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CONTAMINATION OF PROPOLIS USED AS A DIETARY SUPPLEMENT

Adam Roman, Ewa Popiela-Pleban

ABSTRACT

The aim of the study was to determine the extent of chosen toxic elements (zinc, copper, lead, arsenic and cadmium) bioaccumulation in propolis collected in Opole area. The present study demonstrates that propolis can be used as a dietary supplement. The research material were samples of propolis originated from 3 bee colonies in 30 apiaries (n=3x30=90). Quantitative analysis of studied elements were conducted using Varian ICP-AES plasma spectrometer with mass detection controlled, and CETAC-5000 AT ultrasonic nebulizer. The presence of toxic elements was determined in an examined biological materials. The sequence of accumulation level of studied elements in propolis was as follows: Zn>>Cu>Pb>As>Cd. An average concentration of zinc, copper, lead, arsenic and cadmium amounted to 56.28, 7.12, 6.91, 0.745, 0.218 $mg.kg^{-1}$, respectively. Only the copper average content in propolis was within acceptable standards, whereas the mean contents of other elements greatly exceed these standards. All portions of propolis should be subjected to toxicological testing before applying these samples for internal use.

Keywords: propolis, heavy metals, arsenic, cadmium, copper, lead, zinc, accumulation, dietary supplement

INTRODUCTION

The industry and motorization development as well as intensive agriculture based on the chemicalization contributed to a massive increase of environment pollution. Among many pollutants there are elements of toxic properties, which natural content in soil and the atmosphere is negligible **(Bayçu and Önal 1993, Celli and Maccagnani 2003, Chłopecka et al. 1996, Jones 1987, Kabata-Pendias and Pendias 1999, Stankovska et al. 2008)**. They are widely used in the human economy, therefore they are common environmental pollutants. Even at low concentrations they may cause a lot of diseases and abnormalities in the functioning of the human organism.

Honey bee (*Apis mellifera* L.) is inextricably linked with the external environment, from which it derives not only air and water, but food as well. The products gathered by the bee colony are kind of pooled samples derived from such a large area. The pollutants occurring in a given area can also be accumulated in the raw material collected by bees, and in bees itself **(Bogdanov 2006, Iannotti et al. 2000, Roman 2009, Roman 2010)**.

During the processing of raw materials to propolis by worker bees its purification does not take place. Propolis is an extremely complicated material which has many biologically active compounds valuable to the human organism. Most of these substances belong to the fallowing groups flavonoids, diterpene, phenolic acids, aromatic esters, alcohol-phenols, keto-phenols, coumarins, lipid and wax, bio-elements, vitamins, proteins and other compounds **(Bankova et al. 2000, de Casrtro 2001, Marcucci 1995)**. Propolis it contains many minerals and trace elements, which concentration reflects the given region abundance with these chemical elements **(Conti and Botrè 2001, Roman 1997)**. There is a close correlation between the level of heavy metals accumulation in soil and plants and their content in bee products **(Kabata-Pendias and Pendias 1999, Roman 1997, Roša 2006, Roman 2009 and 2010)**. Therefore, only selected portions of propolis collected from bee are suitable for internal use by humans.

The aim of the study was to determine the accumulation degree of chosen toxic elements (arsenic, cadmium, copper, lead and zinc) in propolis collected from bee colonies in Opole area. Can propolis be used as an additive to food supplements?

MATERIAL A METHODOLOGY

The research material for the investigation were samples of propolis originated from stationary apiaries situated in Opole area. Material was collected from May-June to August 2010. Propolis samples originated from 3 bee colonies in 30 apiaries (n=3x30=90). The individual samples were combined into one pooled sample weighting about 30 g, representative for the particular apiary. Propolis was collected directly from the hives, by scraping down with a sharp instrument from their wooden elements (walls and frames) to a clean plastic containers. The received samples were homogenized by freezing, fragmentation and mixing. The 2000 mg of material from each sample was weighted (with precision of 0.1 mg) and diluted with 20 ml of concentrated, spectrally pure, nitric acid solution produced by Merck company. Next, samples were mineralized using the microwave technique at an elevated pressure in the chip-type MD-2000 station manufactured by CEM-USA. Quantitative analysis of studied elements were conducted using Varian ICP-AES plasma spectrometer with mass detection controlled by P-3202 computer cooperating with Philips Scientific analytical combine (PU-7000 model), and CETAC-5000 AT ultrasonic nebulizer. Quantitative analysis were conducted in Analytical Laboratory of Wroclaw University of Environmental and Life Science (Poland). The one-way analysis of variance (ANOVA) was performed to evaluate differences among groups. Mean concentrations of elements, standard deviations and correlations between elements were calculated. Significance level was taken as $p \leq 0.05$ and $p \leq 0.01$.

