mg/kg)	Recovery (%)
Zn	10	9.98	99.8
Cu	10	10.3	102.0
Fe	10	9.99	99.9
Cr	10	99.6	99.6
Mn	10	10.4	10.4
Pb	10	9.97	99.7
Cd	10	9.94	99.4
As	10	10.2	103.0

Statistical Analysis

The data obtained from these researches were analyzed statistically through analysis of variance (ANOVA) using an appropriate software MSTAT-C from Michigan University, USA. The ANOVA and LSD- tests were utilized to assess the differences of metal concentrations in honey, whereas the significance level for the differences was set at $p<0.05$ and $p<0.01$ as well. Interaction factors were calculated from the table of ANOVA which was according to split-plot experimental design. Factor A represents metals (Zn, Cu, Fe, Cr, Mn, Pb and Cd), Factor B represents localities (Deçan, Podujevë, Mitrovicë and Prishtinë). Interaction between factors was AxB (metal x locality).

RESULTS AND DISCUSSION

Honey is a complex product whose properties as well composition depend primarily from the nectar of plant species, however, other factors worth to be mentioned are bee species, season, geographical area, the method of storage and even the technology and harvest conditions.[17,18] The level of heavy metals in honey has been the subject of research by many authors around the world.[19-21] This study is one of the few that presents the results concerning trace elements in honey species that are specific to the Kosovo climate and are often used as medicinal preparations for various problems to the health.

Table-2 shows the concentrations of metals in four honey types (acacia, pine, flower and meadows) from different localities of Kosovo.

Table-2: Heavy Metal Contents in Different Kinds of Honey

Locality	Honey	Content mg/kg						
		Zn	Cu	Fe	Cr	Mn	Pb	Cd
Deçan	Acacia	0.205[1]	0.257	0.310	0.004	0.188	0.038	0.002
	Pine	0.726	0.389	3.195	0.032	0.373	0.080	0.035
	Flower	1.106	0.730	1.150	0.001	1.550	0.012	0.200
	Meadow	0.712	0.318	0.816	0.023	1.149	0.053	0.001
Podujevë	Acacia	0.531	0.180	0.641	0.001	0.260	0.010	0.006
	Pine	0.233	0.060	0.185	0.021	0.020	0.010	0.010
	Flower	0.506	0.012	0.317	0.001	0.002	0.013	0.001
	Meadow	1.313	0.570	0.411	0.001	0.010	0.017	0.001
Mitrovicë	Acacia	1.810	0.341	0.838	0.058	1.370	3.120	0.031
	Pine	3.700	0.514	2.345	0.073	1.740	2.460	0.068
	Flower	2.100	0.726	5.610	0.156	11.500	1.750	0.349
	Meadow	1.700	0.935	3.973	0.199	8.755	6.100	0.017
Prishtinë	Acacia	0.800	0.232	2.082	0.001	0.123	0.127	0.001
	Pine	1.400	0.138	0.950	0.001	0.314	0.005	0.001
	Flower	0.700	0.097	2.134	0.004	0.002	0.090	0.003
	Meadow	0.900	0.219	1.707	0.002	0.150	0.130	0.001
Average		1.150	0.360	1.670	0.040	1.720	0.880	0.050

Note: [1]The results represent the average value of measurements from five replications

Concentrations of metals in honey varied highly and ranged as follows (mg/kg): Zn 0.205-3.700; Cu 0.012-0.935; Fe 0.185-5.610; Cr 0.001-0.199; Mn 0.010-11.500; Pb 0.010-6.100 and Cd 0.001-0.349 (Tab.2). Variation of the content of metals in honey has been shown also from the other author's worldwide.[22-24]

