

## CONCENTRATIONS OF HEAVY METALS IN MULTIFLORAL HONEY FROM THE DIFFERENT TERRESTRIAL ECOSYSTEMS OF THE CARPATHIANS

Oleh Klym ORCID , Olha Stadnytska ORCID 

Institute of Agriculture of the Carpathian Region, 5 Hrushevskoho street, Obroshyno, Pustomyty district, Lviv region, 81115, Ukraine

### ABSTRACT

The aim of this study was to analyse the intensity of heavy metal accumulation in the multifloral honey collected from the beehives located in the mountain, foothill and forest steppe areas of the Carpathian region. The sources of emissions of heavy metals and their release into the environment differ, but they mostly have a technological origin as a consequence of urbanization and industrialization. It was found that the content of iron, zinc, copper, chromium, nickel and lead in the natural multifloral honey collected from the beehives located in the mountain, foothill and forest steppe areas of the Carpathian region did not exceed the standards. However, with the increasing intensity of an industrial impact on the environment, natural multifloral honey contains more heavy metals.

**Key words:** heavy metals, multifloral honey, bees, Carpathian region

### INTRODUCTION

Honey is the most popular apiarian product with a rich chemical composition and medical properties. Honey, propolis and pollen may affect the capability of eliminating heavy metals from an organism [Kędzia and Holderna-Kędzia 2009]. Honey bees (*Apis mellifera* L.) directly depend on the environment that provides them with feed. The area worked by bees from one bee family is usually 7.0 to 30 km<sup>2</sup> [Yazgan et al. 2006]. Therefore, bee products originate from a large area and may constitute a very good indicator of environmental pollution.

The development of industry, agriculture, energetics, transport and intensive mining has led to the situation in which air, water, soil and plants contain toxic mineral elements [Zhulai 1989, Celechovska and Vorlova 2001]. Migration of heavy metals in the environment resulted in their accumulation in soils, plants, tissues of honey bees and bee products [Lavrenov 2004, Horn 2007]. As a result, the character (altered flowering time) and allocation of vegetation (replacement of some honey plant species) changed, which led to the deterioration in the honey quality [Celechovska and Vorlova 2001, Lavrenov

2004, Kuzmina 2008]. The content of mineral components depends on honey type, collection period and the area of its origin [Kanoniuk et al. 2004].

Heavy metals in the pollen basket, honeycombs and multifloral honey characterize the intensity of the urbanization and industrialization of the environment [Zhulai 1989, Horn 2007]. Heavy metal content in honey may serve as an indicator of environmental condition. Numerous studies have shown the relationship between the honey mineral composition and the level of environmental pollution [Nasiruddin Khan et al. 2006, Roman and Popiela 2011]. The presence of metals in honey may pose a threat for human health. In recent years, the content of heavy metals in honey has been determined in countries such as China [Ru et al. 2013], Iran [Mahmoudi et al. 2015, Sobhanardakani and Kianpour 2016], Spain [Frias et al. 2008], Romania [Ciobanu and Rădulescu 2016] and Poland [Formicki et al. 2013].

In addition, heavy metals in the above-mentioned products characterize different terrestrial ecosystems. Carpathian region includes areas such as: mountain, foothill and forest steppe. Taking into account the above considerations, there is scientific and practical interest in

**Table 1.** Heavy metals in the multifloral honey, mg/kg of natural weight ( $M \pm m$ ,  $n = 3$ )

**Tabela 1.** Metale ciężkie w miodzie poliflornym, mg/kg masy naturalnej ( $M \pm m$ ,  $n = 3$ )

| Element – Pierwiastek    | Terrestrial ecosystems of the Carpathians – Ekosystemy lądowe regionu karpackiego |                               |                               |
|--------------------------|---|-------------------------------|-------------------------------|
|                          | Mountain – Góry   | Foothill – Pogórze            | Forest steppe – Step leśny    |
| Iron, Fe – Żelazo, Fe    | 2.40 ± 0.112  | 3.96 ± 0.099***               | 5.66 ± 0.202***               |
| Zinc, Zn – Cynk, Zn      | 2.40 ± 0.107  | 4.19 ± 0.109***               | 6.83 ± 0.254***               |
| Copper, Cu – Miedź, Cu   | 0.19 ± 0.011  | 0.41 ± 0.017***               | 0.76 ± 0.031***               |
| Chromium, Cr – Chrom, Cr | 0.21 ± 0.011  | 0.42 ± 0.020***               | 0.80 ± 0.031***               |
| Nickel, Ni – Nikiel, Ni  | 0.18 ± 0.011  | 0.41 ± 0.017***               | 0.60 ± 0.020***               |
| Lead, Pb – Ołów, Pb      | 0.03 ± 0.003  | 0.08 ± 0.003***               | 0.12 ± 0.005***               |
| Cadmium, Cd – Kadm, Cd   | Trace levels – Ilości śladowe   | Trace levels – Ilości śladowe | Trace levels – Ilości śladowe |

\* $P \leq 0.05$ –0.02, \*\* $P \leq 0.01$ , \*\*\* $P \leq 0.001$ .

the study of heavy metals in the multifloral honey collected from the beehives located in the mountain, foothill and forest steppe areas of the Carpathian region.

