Journal of Microbiology, Biotechnology and Food Sciences

Lazor et al. 2012 : 1 (4) 522-533

REGULAR ARTICLE

MONITORING OF AIR POLLUTION AND ATMOSPHERIC DEPOSITION OF HEAVY METALS BY ANALYSIS OF HONEY

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ABSTRACT

Honey is an important food for the human nutrition. Changes in the quality of bee honey are also caused by the contamination with micro-polluting agents, toxic to consumers. Heavy metals in honey are of interest not only for quality control, but can be used also as an environmental indicator. In this work honey samples were collected in different places of Slovakia. From the experimental results the average values for concentration heavy metals in honey in site Prievidza 2006 - 2008 were: 0.0599 $mg.kg^{-1}$ As, 0.0948 $mg.kg^{-1}$ Cd, 0.0747 $mg.kg^{-1}$ Cr, 0.0394 $mg.kg^{-1}$ Hg and 0.1252 $mg.kg^{-1}$ Pb; in site Šaľa 2006 - 2008: 0.0862 $mg.kg^{-1}$ As, 0.0942 $mg.kg^{-1}$ Cd, 0.0736 $mg.kg^{-1}$ Cr, 0.0341 $mg.kg^{-1}$ Hg and 0.1626 $mg.kg^{-1}$ Pb. The results suggested that honey could be used to detect contaminating agents in the environment.

Key words: honey, contamination of honey, heavy metals, Slovakian regions

INTRODUCTION

The heavy metals cadmium, lead and mercury are common air pollutants, being emitted mainly as a result of various industrial activities. Although the atmospheric levels are low, they contribute to the deposition and build-up in soils. Heavy metals are persistent in the environment and are subject of bioaccumulation in food-chains. In monitoring system also continual connection to interlocking partial monitoring systems, especially to atmosphere, water, soil, and geological factors (subsoil) i.e. enters into food chain. Health safety of foodstuffs includes special attention as it directly affects the health state of population in Slovak Republic. Various contaminants are natural part of our ecosystem and on minimalization of their occurrence in foodstuffs special treatments are necessary in whole food chain from controlling of soil and inputs to soil through phyto-sanitary and veterinary control and control of feeds to monitoring of other factors of food chain that are closely related to safety of foodstuffs. Long-range trans-boundary air pollution is only one source of exposure to these metals but, because of their persistence and potential for global atmospheric transfer, atmospheric emissions affect even the most remote regions. Emissions of heavy metals (HM) in Slovak republic from industry have exerted declining trend since 1990. Declining trend of emissions of most heavy metals affected the shut-down of some productions, large reconstruction of separating devices and change of used raw materials. However, in 2006 in comparison to previous years, an increase of emissions of Pb, Hg, Cr, As, Ni, Cu and Zn in combustion processes in industry as well as an increase of emissions of Pb, Cd, As, Ni, Cu and Zn in industrial technologies were recorded. Motivations for controlling of HM concentrations are diverse. Some of these concentrations are dangerous to health or to the environment (e.g. mercury, cadmium, lead, chromium), some may cause corrosion (e.g. zinc, lead), some are harmful in other ways (e.g. arsenic may pollute catalysts). Within the European community the eleven elements of highest concern are arsenic, cadmium, cobalt, chromium, copper, mercury, manganese, nickel, lead, tin, and thallium, the emissions of which are regulated in waste incinerators **(Hogan, 2010)**. Some of these elements are actually necessary for humans in minute amounts (cobalt, copper, chromium, manganese, nickel) while others are carcinogenic or toxic, affecting, among others, the central nervous system (manganese, mercury, lead, arsenic), the kidneys or liver (mercury, lead, cadmium, copper) or skin, bones, or teeth (nickel, cadmium, copper, chromium). The heavy metals Pb, Cd and Hg are not part of the family of trace elements which in small quantities are essential for human health e.g. copper or zinc. If absorbed into the human organism in sufficient quantity they will cause damage. Nutrition is the principal path of pollution of the human organism with Pb, Cd and Hg. Depending on the heavy metal the direct transfer via deposition on nutritive plants or indirect paths via the soil pore water or surface waters are of varying significance. Neither animals, plants nor microorganisms require Pb, Cd or Hg for their metabolic processes. Once certain concentrations in the environment have been exceeded the three heavy metals have harmful effects. These range from individual deaths with the concomitant reduction in the number of individuals, stunted growth, visible leaf damage and reproductive disorders to changes in physiological processes and the impairment of microbiological metabolic processes. Most of the data on the effects come from laboratory research and have been partly confirmed by (very much more laborious) field trials. The results of investigations into the effects form the basis of the determination of critical concentrations in soil water in the critical loads concept. The examination of humanotoxic effects in the context of the critical loads concept focuses in respect of each particular heavy metal (Pb, Cd, Hg) on its transfer via soil and/or water into human nutrition or drinking water systems. Other sources of pollution with consequences for human health such as house dust or paints are not taken into consideration. Honey is an important food for the human nutrition. Honey possesses valuable nourishing, healing and prophylactic properties which result from its chemical composition **(Tuzen and Duran, 2002)**. Honey bees are good biological indicators because they indicate the chemical impairment of the environment they live in through two signals: the high mortality and the residues present in their bodies or in beehive products (in the elements and other contaminants like heavy metals and radionuclides) that may be detected by means of suitable laboratory analyses **(Celli, 1994)**. Several ethological and morphological characteristics make the honey bee a reliable ecological detector: it is an easy-to-breed, almost ubiquitous organism, with modest food requirements; its body is covered with hairs, which make it particularly suitable to hold the materials and substances it comes into contact with; it is highly sensitive to most plant protection products, revealing when they are improperly spread through the environment (e.g. during flowering, in the presence of wind, etc.); its very high rate of reproduction and relatively short average lifespan, causes the colony to undergo rapid, continuous regeneration; its great mobility and wide flying range allows a vast area to be monitored; it is highly efficient in ground surveys (numerous inspections per day). Furthermore, almost all environmental sectors (soil, vegetation, water, air) are sampled by honey bees (Fig. 1), providing numerous indicators (through foraging) for each season. Finally, a variety of materials are brought into the hive (nectar, pollen, honeydew, propolis and water) and stored according to verifiable criteria.