Zinc (Zn), this element is reported to be a part of 200 different enzymes. It has a role in nucleic acids metabolism, in the synthesis of proteins, in the replication of the cells, tissue repair, bone mineralization and proper distribution of vitamin A. Low content of Zn in the body causes anemia, slowing down the growth rate, and if lack in children causes lower growth and slower mental development. In this research, it was determined in the concentration from 0.205 to 3.700 mg/kg. The Zn concentrations (Fig.-1) found ranged according to the mean values in the order (mg/kg): pine honey (1.515) > meadow honey (1.156) > flower honey (1.103) > acacia honey (0.837). The highest concentration as a mean was seen in honey from pine and the lowest one in honey from acacia. In Croatia the highest concentrations were measured in forest honey (2.95 mg/kg), while the mean values for other honey types ranged in this order (m/kg): flower honey (1.22) > sage (0.94) > chestnut (0.89) > acacia (0.55), whereas according to some authors the zinc concentration in honey ranged from 1.734 to 245.205, which level is far away from our results.[25-27]

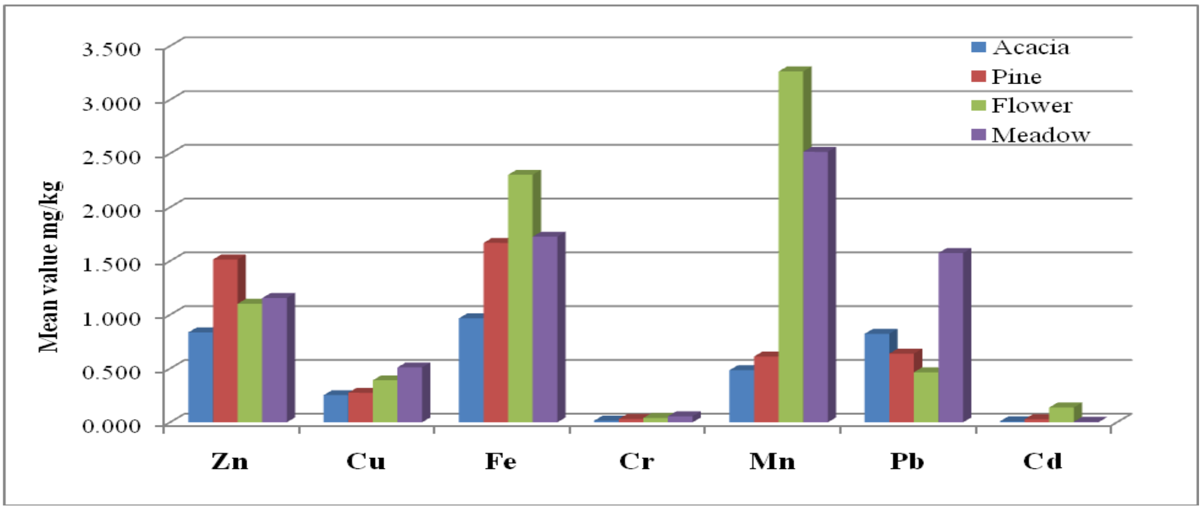

Fig.-1: Metal Concentration by Honey Type

Copper (Cu), as a trace element is important in human health and its role is regarded as the Fe absorption and in this respect is essential for erythrocytes production in blood. The deficiency of the Cu in the organisms causes anemia. In this research (Fig.-1), Cu concentrations ranged from 0.012-0.935 mg/kg, with mean values in this order (mg/kg): meadow honey (0.511) > flower honey (0.391) > pine honey (0.275) > acacia honey (0.253). Opposite to our results, there are some reports from different authors. The Cu concentrations of honey found in Croatia are significantly higher than in this study, and the mean values are in this order (mg/kg): forest honey (19.600) > acacia (18.600) > sage (7.850) > chestnut (6.190) > flower honey (4.380).[25] Even in Turkey were reported higher average values of Cu content in honey (0.223 to 198.361).[26,28] Iron (Fe), is an important element for humans and very essential for plants. It is contained in all body cells and as a part of hemoglobin as well myoglobin is irreplaceable concerning the oxygen transport in the blood and to the muscles. In the case of Fe deficiency in the body causes anemia. In this research (Table-2), Fe was determined at concentrations of 0.185 to 5.610 mg/kg. The highest value was observed in the flower honey sample. The order of mean Fe values (Fig.-1) in different brands of honey is (mg/kg): flower honey (2.303) > meadow honey (1.727) > pine honey (1.669) > acacia (0.968). However, as reported Fe concentrations (mg/kg), in Croatia, are found to some extent similar to our results and the mean metal value in all honey types was recorded as follows: forest 5.170, sage 4.170, chestnut 3.570, floral 3.290 and acacia 2.770.[25] Significantly higher mean values of Fe content have been reported from others and were from 3.506 to 1278.778 mg/kg.[26]