The aim of this study was to analyse the intensity of heavy metal accumulation in the multifloral honey collected from the beehives located in the mountain, foothill and forest steppe areas of the Carpathian region.

## MATERIAL AND METHODS

The multifloral honey was collected from three beehives in three apiaries located in the mountain (Slavska village in the Skole district), foothill (Stynava village in the Stryi district) and forest steppe (Myklashiv village in the Pustomyty district) areas of the Carpathian region. The honey was homogenized by its thorough mixing. The samples were pooled. Heavy metals (iron, zinc, copper, chromium, nickel, lead and cadmium) in the studied multifloral honey were determined by atomic absorption spectrophotometer (S-115 PK).

The obtained data were analysed statistically using Student's *t*-test. Moreover, an arithmetic mean ( $M$ ) and the standard error of the mean ( $\pm m$ ) were calculated. The differences in the means were considered statistically significant at  $P \leq 0.05$ . The Origin 6.0 (OriginLab Corporation, Northampton, MA, USA) and Excel (Microsoft Inc., Redmond, WA, USA) programs were used for statistical analysis.

## RESULTS AND DISCUSSION

Honey may contain different amounts of toxic metals, which depend on its origin and species. The contents of selected elements in the honey are presented in Table 1. In the present study, quite a clear regionalization of elements occurrence was found in the analyzed honey samples.

Toxic elements such as lead, cadmium and arsenic are controlled in natural flower honey according to the state standards of Ukraine (SSTU 4497:2005) [Lavrenko 2004, Horn 2007]. Natural flower honey is considered to be good if it does not contain more than 1.0

and 0.05 mg · kg<sup>-1</sup> of lead and cadmium, respectively. Good flower honey should not contain even the smallest amounts of arsenic [Zhulai 1989]. The honey lead (Pb) content determined in the present study ranged from 0.03 to 0.12 mg · kg<sup>-1</sup> (Table 1). Spodniewska and Romaniuk [2007] also reported a low lead content in honey from the Warmia and Mazury Province (in the honey samples from the municipalities without intensive soil and plant cultivation or heavy traffic routes).

It can be stated that the content of iron, zinc, copper, chromium, nickel and lead in the natural multifloral honey collected from the beehives located in the foothill and forest steppe areas of the Carpathian region does not exceed the standards. However, natural multifloral honey taken from the beehives located in the forest steppe and foothill areas of the Carpathian region contained more iron, zinc, copper, chromium, nickel and lead in comparison with the multifloral honey taken from the beehives located in the mountain area (Table 1).

Copper and zinc are microelements essential for the normal development of an organism and their low content in honey is undesired. These elements may also reflect the level of environmental pollution [Wieczorek et al. 2006]. In the present study, the limits were not exceeded (Table 1). A much higher concentration of these elements in honey was found by Roman and Popiela [2011] in Poland (Cu 1.03–7.72 mg · kg<sup>-1</sup>, Zn 1.13–13.92 mg · kg<sup>-1</sup>). Kacaniová et al. [2009] reported the content of zinc ranging from 0.25 to 3.82 mg · kg<sup>-1</sup> and that of copper from 0.12 to 1.37 mg · kg<sup>-1</sup> in the honey from Slovakia. In the multifloral honey from Italy [Perna et al. 2012], the zinc content was higher (9.39 mg · kg<sup>-1</sup>) in comparison with the results obtained in the present study, whereas the multifloral honey from Turkey [Kucuk et al. 2007] contained much lower amounts of copper (0.09 mg · kg<sup>-1</sup>) and zinc (0.54 mg · kg<sup>-1</sup>).

In the studied honey samples, the trace amounts of cadmium were also determined, whereas Tuzen et al. [2007] reported the cadmium levels in Turkish honey ranging between 0.0009 and 0.0179 mg · kg<sup>-1</sup>. Also, the low cadmium content in the honey from the Warmia

and Mazury Province was found by [Spodniewska and Romaniuk \[2007\]](#), who showed that the content of this element in the analyzed samples was much lower than that of lead.