RESULTS AND DISCUSSION

Toxicological status of propolis fully reflects the harmful compounds and toxic elements contamination of raw materials and the environment. The concentration of some toxic elements in the propolis samples is high variability. Numerous authors draw an attention to the fact that there is a high correlation between the concentration level of minerals in bee products and their content in an external environment **(Barišic et al. 2002, Bayçu and Önal 1993, Conti and Botrè 2001, Leita et al. 1996, Rashed et al. 2009, Roša 2006)**. Present study demonstrated that propolis was contaminated with toxic elements. This is confirmed by numerous research of other authors, which showed high concentrations of heavy metals in the propolis **(Conti and Botrè 2001, Cvek et al. 2008, Kulevanova et al. 1995)** also claimed that propolis is much more contaminated with heavy metals than any other bee products. The study results are presented in Table 1. Very high level in this product was observed in the case of zinc, which averaged 55.79 $mg \cdot kg^{-1}$ and in individual samples ranged from 10.91 to 115.22 $mg \cdot kg^{-1}$. The content of this element observed by **Dogan et al. (2006)** in propolis samples from different regions of Turkey was 176-676 $mg \cdot kg^{-1}$. **Cvek et al. (2008)** demonstrated even the value of 9326 $mg \cdot kg^{-1}$. In turn, the maximum copper content was determined as 18.32 $mg \cdot kg^{-1}$, and the average was 8.94 $mg \cdot kg^{-1}$. **Roman (1997)** observed several times higher concentration of that metal in the propolis from the region of Głogów which ranged from 23.51 to 34.15 $mg \cdot kg^{-1}$, while **Dogan et al. (2006)** received higher value in the propolis from different regions of Turkey - an average of i.e. 45-96 mg $Cu \cdot kg^{-1}$. Lead is one of the most burdensome heavy metals in the environment. Its concentration was also high in propolis and reached an average level of 6.54 $mg \cdot kg^{-1}$ (maximum 18.29 $mg \cdot kg^{-1}$). In other studies, **Roman (2000)** showed the mean level of lead concentration amounted to 18.39 mg $Pb \cdot kg^{-1}$ in the propolis from Głogów and from Rudna area from 6.73-17.83 $mg \cdot kg^{-1}$ on average **(Roman 2000)**. Similar results were obtained when examining the mean concentration of this element in propolis from the region of Opole (6.62-13.63 $mg \cdot kg^{-1}$). It can be thus concluded that lead concentration in propolis was high and relatively stable. The present study demonstrated that arsenic content in propolis amounted to 0.698 $mg \cdot kg^{-1}$ on average, and in individual samples ranged from 0.007 to 1.806 $mg \cdot kg^{-1}$. The average concentration of arsenic should be considered as a very high in the propolis. Very similar results were shown by **Roman (1997)** in propolis originating from the Opole and LGOM area, i.e. 0.561 and 0.670 $mg \cdot kg^{-1}$, respectively. This is most likely the result of a significant contamination of soils in the areas of metallurgical and chemical industries and extensive urban areas, where the accumulation of arsenic in the soil reaches up to 2500 $mg \cdot kg^{-1}$ **(Kulevanova et al. 1995)**. Generally, the cadmium concentration in the examined bee product was the smallest and was an average of 0.203 $mg \cdot kg^{-1}$. However, the dispersion of results was very wide from 0.006 to 0.811 $mg \cdot kg^{-1}$. In earlier studies conducted by **Roman (1997)**, lower levels of cadmium, ranged from 0.043 to 0.116 $mg \cdot kg^{-1}$, were observed in propolis originating from the region of the copper industry, but significantly higher in the product coming from the region of the cement industry - an average of 0.513 to 0.795 $mg \cdot kg^{-1}$. Moreover, **Roman (2000)** noted higher cadmium content in the propolis from the Wałbrzych region (0.260 $mg \cdot kg^{-1}$).