Chromium (Cr), as an element in the trace, has an important role regarding the metabolism of the carbohydrates, as a part of glucose tolerance factors, and according to the other authors, is included in the cardiovascular risk and metabolic syndrome.[29] Mean concentrations of Cr in honey in this study were 0.056 to meadow honey, 0.041 to flower honey, 0.032 to pine honey and 0.016 to acacia honey (Fig.-1). In Turkey, they were determined in the ranges of 2.4-37.7 and 0.126-7.964 from other authors.[26,30]

Manganese (Mn) as a metal is an element that is essential for organisms and is an ingredient that is a part of various enzymes. Mn is important regarding bone development, cholesterol metabolism, sterility prevention, proper pancreatic function. In the four types of honey in this study, Mn was determined from 0.010 to 11.500 mg/kg. The lowest mean value was recorded in honey from acacia (0.485 mg/kg) whereas the highest in flower honey (3.264 mg/kg). Opposite of our results in Turkey was reported a higher concentration of this metal in honey (0.096-29.496).[26]

Lead (Pb), in this research, ranged as a mean concentration (mg/kg) in the following order. The highest concentrations of Pb (1.575 mg/kg) was determined in meadow honey (Fig.-1), which is lower than the mean value of 289 mg/kg in flower honey reported from Italy or in the range 31.5 to 46.32 mg/kg in floral species from Tenerife[31,32]. Higher Pb concentrations were determined also in honey of other countries (mg/kg): Hungary12-99 mg/kg, Turkey 1.51-55.3 mg/kg.[33-35]

Cadmium (Cd) also was determined in this study. The highest concentrations of Cd of 0.138 mg/kg as a mean value were recorded in flower honey and were 4.78 to 27.6 times higher compared to that in other honey types (Fig.-1). Similar high Cd concentrations from the range of 10.9-17.9 mg/kg were recorded in flowering honey types in Turkey and 8-18 mg/kg in Italy, respectively.[4,30,31]

Metal concentrations in different kinds of honey as for the localities are presented in Fig.-2. The values presented are the mean content recorded for each respective element. The data regarding heavy metals concentrations were compared with the data established by EU legislation[36].

The Zn content ranged between 0.646 and 2.328 mg/kg, with a mean concentration of 1.153 mg/kg. The highest level of Zn was recorded in honey samples from Mitrovicë (2.328 mg/kg). The values are more or less in similar range with values reported in Italian honeys (3.205 mg/kg), Slovenian honeys (3.610 mg/kg), in Romanian honey (0.1477-1.48 mg/kg), in China (1.329 mg/kg as a mean) and in case of Iran honey (2.03-6.54 mg/kg).[37-41]

The Cu content ranged from 0.172 mg/kg in Prishtinë and 0.629 mg/kg from Mitrovicë, with a mean concentration of 0.358 mg/kg. The Cu content in honey from other countries ranged more widely, as 1.074 mg/kg in Croatian honey, 0.900 mg/kg in Italian honey, 0.25–1.10 mg/kg in Turkey and 3.22 mg/kg in Slovenian honeys.[9,30,41,42] The copper can be introduced in honey from the copper fungicides that are used in agriculture as well via agricultural chemicals.

