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Original Research

Determination of Pesticides, Heavy Metals, Radioactive Substances, and Antibiotic Residues in Honey

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Abstract

During the war in Bosnia and Herzegovina a large number of chemicals were released into the environment. This research was conducted to evaluate the health safety of honey (meadow, chestnut, acacia, amphorae, and honeydew) produced at 18 different locations in the region of Una-Sana Canton in the northwestern part of Bosnia and Herzegovina. We determined the pesticides, heavy metals, radioactive elements, and antibiotic residues in 46 honey samples. The content of pesticides and heavy metals (Pb, Cd, As) were not found, and the concentration of Cu, Fe, Zn in all honey samples was in accordance with EU standards. Statistically significant differences (ANOVA) in honey samples were found according to concentrations of Cu (0.09-0.18 mg/kg), Fe (0.29-0.92 mg/kg), and Zn (0.18-1.38 mg/kg). The radionuclide content was determined by means of low-level (<5 Bq/kg), using high-resolution gamma-spectroscopy (^{137}Cs). Antibiotic residues were carried out by disc plate method of microbiological antibiotic assay and all honey samples were negative. The results of this work indicate that this area is not polluted and is suitable for the development of beekeeping.

Keywords: honey, health safety, pollution

Introduction

In the past decades an intensive development has taken place in human activities, which globally resulted in enhanced contamination of the environment. That is why research deals more frequently with various contaminants that affect human health through the food chain. The question arises whether this problem exists in the region of Una-Sana Canton (USC), in the northwestern part of Bosnia and Herzegovina (B&H). In recent years several publications have confirmed that the area, with its population of 300,000 in 4,125 km^2, falls into the non-contaminated category [1-3]. As a result, 225 km^2 of the area were pronounced to be protected, with very high quality ratings according to IUCN criteria. Our aim was to do further research on health safety and analyze 46 samples of honey in 18 different locations in the above-mentioned area.

Our research was based on the knowledge that bees and their products are excellent biological indicators of pollution and of chemical impact on the environment. Heavy metals, pesticides, radioactivity, and antibiotics all endanger bee families. Pesticides might cause a high mortality rate among bees. The presence of contaminants such as pesticides, heavy metals, radionuclide, or their residues in tis-

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sues of bees and in honey or other bee products can be discovered by suitable laboratory analyses [4, 5]. The application of antibiotics and other chemicals in the treatment of bee diseases [6] is also considered as a threat, and this was the reason 280,000 tons of honey was removed from the market in 2005 worldwide [7].

However, honey can also be contaminated by less direct activities [8]. Heavy metals, which appear in the environment as a result of industrial activity and traffic, can contaminate bee colonies and their products. Lead and chrome contents of honey have been widely investigated. In the last decade all research results show lower concentration than 1 mg/kg in the case of lead and 0.1 mg/kg in the case of cadmium, which are MRL proposed for the EU [9]. Lead concentration in honey ranges from 0.012 mg/kg in Turkey [10] to 0.28 mg/kg in Spain [11], while cadmium concentration ranges from 0.0016 mg/kg in Hungarian [12] to 0.04 mg/kg in Spanish samples. Chrome concentration is also low, from 0.0018 to 4.25 mg/kg in Hungarian [12] and Polish samples [13], respectively. In the case of arsenic, low values can be found between 0.0066 and 0.05 mg/kg, where the lowest value refers to honey from Italy [14], and the maximum value to honey from Spain [11].

While heavy metal concentration in honey has been researched widely, literature data about the presence of pesticides in honey are sparse. This presence has been examined in several areas in Italy, providing information about the diffusion of pesticides within agricultural environments, and as a side effect of crop protection practices [15-17] in Greece [18]. Most results show that residues range from 0.001 to 0.005 mg/kg.

In the case of organophosphates and organochlorine, the investigation discovered that none of these showed higher values than the detection limit, which varied between 0.005 and 0.050 mg/kg [19] while in the case of imidacloprid the values were even below the limit of detection devices from 0.002 mg/kg [20, 21].

After the nuclear disaster in Chernobyl, research and studies on radioactivity in honey mainly focussed on ^{137}Cs. The results reported values between 8 and 51 Bq/kg for Slovenian, and between 1 and 21 Bq/kg for Croatian honey [22].