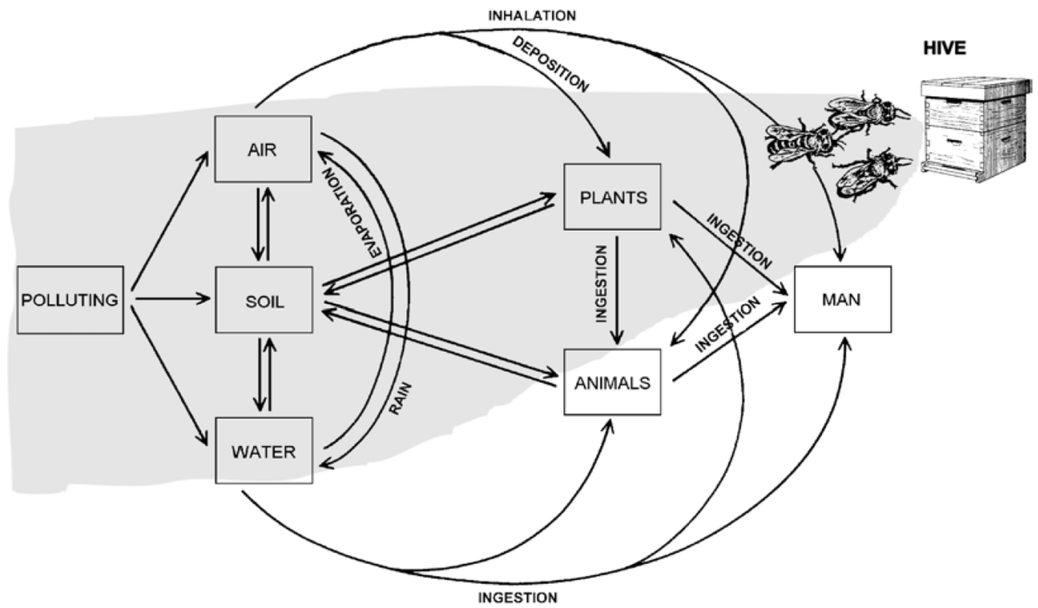

Figure 1 Chart of polluting substance diffusion in the environment. The grey area shows the environmental sectors visited by the honey bees.