As can be seen from [Table 1](#), the natural multifloral honey taken from the beehives located in the forest steppe of the Carpathian region, contained the greatest amounts of the above-mentioned heavy metals. It can point towards the level of the urbanization and industrialization of this terrestrial ecosystem and the influence of heavy metals on bee and human organisms.

With the increasing intensity of an industrial impact on the environment, natural multifloral honey contains more iron, zinc, copper, chromium, nickel and lead. Some traces of toxic cadmium are found in the studied natural multifloral honey. [Jabłoński et al. \[1995\]](#) showed that the multifloral honey is a product containing much lower amounts of toxic metals, which is associated with the process of purifying the honey raw material during its processing for honey in the bee's honey sac. More than 20% of heavy metals are captured from the raw material by the bee's gastrointestinal system. Therefore, the smaller amounts of these elements are found in the honey compared with the raw material.

## CONCLUSIONS

The content of iron, zinc, copper, chromium, nickel and lead in the natural multifloral honey, taken from the beehives located in the mountain, foothill and forest steppe areas of the Carpathian region does not exceed the standards. However, with the increasing intensity of an industrial impact on the environment, natural multifloral honey contains more heavy metals. The low content of heavy metals in the Ukrainian honey is not dangerous for consumers.

## ACKNOWLEDGEMENT

The study was funded as part of the research units' statutory activity.

## REFERENCES

- Celechovska, O., Vorlova, L. (2001). Groups of honey – physicochemical properties and heavy metals. *Acta Vet. Brno*, 70(1), 91–95. DOI: [10.2754/avb200170010091](https://doi.org/10.2754/avb200170010091).
- Ciobanu, O., Rădulescu, H. (2016). Monitoring of heavy metals residues in honey. *Research Journal of Agricultural Science*, 48(3), 9–13.
- Formicki, G., Greń, A., Stawarz, R., Zyśk, B., Gał, A. (2013). Metal Content in Honey, Propolis, Wax, and Bee Pollen and Implications for Metal Pollution Monitoring. *Pol. J. Environ. Stud.*, 22(1), 99–106.
- Frias, I., Rubio, C., Gonzalez-Iglesias, T., Gutierrez, A.J., Gonzalez-Weller, D., Hardisson, A. (2008). Metals in fresh honeys from Tenerife Island, Spain. *Bull Environ. Contam Toxicol.*, 80(1), 30–33. DOI: [10.1007/s00128-007-9301-9](https://doi.org/10.1007/s00128-007-9301-9).
- Horn, H. (2007). All about honey. M: Astrel 141-152, [in Russian].
- Jabłoński, B., Kołtowski, Z., Marcinkowski, J., Rybak-Chmielewska, H., Szczesna, T. (1995). Contamination of nectar, honey and pollen collected from roadside plants [Zawartość metali ciężkich (Pb, Cd i Cu) w nektarze, miodzie i pyłku pochodzącym z roślin rosnących przy szlakach komunikacyjnych]. *Pszczel. Zesz. Nauk.*, 39(2), 129–144 [in Polish].
- Kacaniová, M., Knazovická, V., Melich, M., Fikselová, M., Massanyi, P., Stawarz, R., Hascik, P., Pechociak, T., Kuczowska, A., Putala, A. (2009). Environmental concentration of selected elements and relation to physicochemical parameters in honey. *J. Environ. Sci. Health Part A* 44, 414–422. DOI: [10.1080/10934520802659802](https://doi.org/10.1080/10934520802659802).
- Kanoniuk, D., Podgórski, G., Unkiewicz-Winiarczyk, A. (2004). Content of Ca, Mg, Fe and Cd in nectarous and honeydew honeys from non-urbanized and urbanized areas [Zawartość Ca, Mg, Fe i Cd w miodach nektarowych i spadziowych z terenów niezurbanizowanych i zurbanizowanych]. *Roczn. PZH*, 55 (Suppl), 77–80 [in Polish].
- Kędzia, B., Hołderna-Kędzia, E. (2009). Elimination of toxic metals from the human body with use of bee-products [Usuwanie metali szkodliwych dla zdrowia z organizmu za pomocą produktów pszczelich]. *Herba Polonica*, 55(1), 98–108 [in Polish].
- Kucuk, M., Kolayli, S., Karaoglu, S., Ulusoy, E., Baltaci, C., Candan, F. (2007). Biological activities and chemical composition of three honeys of different types from Anatolia. *Food Chem.*, 100, 526–534. DOI: [10.1016/j.foodchem.2005.10.010](https://doi.org/10.1016/j.foodchem.2005.10.010).
- Kuzmina, K. (2008). Only natural honey has antibacterial effect. *Apiary*, 12, 26–27 [in Ukrainian].
- Laurenov, V.K. (2004). All about honey and other bee products. M: AST Donetsk: Stalker, 256 [in Russian].
- Mahmoudi, R., Mardani, K., Rahimi, B. (2015). Analysis of heavy metals in honey from north-western regions of Iran. *J. Chemical Health Risks*, 5, 251–256.
- Nasiruddin Khan, M., Qaiser, M., Mubashir Raza, S., Rehman, M. (2006). Physicochemical properties and pollen spectrum of imported and local samples of blossom honey from the Pakistani market. *Int. J. Food Sci. Technol.*, 41, 775–781. DOI: [10.1111/j.1365-2621.2005.01079.x](https://doi.org/10.1111/j.1365-2621.2005.01079.x).
- Perna, A., Simonetti, A., Intaglietta, I., Sofo, A., Gambacorta, E. (2012). Metal content of southern Italy honey of different botanical origins and its correlation with polyphenol content and antioxidant activity. *Int. J. Food Sci. Technol.*, 47, 1909–1917. DOI: [10.1111/j.1365-2621.2012.03050.x](https://doi.org/10.1111/j.1365-2621.2012.03050.x).
- Roman, A., Popiela, E. (2011). Studies of chosen toxic elements concentration in multiflower bee honey. *Potravinárstvo*, 5, 67–69 DOI: [10.5219/134](https://doi.org/10.5219/134).
- Ru, Q.M., Feng, Q., He, J.Z. (2013). Risk assessment of heavy metals in honey consumed in Zhejiang province, southeastern China. *Food Chem. Toxicol.*, 53, 256–262. DOI: [10.1016/j.fct.2012.12.015](https://doi.org/10.1016/j.fct.2012.12.015).
- Sobhanardakani, S., Kianpour, M. (2016). Heavy Metal Levels and Potential Health Risk Assessment in Honey Consumed