In the case of antibiotics, specific MRL values have not been known because their presence in honey and bee products was not expected. Literature data suggests that although the presence of acaricide (coumaphos, chlorfenvinphos, fluvalinate, amitraz) and antibiotics (tetracyclines, oxitetracyclines) can be detected in the case of coumaphos in 14% of the samples, the detected residues do not represent a risk for food safety [23]. In 80 honey samples from Germany, using two methods by which the limit of detection is defined to 0.07 μg/kg, antibiotics could not be detected [24].

USC is an area in which the production of honey and bee products has been defined as promising, which is supported by the fact that from 2005 up to the present the number of registered beekeepers has risen from about 200 to approximately 1,000 [25]. On the other hand, the production of honey has grown only from 264 tons in 2005 to 325 tons in 2008 [26]. The number of bee families in 2008 was 27,410. The control of honey and bee products is carried out by the Veterinary Institute of USC, which serves as the referential institution accredited by BAS EN ISO/IEC 9001:2001 and 17025:2006 standards.

Since research in this area of B&H has rarely included the content of those hazardous substances that are characteristic to honey and honey products, this work aims to investigate more aspects of present environmental contamination and the influence of pesticides, heavy metals, radioactive elements, and antibiotic residues on honey.

Experimental Procedures

Forty six samples of unifloral and multifloral honeys (meadow 14, chestnut 18, acacia 4, and amphorae 2, plus blends of honeydew honey with blossom 8) from 18 locations in northwestern B&H were analyzed in this research during august 2008 and were not botanically typified. The field of sampling is presented in Table 1 and Fig. 1. Sampling was conducted directly from the hive sites or in producer's filling processing plants using the standard sampling method described in the Official Gazette of the Republic of Croatia [27]. Samples were filled into glass containers to the top, closed and stored in a refrigerator at 4ºC until analysis.

In our research the honey samples, which had been taken from honey producers, were typified and labelled by the Veterinary Institute of USC and Veterinary Faculty at the University of Sarajevo on the basis of a chemical-physical analysis and an analysis of pollen.

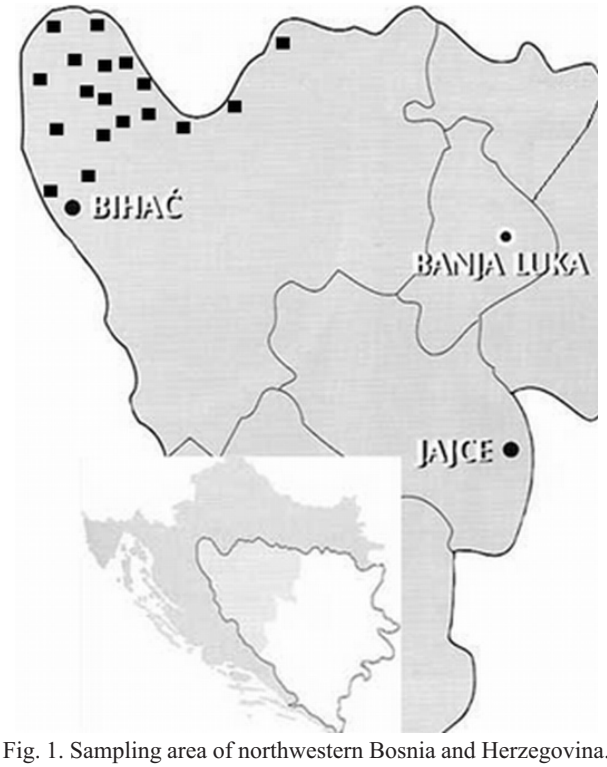

Fig. 1. Sampling area of northwestern Bosnia and Herzegovina.

Table 1. List of honey samples, type brand, label, and location of production.