The results of the present study and review of other authors results allow to conclude unequivocally that the level of toxic trace elements concentration in propolis depends on the state of environmental pollution in the area of the material sampling.

Only the copper average content in propolis did not exceed the maximum allowable standards (Polish Standard) (PN 1998), which is set at 10 $mg.kg^{-1}$. In case of other elements mean values were significantly exceeded. It demonstrates that bee products show significant contamination level. Therefore, only selected portions of propolis can be used to produce natural medicines and dietary supplements for humans. Each lot of propolis intended for that purpose must be subjected to toxicological analysis.

CONCLUSION

In present study propolis has shown high contamination level by toxic elements. Zinc, lead and arsenic showed the highest level of concentration of studied elements in relation to current standards. The sequence of accumulation level of studied elements in propolis was as follows: Zn>>Cu>Pb>As>Cd.

All portions of propolis should be subjected to toxicological testing before applying these samples for internal use as a dietary supplement.

Table 1 Concentration of chosen elements in bee propolis (N=90)

No. of sample	Chemical elements (in $mg \cdot kg^{-1}$)				
	As	Cd	Cu	Pb	Zn
Minimum	0.007	0.006	1.09	0.39	10.91
Maximum	1.806	0.811	18.32	18.29	115.22
Average	**0.698** ^{A}	**0.203** ^{B}	**8.94** ^{C}	**6.54** ^{D}	**55.79** ^{E}
SD	0.577	0.184	4.89	4.51	33.21
Variation coefficient in %	81.7	90.6	54.7	69.0	59.5
LOQ	$\leq$ 0.001	$\leq$ 0.01	$\leq$ 0.01	$\leq$ 0.100	$\leq$ 0.001

A, B, C, D, E - differences between the elements assessed highly significant on a level of $p \leq 0.01$

REFERENCES

BANKOVA, V. S, CASTRO S. L, MARCUCCI M. C. 2000. Propolis: recent advances in chemistry and plant origin. In *Apidologie*, vol. 31, 2000, p. 3-15

BARIŠIC, D., BROMENSHENK, J. J., KEZIÆ, N., VERTAÈNIK, A. 2002. The role of honey bees in environmental monitoring in Croatia. In *Honey Bees: Estimating the Environmental Impact of Chemicals*, p. 160-185.

BAYÇU, G., ÖNAL, M. 1993. An Investigation of the Levels of Cadmium and Lead in the Soil and in the Leaves of Selected Specimens of Ailanthus altissima Found Growing Beside a Freeway in Istanbul. In *The Journal of Biology*, vol. 56, 1993, p. 21-34.

BOGDANOV, S. 2006. Contaminants of bee products. In *Apidologie*, vol. 37, 2006, p. 1-18.

CELLI, G., MACCAGNANI B. 2003. Honey bees as bioindicators of environmental pollution. In *Bulletin of Insectology*, vol. 56, 2003, p. 137-139.

CHŁOPECKA, A., BACON, J. R., WILSON, M. J., KAY, J. 1996. Forms of cadmium, lead, and zinc in contaminated soils from southwest Poland. In *Journal of Environmental Quality*, vol. 25, 1996, p. 69-79.

CONTI, M. E., BOTRÈ, F. 2001. Honeybees and their products as potential bioindicators of heavy metals contamination. In *Environmental Monitoring and Assessment*, vol. 69, 2001, p. 267-282.

CVEK, J., MEDIĆ-ŠARIĆ, M., WITALI, D., VEDRINA-DRAGOJEVIĆ, I., SMIT, Z., TOMIĆ, S. 2008. The content of essential and toxic elements in Croatian propolis samples and their tinctures. In *Journal of Apicultural Research*, vol. 47, 2008, p. 35-45.