The iron is second the most present metal analyzed in honey. The iron level ranged from 0.389 mg/kg in Podujevë to 3.192 mg/kg in Mitrovicë, with an average concentration of 1.667 mg/kg. Fe content was not in a similar range with the data that were reported in honey from other countries, United Arab Emirates (1.15–110.79 mg/kg), Turkey (1.8–10.2 mg/kg), Canary Island (0.40–52.51 mg/kg), in Thailand (as a mean of 2.77 mg/kg) and Saudi honey (8.86–13.3 mg/kg).[30,43-46]

The highest concentration of Cr was observed in the case of honey samples from Mitrovicë (0.122 mg/kg). The Cr content was between 0.002 and 0.122 mg/kg, with a mean concentration of 0.036 mg/kg. The honey analyzed contained Mn from 0.073 mg/kg in Podujevë to 5.841 mg/kg in honey from Mitrovicë, with a mean concentration of 1.719 mg/kg. The values are in line with those reported from Czech Republic (1.16–8.89 mg/kg), United Arab Emirates (below <0.01–10.31) and Turkey (0.32–4.56 mg/kg).[30,44,47] The Pb content ranged between 0.013 mg/kg in Podujevë and 3.358 mg/kg from Mitrovicë, with an average concentration of 0.876 mg/kg. None of the samples analyzed exceed the maximum allowed level established by the UE regulations (0.05 mg/kg) [36].

The Cd content ranged from 0.002 to 0.116 mg/kg in honey from Prishtina and Mitrovicë, with a mean value of 0.046 mg/kg. These data are fair away lower comparing to the maximum set by EU legislation (0.05 mg/kg). The Cd content in honey was different from one country to another, in Turkey and Croatia it ranges from 2.1–38.0 μg/kg and in Poland it is 15.0 μg/kg.[30,48-50] The values reported for Cd in this study were in the same range as the honey from China and Turkey.[30, 39]

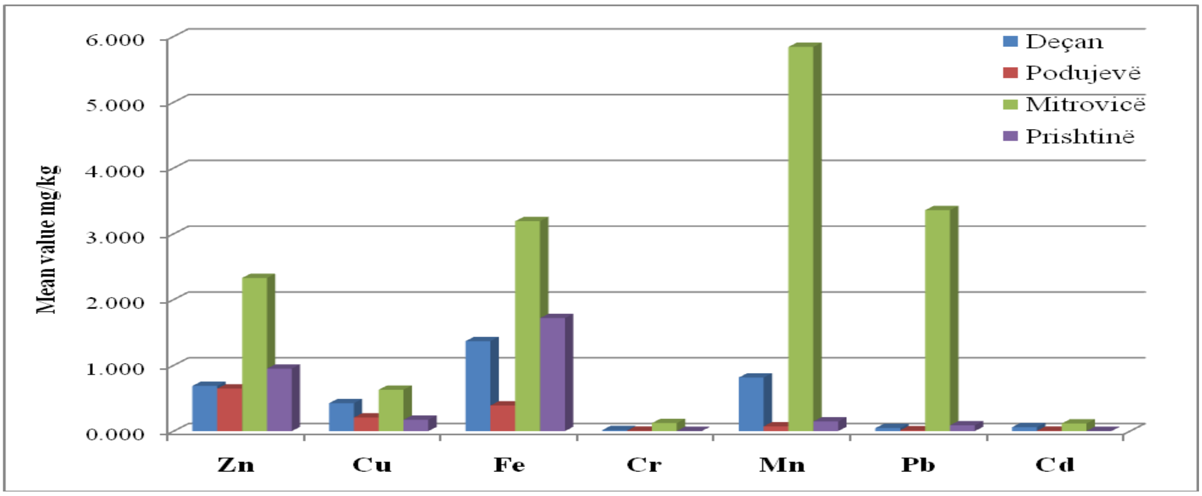

Fig.-2: Metal Concentration in Honey by Localities

Based on the results of the ANOVA (Table-3), it was shown that there exist significant differences of different levels regarding the content of metals (Factor A), the locality (Factor B) and factor interactions (AxB).