Type of honey	Sample label *	Location of production	Location number
Meadow honey (MH)	MH-1 MH-1a	Velika Kladuša	1
	MH-2 MH-2a	Velika Kladuša	2
	MH-3 MH-3a	Bihać	3
	MH-4 MH-4a	Velika Kladuša	4
	MH-5 MH-5a	Velika Kladuša	5
	MH-6 MH-6a	Bihać	6
	MH-7 MH-7a	Bužim I	7
Chestnut honey (CH) (*Castanea sativa*)	CH-1 CH-1a	Velika Kladuša	8
	CH-2 CH-2a	Velika Kladuša	9
	CH-3 CH-3a	Cazin	10
	CH-4 CH-4a	Velika Kladuša	11
	CH-5 CH-5a	Bužim II	12
	CH-6 CH-6a	Velika Kladuša	2
	CH-7 CH-7a	Velika Kladuša	13
	CH-8 CH-8a	Bužim I	7
	CH-9 CH-9a	Cazin II	14
Acacia honey (AcH) (*Robinia pseudoacacia*)	AcH-1 AcH-1a	Bosanska Krupa	15
	AcH-2 AcH-2a	Bosanska Dubica	16
Amphorae honey (AH) (*Amphora fructicosa*)	AH-1 AH-1a	Bosanska Dubica	17
Honeydew (HH)	HH-1 HH-1a	Velika Kladuša	13
	HH-2 HH-2a	Velika Kladuša	18
	HH-3 HH-3a	Velika Kladuša	7
	HH-4 HH-4a	Velika Kladuša	11

* Number and "a" letter in the column indicate two honey samples from different producers at the same location.

Method for Determination of Pesticides in Honey Samples

The analysis of pesticides in honey was made using a method recommended by AOAC (1986) [28]. Pesticides are extracted from samples using a honey mixture of acetonitrile and water, after which the extract was purified by letting through the columns with solid sorbent C_{18}. A column is conditional by acetonitril, elution was made by a mixture of acetonitrile and toluene. Separation, identification and quantitative determination of pesticides were made by injection of aliquots of concentrated extract into capillary column gas chromatograph Hewlett Packard HP 6890 Series with a mass detector HP 5973 Mass Selective Detector. The capillary column HP-5MS Agilent 19019S-433, dimensions 30 m × 250 μm × 0.25 μm, and the carrier gas flow of 2.0 mL/min was used. A starting column temperature was 70ºC, then increased to 25ºC/min to 150ºC, followed by 3ºC/min to 200ºC and finally 8ºC/min to 280ºC (10 minutes). One run per sample lasted 41.07 minutes. Obtained chromatogram analysis was performed using the DRS software, which consists of three parts. This is the G1716 Agilent Deconvolution Reporting software, which for analysis of target compounds used data from the Agilent GS/MSD ChemStation (I), with the help of NIST Automated Mass Spectral Deconvolution and Identification Software (AMDIS) (II), and NIST Mass Spectral Search Program (III). The final report (MSD Deconvolution Report) is a combination of these three techniques.

Method for Determination of Heavy Metals in Samples of Honey

Analysis of lead (Pb), zinc (Zn), cadmium (Cd), copper (Cu), and iron (Fe) was conducted on a SpectrAA 200 Varian flame atomic absorption spectrophotometer. Samples were prepared so that 10 g of honey was burned until turned into ashes, and the rest was calcined 13 h in a furnace at 450ºC. Residual ash was dissolved in 10 mL 0.5 M HNO_3, and filtered through quantitative filter paper, according to the US EPA (1983) [29]. Preparation of honey samples for the determination of arsenic was made by wet digestion according to the same reference. During the analysis of all samples to create calibration standards, the basic standards of concentration 1000 ppm – Merck were used.

Method for Determination of Radioactivity in Honey Samples

To determine the activities of ^{137}Cs we used line gamma radiation energy 661.6 key. Time measurement was from 3,600 according to IAEA (1989) [30]. Measurements were performed on a semiconducting gamma-spectroscope with HPGe detector (8192 channels) with a computer. For processing data Gamma Vision 32 software was used.

Method for Determining Residues of Antibiotics and Sulfonamides in Honey

Samples were tested using the microbiological method, the so-called "Four plates" for determination of antibiotic and sulfonamide residues based on an inhibition zone. Samples of honey are inactivated warmed to 80ºC during 10 minutes and tested for Nutritive substrates that are inoculated following test microorganisms:

I base pH 6 – *Bacillus subtilis* ATCC 6633
II base pH 8 – *Bacillus subtilis* ATCC 6633

Table 2. Concentration (mg/kg) of some metals in honey samples from northwestern B&H.