DE CASRTRO, S. L. 2001. Propolis: biological properties and pharmacological activities. Therapeutic uses of this bee-product. In *Annual Review Biomedical Sciences*, vol. 3, 2001, p. 49-83.

DOGAN, M., SILICI, S., SARAYMEN, R., ILHAN, I. O. 2006. Element content of propolis from different regions of Turkey. In *Acta Alimentaria*, vol. 35, 2006, p.127-130.

IANNOTTI, O., MINCIGRUCCI, G., BRICCHI, E., FRENGUELLI, G. 2000. Pollen viability as a bio-indicator of air quality. In *Aerobiologia., vol.* 16, 2000, p. 361-365.

JONES, K. C. 1987. Honey as an indicator of heavy metal contamination. In *Water, Air, and Soil Pollution*, vol. 33, 1987, p. 179-189.

KABATA-PENDIAS, A., PENDIAS, H. 1999. *Biogeochemistry of vestigial elements.* PWN, Warszawa, 74-288.

KULEVANOVA, S., STAFILOV, T., DOREVSKI, K. 1995. Determination of some macroelements in propolis by atomic absorption spectrometry. In *Acta Pharmaceutica*, vol. 45, 1995, p. 45-52.

LEITA, L., MUHLBACHOVA, G., CESCO, S., BARBATTINI, R., MONDINI, C. 1996. Investigation of the use of honey bees and honey bee products to assess heavy metals contamination. In *Environmental Monitoring and Assessment*, vol. 43, 1996, p. 1-9.

MARCUCCI, M. C. 1995. Propolis: Chemical composition, biological properties and therapeutic activity. In *Apidologie*, vol. 26, 1995, p. 83-99.

PN-R-78891:1996 *Propolis - kit pszczeli. Dz. U. Nr 139. poz. 300, 1998.*

RASHED, M. N., EL-HATY, M. T. A., MOHAMED, S. M. 2009. Bee honey as environmental indicator for pollution with heavy metals. In *Toxicological and Environmental Chemistry*, vol. 91, 2009, p. 389-403.

ROMAN, A. 1997. Bees and their products as pollution bioindicator in the copper (LGOM) and lime-cement (Opole) industry areas. In *Scientific Exercise Books of Agricultural University in Wroclaw*, 323, p. 175-193.

ROMAN, A. 2000. The study comparative contents Cd, Pb and Zn in honey, propolis and wax come from regions of Wałbrzych and Głogów. In *Apicultural Scientific Exercise Books*, Suplement No 1, p. 76-77.

ROMAN, A. 2009. Concentration of Chosen Trace Elements of Toxic Properties in Bee Pollen Loads. In *Polish Journal of Environmental Studies*, vol. 18, 2009, no. 2, p. 265-272.

ROMAN, A. 2010. Level of Copper, Selenium, Lead, and Cadmium in Forager Bees. In *Polish Journal of Environmental Studies*, vol.19, 2010, no. 3, p. 663-669.

ROŠA, J. 2006. Tannins and microelements in the cells of silver fir tree needles (*Abies alba* Mill.) and the microelements in bee honey as indicators of the silver fir forests condition in the area of Gorski Kotar. In *Sumarski List*, vol. 130, 2006, p. 493-509.

STANKOVSKA, E., STAFILOV, T., ŠAJN, R. 2008. Monitoring of trace elements in honey from the Republic of Macedonia by atomic absorption spectrometry. In *Environmental Monitoring and Assessment*, vol. 142, 2008, no. 1-3, p. 117-126.

Contact address:

Adam Roman, dr. hab. ing. Wrocław University of Environmental and Life Science, Department of Environment Hygiene and Animal Welfare, Chełmońskiego 38C, 51- 630 Wrocław, Poland, E-mail: adam.roman@up.wroc.pl

Ewa Popiela-Pleban, Wrocław University of Environmental and Life Science, Department of Environment Hygiene and Animal Welfare, Chełmońskiego 38C, 51- 630 Wrocław, Poland, E-mail: ewa.popiela@up.wroc.pl