Regardless of the type of metals (Factor A) the highest concentration was found in Mn (1.719 mg/kg) whereas the lowest in Cd (0.046 mg/kg). The differences found statistically are highly significant. As for other metals, their concentration as an average value was different as well. Thus the differences between the concentrations of Mn and Zn are not significant and the values are approximate (1.719 and 1.667 mg/kg). The differences in metal concentration between Cr and Cd are not as significant as between Zn and Pb (Table-3).

Significant differences are shown also concerning the localities where the samples of the honey were taken for analysis (Factor B). The content of heavy metals as average was higher in Mitrovicë (2.227 mg/kg), while the lowest in analyzed honey samples of locality Prishtinë (0.487 mg/kg), and the differences observed are highly significant.

Table-3: Metal Concentration (mg/kg) in Honey of Different Localities- ANOVA

Metals (A)	Locality (B) Deçan	Podujevë	Mitrovicë	Prishtinë	Mean (A)
Zn	0.687	0.683	2.328	0.950	1.162
Cu	0.424	0.206	0.629	0.172	0.358
Fe	1.368	0.389	3.192	1.718	1.667
Cr	0.015	0.006	0.122	0.002	0.036Ns
Mn	0.815	0.073	5.841	0.147	1.719**
Pb	0.046	0.013	3.358	0.421	0.960
Cd	0.060	0.005	0.116	0.002	0.046Ns
Mean (B)	0.488*	0.196*	2.227**	0.487**	Interaction A x B

Faktor		A	B	A x B
LSD	1%	0.3214	0.2235	0.6093
	5 %	0.4506	0.2942	0.8201

Legend: Ns = No significant, * = significant, ** = highly significant

Differences about metal content in the locality of Deçan and Prishtinë are not significant and the values of the mean concentration ranged as 0.488 and 0.487 mg/kg. Differences between honey samples from Deçan and Prishtinë compared to Podujevë are significant and the ranges were 0.488 mg/kg in Deçan, 0.487 mg/kg in Prishtinë, whereas in Podujevë 0.196 mg/kg.

According to our opinion, we have expected this given that in Mitrovicë there is a metallurgical plant "Trepça" where zinc, manganese, iron and lead are produced and the contamination of air, soil as well water is higher compared with other localities involved in these researches.
Regarding factor interaction, AxB, statistically significant differences of different levels of significances were found as well (Table-3).

CONCLUSION

Ecosystem pollution via heavy metals recently has become a global concern and this is due to its adverse effect on living organisms when they are in contact with these metals. They can be toxic and of a detrimental effect since they possess the ability to accumulate and retain inside the body of different organisms. The data we have obtained through these researches showed that the different honey types coming from four localities of Kosovo have had different metal compositions. Regarding the study on the monitoring of metal concentrations in honey the analysis of this research showed the presence of the metals following this order: Mn (1.720 mg/kg), Fe (1.670 mg/kg), Zinc (1.150 mg/kg), Pb (0.880 mg/kg), Cu (0.360 mg/kg), Cd (0.046 mg/kg) and Cr (0.040 mg/kg). Honey contamination with Cd was found less than the minimum permissible limits set by EU (0.05 mg/kg), whereas Pb was higher than the limit. Honey produced in different regions of Kosovo does not completely lack heavy metals but they are at a satisfactory level and good quality for human consumption. The highest value of metals was recorded in honey produced in Mitrovicë (2.227 mg/kg) comparing to the other localities where the values were lower and as a mean was in this order, Deçan (0.488 mg/kg), Podujevë (0.196 mg/kg) and Prishtinë (0.487 mg/kg). The high contamination of the honey in Mitrovicë to our opinion is due to the proximity of metal industry which is located here, use of the huge amount of fertilizers in agriculture crops, and perhaps other factors that for sure might influence. Since humans use honey as food this kind of research will be very important for risk assessment by relevant institutions that are responsible to monitor environmental pollution, food safety as well as food security in our country.

ACKNOWLEDGMENT

We wish to thank the staff of the Faculty of Agriculture and Veterinary, the University of Prishtina for their immense help and assistance in the laboratory and fieldwork

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