Honey, a viscous and aromatic product prepared by bees mainly from nectar or flowers or honey dew **(Dustmann, 1993)**, is an excellent and widely used food that is popular all over the world. In addition, the biological role of honey intake may be important in cases of heavy metal contamination. Honey can characterise the level of soil, plant and air pollution (Jones, 1987) in an area of some square kilometres.

Considered as an analytical sample, honey is one of the most complex mixtures of carbohydrates produced in nature. In common honey, mono- and disaccharides constitute 80–85 % (w/w), water is around 15–20% (w/w) and other organic compounds and inorganic ions being present to a minor extent **(Sanna et al., 2000)**. However, the minor components are often of great importance from many points of view. The evaluation of heavy metals content in honey has a twofold significance: the former one lies in the toxicity of theses metals, with the consequent necessity to develop adequate analytical procedures for their monitoring; the latter one is suggested by the possibility of using bees and their products as bio-indicator of possible environmental pollution, taking advantage of both the large covering area where they live and of the concentration effect of the possibly present environmental pollutants into the "products" of bees **(Rowarth, 1990)**. The mineral content of honey is recognised as an environmental indicator at least since, when Crane published the first data on metals content in honey collected near or far from highways **(Buldini *et al.,* 2001)**. The aim of our work was to carry out an objective monitoring contamination of samples of honey bee with selected heavy metals in sites of Nitriansky and Trenčiansky regions in areas affected by emission - immission activity with using of results to determinate the exact area from the standpoint of its metallic loading in years 2006 - 2008.

MATERIAL AND METHODS

Samples of bee honey were taken in sites in district of Prievidza (Oslany, Bystričany, Čereňany, Nováky and Diviaky nad Nitricou) and district of Šaľa (Horná Kráľová, Močenok, Trnovec nad Váhom, Diakovce and Tešedíkovo) with cooperation of beekeepers in years 2006 and 2008. Processing of samples and analysis of contents of arsenic, cadmium, chromium, mercury and lead by method of atomic absorption spectrophotometry were used for immission loading of individual areas assessment.

Partial honey samples were collected at intervals of 2 weeks during the laying period honey each year. thus the resulting average sample of honey was subjected to analysis. Determination of minerals in honey samples: Five ml of 0.10 N of either Nitric acid or HCl plus 1 ml of hydrogen peroxide was added to a beaker containing the ash of 5g of honey sample, stirred and then heated the mixture on a hotplate to almost complete dryness. Two ml of the HCl (0.1 N) was added to the contents of the beaker, transferred to volumetric flask and diluted with dionized water to 10 ml. Metals were measured by using Atomic Absorption Spectrometry (AAS – VARIAN 240), equipped with a hollow cathode lamp, a 10 cm long slot-burner head and air/acetylene flame.

RESULTS AND DISCUSSION

Results of determination of selected heavy metals contents in individual sites in district Šaľa and Prievidza in years 2006 - 2008 were processed into Tables 1 - 6. In Prievidza locality the average content of As in samples of honey was in 2006 in average 0.0561 $mg.kg^{-1}$, 0.0603 $mg.kg^{-1}$ in 2007 and 0.0634 $mg.kg^{-1}$ in 2008 (tables 1, 3, 7). Arsenic has got into environment in Prievidza locality also by combustion of coal. Coal from Nováky, according to **Baláž and Turčániová (1998)** contains sulphur in total amount 3.00 %, arsenic - 264 000 $mg.t^{-1}$, ash - 28.21 %, moisture - 8.21 %, and iron - 1.44 %. Arsenic could be, or not an essential nutrient for humans and if so, then it is not estimated its daily intake **(Bogdanov *et***