- in the West of Iran. *Avicenna J. Environ. Health Eng.*, 3(2):e7795. DOI: [10.5812/ajehe.7795](https://doi.org/10.5812/ajehe.7795).
- Spodniewska, A., Romaniuk, K. (2007). Concentration of lead and cadmium in honey in randomly chosen apiaries of the Warmia and Mazury province [Zawartość ołowiu i kadmu w miodzie w wybranych pasiekach województwa warmińsko-mazurskiego]. *Medycyna Wet.* 2007, 63(5), 602–603 [in Polish].
- Tuzen, M., Silici, S., Mendil, D., Soylak, M. (2007). Trace element levels in honeys from different regions of Turkey. *Food Chemistry*, 103, 325–330. DOI: [10.1016/j.foodchem.2006.07.053](https://doi.org/10.1016/j.foodchem.2006.07.053).
- Wieczorek, J., Wieczorek, Z., Mozolewski, W. (2006). Can bee honey serve as an environmental marker? *Polish J. Environ. Stud.*, 15, 203–207.
- Yazgan, S., Horn, H., Isengard, H.-D. (2006). Honey as bio indicator by screening the heavy metal content of the environment – *Deutsche Lebensmittel-Rundschau. Zeitschrift für Lebensmittelkunde und Lebensmittelrecht*, 102(5), 192–194.
- Zhulai, V.Y. (1989). Minerals in different kinds of honey. *Apiary*, 9, 316 [in Ukrainian].

## KONCENTRACJA METALI CIĘŻKI W MIODZIE WIELOKWIATOWYM W RÓŻNYCH EKOSYSTEMACH KARPAT

### STRESZCZENIE

Celem badań było zbadanie intensywności akumulacji metali ciężkich w miodzie wielokwiatowym pobranym z uli zlokalizowanych na obszarach górskich, podgórskich i leśnych w rejonie karpackim. Źródła emisji metali ciężkich i ich uwalnianie do środowiska są różne, ale w większości mają one technologiczne pochodzenie związane z urbanizacją i industrializacją. Stwierdzono, że zawartość żelaza, cynku, miedzi, chromu, niklu i ołowiu w naturalnym miodzie wielokwiatowym, pobranym z uli zlokalizowanych na obszarach górskich, podgórskich i leśnych w regionie karpackim nie przekracza standardów. Jednak wraz ze wzrostem intensywności przemysłowego oddziaływania na środowisko naturalny miód wielokwiatowy zawiera więcej metali ciężkich.

**Słowa kluczowe:** metale ciężkie, miód wielokwiatowy, pszczoły, region karpacki