Honey type	Pb	Cd	As	Cu	Fe	Zn
MH	<0.1	<0.02	<0.025	0.14±0.14 [a]	0.91±0.49 [a]	1.29±0.79 [a]
CH	<0.1	<0.02	<0.025	0.17±0.03 [b]	0.46±0.18 [b]	1.38±2.64 [a]
AcH	<0.1	<0.02	<0.025	0.09±0.03 [c]	0.29±0.09 [b]	1.29±1.36 [a]
AH	<0.1	<0.02	<0.025	0.10±0.01 [a, c]	0.59±0.07 [a, b]	0.29±0.05 [b]
HH	<0.1	<0.02	<0.025	0.17±0.04 [b]	0.82±0.32 [a]	0.175±0.09 [b]

Data are expressed as mean value of all analyzed honey samples ± SD (standard deviation).
The same letter in the same column indicates no significant differences (Duncan's test, $p<0.05$).

III base pH 8 – *Sarcina/utea* (*Coccuria varians*) ATCC 9341
IV base PH 7.4 with the addition of trimetoprima – *Sarcina/utea* (*Coccuria varians*) ATCC 9341 [31]

Statistical Analysis

One-way analysis of variance (ANOVA) and multiple comparisons (Duncan's *post-hoc* test) were used to evaluate the significant difference of the data at $p<0.05$. Data were expressed as means ± standard deviation (SD).

Results and Discussion

In all 46 samples of honey, no presence of a series of pesticides was unrecognized by: hexachlorciklohexan, lindane, DDT (DDD+DDE), heptachlor+heptachlorepoxide, endosulfan, aldrin, dieldrin, chlordane, chlorpirifos, dichlorvosa, dimetoat, diazinon, malation, paration, fosalon, atrazine, simazin, and dichlorbenil. Detection limit for all pesticides was 0.001 mg/kg. According to the maximum permitted levels for certain contaminants in food [32], MDK for honey ranged from 1.0-0.01 mg/kg.

During the honey analysis, the stress was put on the detection of the metals Pb, Cd, As, Cu, Fe, and Zn. Table 2 presents the concentration (mg/kg) of some metals in honey samples from northwestern B&H. The results are presented as mean values ± standard deviations of all the analyzed honeys from different locations according to type of honey. The obtained results support the hypothesis that very little metal pollution enters honey. B&H standards permit <0.5 mg/kg Pb, <0.03 mg/kg Cd, <0.5 mg/kg As, <1 mg/kg Cu, <20 mg/kg Fe, and <10 mg/kg Zn in samples [32]. The results show that lead concentrations <0.1 mg/kg were detected in all honey samples, while the concentration of cadmium was <0.02 mg/kg and arsenic <0.025 mg/kg of honey. When it comes to copper determined, the average concentration in all samples is ranked from 0.09 mg/kg (AcH) to 0.18 mg/kg (HH). Iron is found in the concentration of 0.29 mg/kg in acacia honey and 0.92 mg/kg in strained honeys. The lowest proportion of zinc was found in mixed honey samples (0.18 mg/kg), while the highest proportion (1.38 mg/kg) was found in samples of chestnut honey. Samples of chestnut honey (number 8), taken from the municipality area of Bužim (location 7), has a high zinc proportion (8.40 mg/kg), but a sample of chestnut honey (number CH-6a) from a micro location in the Pećigrad area of Velika Kladuša (location 2) has an even larger zinc share of 2.32 mg/kg. And a strained honey sample (number MH-4a) taken from a micro location in Jahovice, in the area of Velika Kladuša (location 4), which is in the vicinity of locations 2 and 7, has 2.79 mg of zinc per kilogram of honey. Zinc is an essential element that occurs in the human body in larger quantities. Its concentration in humans is about 1.8 g. According to the RDA [33], allowed daily allowance for men is 15 mg, and for women 12 mg. Consumption of 100 g honey from Bužim locations (the highest value found in the survey) with its portion of zinc, man enters 0.84 mg of zinc, which meets 5.59% of the allowed limit reached by the daily input of zinc, which cannot be called a critical value. These levels of metals may be appropriate for honey. The represented results (Table 2) show that there were statistically significant differences (ANOVA, Duncan's *post-hoc* test $p<0.05$) in the concentration of Cu, Fe and Zn between different types of honey.

Considering the level of radioactive ^{137}Cs, determined values from nine different locations are shown in Table 3. Results of activity measurement (Bq/kg dry substance) show that the highest levels of active ^{137}Cs was determined in a chestnut honey sample (sample CH-5) taken from the Bužim II site (3.8±0.4 Bq/kg), while the next highest value (2.1±0.4 Bq/kg) was measured at the Bužim I location geographically close by (sample CH-8). In both cases, the samples were taken from chestnut honey. The represented results (Table 3) show that there were statistically significant differences (ANOVA, Duncan's *post-hoc* test $p<0.05$) according to the level of radioactive activity ^{137}Cs (Bq/kg) in different types of honey samples.