***al.*, 2003)**. Interaction of arsenic with others nutrient factors are mainly not known, with exception of mutual antagonism with selenium **(McLaughlin *et al.*, 1999)**. Determined average concentrations in Šaľa locality in individual sites in samples of honey were in range in 2006 in average 0.1085 $mg.kg^{-1}$, 0.0527 $mg.kg^{-1}$ in 2007 and 0.0974 $mg.kg^{-1}$ in 2008 (table 2, 4, 6). Arsenic is notoriously poisonous to multicellular life because of the interaction of arsenic ions with protein thiols. Arsenic and its compounds, especially the trioxide, are used in the production of pesticides (treated wood products), herbicides, and insecticides. These applications are declining, however, as many of these compounds are being phased out. Arsenic poisoning from naturally occurring arsenic compounds in drinking water remains a problem in many parts of the world.

Average concentrations of Cd assessed in honey in sites of Prievidza district in 2006 were in range of average 0.0850 $mg.kg^{-1}$, 0.0961 $mg.kg^{-1}$ in 2007 and 0.1034 $mg.kg^{-1}$ in 2008 (Table 1, 3, 7). The most significant share on distributing of HM into soil has had phosphorous fertilizers produced on base of Algerian apatites that contain relatively high portion of Cd. **Danihelka (1996)** reported that cadmium content in coal combusted in power station Nováky 1.86 - 2.16 ppm.

In Šaľa locality average contents of Cd in 2006 - 0.0937 $mg.kg^{-1}$, in 2007 - 0.0997 $mg.kg^{-1}$ and 0.0893 $mg.kg^{-1}$ in 2008, respectively were assessed (Table 2, 4, 6). Basic source of cadmium enter into food chain is the application of phosphorous fertilizers, production of iron, steel and combustion of coal. Generally it is accepted that humans are the most sensitive receptors on intake of cadmium from environment, especially through food chain **(Smolders, 2004)**. Chromium is a steely gray and non-oxidating hard metal that is in basic state malleable and lustrous **(Costa and Klein, 2006)**. Usually it occurs in oxidation states II+, III+ and VI+, while III+ is the most stable, and thus in environmental components does not have any significant transfer and does not exert any toxic effects for plants and animals **(Sokolov and Černikov, 1999)**. In nature this element is part of ores, mainly chromite ($FeCr_2O_4$) and krokoite ($PbCrO_4$). Our measured concentrations of chromium were in average 0.0709 $mg.kg^{-1}$ (2007), 0.0766 $mg.kg^{-1}$ (2006) and 0.0734 $mg.kg^{-1}$ (2008) in samples in district Šaľa. In Prievidza locality in 2006 assessed average content of Cr presented 0.0733 $mg.kg^{-1}$, 0.0819 $mg.kg^{-1}$ in 2007 and 0.0689 $mg.kg^{-1}$ in 2008 (table 1, 3, 7). In contrary to Cr(VI+) that is known by its mobility and also its toxicity it occurs in form of solution $HCrO_4^-$, CrO_4^{2-} and $Cr_2O_7^{2-}$ **(Grebenjuk and Charina, 2003)** . Both Cr (VI+) also Cr (III+) can be absorbed directly by skin in relation to physical state, anion form, concentration and pH of solution **(Costa and Klein, 2006)**. Heavy metal pollution can arise from many sources but most commonly arises from the purification of metals, e.g., the smelting of copper and the preparation of nuclear fuels. Electroplating is the primary source of chromium and cadmium. Through precipitation of their compounds or by ion exchange into soils and muds, heavy metal pollutants can localize and lay dormant. Unlike organic pollutants, heavy metals do not decay and thus pose a different kind of challenge for remediation. Currently, plants or microorganisms are tentatively used to remove some heavy metals such as mercury. Plants which exhibit hyper accumulation can be used to remove heavy metals from soils by concentrating them in their bio matter. Some treatment of mining tailings has occurred where the vegetation is then incinerated to recover the heavy metals.

Average contents of Hg in analysed samples in sites of district Prievidza gained in 2006 were 0.0413 $mg.kg^{-1}$, 0.0379 $mg.kg^{-1}$ in 2007 and 0.0390 $mg.kg^{-1}$ in 2008 (Tables 1, 3, 7). Mercury and its compounds are highly toxic for plants and wild animals **(Buldini et al., 2001)**, while mercury is persistent contaminant and in environment it could be transformed on methyl-mercury – the most toxic form that easily gets through placental barrier and hematoencephalic barrier and could damage developing brain **(Aksenova, 2000; Beljaeva, 2000)**. Content of Hg assessed in sites in locality Šaľa was in 2006 in average 0.0199 $mg.kg^{-1}$, 0.0648 $mg.kg^{-1}$ in 2007 and 0.0175 $mg.kg^{-1}$ in 2008 (Tables 2, 4, 6).

Preindustrial deposition rates of mercury from the atmosphere may be about 4 ng/(1 L of ice deposit). Although that can be considered a natural level of exposure, regional or global sources have significant effects. Volcanic eruptions can increase the atmospheric source by 4–6 times. [It could get to foodstuffs from natural, but also human sources. In comparison to low concentrations of mercury in consumable vegetation, mushrooms accumulated higher amount of total mercury and methyl-mercury **(Cappon, Smith, 2003)**.

A serious industrial disaster was the dumping of mercury compounds into Minamata Bay, Japan. It is estimated that over 3,000 people suffered various deformities, severe mercury poisoning symptoms or death from what became known as Minamata disease. The most toxic forms of mercury are its organic compounds, such as dimethyl mercury and methyl mercury. However, inorganic compounds, such as cinnabar are also highly toxic by ingestion or inhalation. Mercury can cause both chronic and acute poisoning.

Lead is a poisonous metal that can damage nervous connections (especially in young children) and cause blood and brain disorders. Lead poisoning typically results from ingestion of food or water contaminated with lead; but may also occur after accidental ingestion of contaminated soil, dust, or lead based paint **(Aksenova, 2000; Beljaeva, 2000)**. Not least interesting source of contamination of environment by compounds of lead is the transportation, especially as consequence of used lead anti-detonation means in engine fuel **(Dobrovol'skij and Nikitin, 1990)**. In most studies lead input to foodstuffs is considered to be app. 70 % of total daily intake of absorbed lead from all sources. Portion of total intake from foodstuffs depends on concentration of lead in atmosphere, water and other sources **(Rojas et al., 1999)**. Content of lead in our samples in average presented value 0.1385 $mg.kg^{1}$ obtained in sites of Prievidza locality in 2006, 0.1535 $mg.kg^{-1}$ in 2007 and 0.0835 $mg.kg^{-1}$ in 2008. In sites of Šaľa locality the average assessed values of Pb in 2006 presented value 0.1674 $mg.kg^{-1}$ in 2007 - 0.1495 $mg.kg^{-1}$ and 0.1709 $mg.kg^{-1}$ in 2008 (Tables 2, 4, 6).

Long-term exposure to lead or its salts (especially soluble salts or the strong oxidant PbO_2) can cause nephropathy, and colic-like abdominal pains. The effects of lead are the same whether it enters the body through breathing or swallowing. Lead can affect almost every organ and system in the body. The main target for lead toxicity is the nervous system, both in adults and children. Long-term exposure of adults can result in decreased performance in some tests that measure functions of the nervous system **(Smolders, 2004)**.