Plants enter radionuclides indirectly through roots from soil and through direct deposition of radionuclides in the leaf or in the flower [34]. No similar research has ever been conducted in B&H, but the researchers from Croatia [35] explored the level of ^{137}Cs in the pine needles, and these areas are well known for their honey made from pine needles. The work defined the range of activities of ^{137}Cs in pine cortex from 6:30±0.60 to 29.2±1.80 Bq/kg dry matter, while in pine needles the measured values were present in a range from 6.90±0:50 to 22.6±0.70 Bq/kg of dry matter.

Table 3. Level of radioactive activity ^{137}Cs (Bq/kg) in honey samples from northwestern B&H.

Location	MH	CH	AcH	AH	HH
1	0.65±0.20 [a]	0.76±0.10 [a]	0.82±0.12 [a]	0.21±0.02	1.3±0.20 [a]
2	0.53±0.10 [a]	0.65±0.10 [a]	0.16±0.02 [b]	-	0.44±0.04 [b]
3	0.47±0.10 [a]	1.1±0.20 [b]	-	-	0.28±0.03 [c]
4	0.97±0.20 [b]	0.53±0.06 [a]	-	-	0.52±0.05 [b]
5	0.28±0.10 [c]	3.8±0.40 [c]	-	-	-
6	0.23±0.10 [c]	<0.07 [d]	-	-	-
7	0.40±0.10 [a]	0.80±0.12 [a]	-	-	-
8	-	2.1±0.20 [e]	-	-	-
9	-	0.77±0.08 [a]	-	-	-

Data are expressed as mean value of replication ± SD (standard deviation).
The same letter in the same column indicates no significant differences (Duncan's test, $p<0.05$).

These values are higher than those measured in honey from the northwestern area of B&H, but the values would be more accurate if values from pine needle honey would be measured. Roša [35] also concluded in this work that the presence of ^{137}Cs in soils in Croatia was a consequence of nuclear testing around the world, and radioactive precipitation after the Chernobyl accident, which also is a possible source of measured radionuclides in this area [35]. However, the activities during the last war in B&H are also taken in consideration. In the sampling area were many battles and various war activites, however these claims should be supported by extensive research of the area.

When antibiotic residues were measured in all 46 samples, no concentrations of antibiotics and sulfonamides were found.

Conclusions

The presence of a series of organochlorine pesticides in 46 different honey samples collected from northwestern Bosnia was not detected. The same conclusion can be given when talking about the presence of Pb, for which concentrations in honey were less than <0.1 mg/kg, Cd <0.02 mg/kg, and As <0.025 mg/kg. There were statistically significant differences (ANOVA, Duncan's *post-hoc* test $p<0.05$) in concentrations of Cu, (0.09-0.18 mg/kg), Fe (0.29-0.92 mg/kg), and Zn (0.18-1.38 mg/kg), and in the level of radioactivity of ^{137}Cs (<0.07-3.8 Bq/kg) between different types of honey. Taking into consideration the level of radioactivity of ^{137}Cs, higher concentrations were detected in the Bužim II (3.8±0.4 Bq/kg) and Bužim I (2.1±0.4 Bq/kg) areas, while all other measured values were significantly lower. No residues of antibiotics and sulfonamides were identified in all the analyzed samples. The results of analysis indicate that this area of B&H is not polluted and is suitable for further development of beekeeping.

Abbreviations

USC – Una Sana Canton
B&H – Bosnia and Herzegovina
IUCN – International Union for Conservation of Nature
MRL – Maximum Residue Levels
EU – European Union
BAS – Institute for Standardization of Bosnia and Herzegovina
ISO – International Organization for Standardization
IEC – International Electrotechnical Commission
MH – Meadow honey
CH – Chestnut honey
AcH – Acacia honey
AH – Amphorae honey
AOAC – Association of Official Analytical Chemists
US EPA – United States Environmental Protection Agency
IAEA – International Atomic Energy Agency
ATCC – American-Type Culture Collection
RDA – Recommended Dietary Allowances

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