Table 1 Average content of selected heavy metals in honey bee ($mg.kg^{-1}$) in sites in district of Prievidza (2006)

site \ metal	As	Cd	Cr	Hg	Pb
Bystričany	0.0691	0.1102	0.0754	0.0496	0.2135
Čereňany	0.0456	0.0368	0.0492	0.0179	0.0674
Diviaky nad Nitricou	0.0538	0.0528	0.0411	0.0231	0.0495
Nováky	0.0796	0.1189	0.0847	0.0572	0.1893
Oslany	0.0322	0.0211	0.0427	0.0173	0.0341

Tab2 Average content of selected heavy metals in honey bee ($mg.kg^{-1}$) in sites in district of Šaľa (2006)

site \ metal	As	Cd	Cr	Hg	Pb
Diakovce	0.0412	0.0976	0.0631	0.0185	0.1646
Horná Kráľová	0.0295	0.0189	0.0379	0.0121	0.0438
Močenok	0.0322	0.0812	0.0596	0.0175	0.1563
Tešedíkovo	0.0397	0.0892	0.0611	0.0149	0.1473
Trnovec nad Váhom	0.2913	0.0879	0.0618	0.0164	0.1576

Tab 3 Average content of selected heavy metals in honey bee (mg.kg^{-1}) in sites in district of Prievidza (2007)

site \ metal	As	Cd	Cr	Hg	Pb
Bystričany	0.0425	0.1179	0.0799	0.0511	0.2273
Čereňany	0.0472	0.0386	0.0496	0.0089	0.0777
Diviaky nad Nitricou	0.0598	0.0713	0.0817	0.0264	0.0537
Nováky	0.0842	0.1281	0.0764	0.0492	0.2152
Oslany	0.0676	0.0284	0.0399	0.0160	0.0400

Tab 4 Average content of selected heavy metals in honey bee (mg.kg^{-1}) in sites in district of Prievidza (2007)

site \ metal	As	Cd	Cr	Hg	Pb
Diakovce	0.0532	0.0983	0.0514	0.2116	0.1672
Horná Kráľová	0.0311	0.0199	0.0630	0.0108	0.0428
Močenok	0.0376	0.0934	0.0647	0.0099	0.1201
Tešedíkovo	0.0475	0.0897	0.0742	0.0168	0.1536
Trnovec nad Váhom	0.0412	0.0975	0.0530	0.0102	0.1141

Tab 5 Average content of selected heavy metals in honey bee (mg.kg^{-1}) in sites in district of Prievidza (2008)

site \ metal	As	Cd	Cr	Hg	Pb
Bystričany	0.0644	0.1796	0.0716	0.0520	0.1995
Čereňany	0.0329	0.0356	0.0511	0.0123	0.0666
Diviaky nad Nitricou	0.0548	0.0573	0.0389	0.0197	0.0532
Nováky	0.0699	0.1200	0.0790	0.0499	0.1644
Oslany	0.0317	0.0211	0.0350	0.0211	0.0300

Tab 6 Average content of selected heavy metals in honey bee (mg.kg^{-1}) in sites in district of Šaľa (2008)

site \ metal	As	Cd	Cr	Hg	Pb
Diakovce	0.0356	0.1031	0.0711	0.0171	0.1720
Horná Kráľová	0.0377	0.0136	0.0365	0.0100	0.0389
Močenok	0.0289	0.0769	0.0499	0.0097	0.1692
Tešedíkovo	0.0401	0.0799	0.0580	0.0137	0.1392
Trnovec nad Váhom	0.2474	0.0836	0.0786	0.0195	0.1643

CONCLUSION

One of the largest problems associated with the persistence of heavy metals is the potential for bioaccumulation and biomagnification causing heavier exposure for some organisms than is present in the environment alone. Coastal fish (such as the smooth toadfish) and seabirds (such as the Atlantic Puffin) are often monitored for the presence of such contaminants **(Aksenova, 2000; Beljaeva, 2000)**. Toxicity is a function of solubility. Insoluble compounds as well as the metallic forms often exhibit negligible toxicity. The toxicity of any metal depends on its ligands. In some cases, organometallic forms, such as dimethyl mercury and tetraethyl lead, can be extremely toxic. In other cases, organometallic derivatives are less toxic such as the cobaltocenium cation. Decontamination for toxic metals is different from organic toxins: because toxic metals are elements, they cannot be destroyed. Toxic metals may be made insoluble or collected, possibly by the aid of chelating agents **(Costa and Klein, 2006)**. Toxic metals can bio-accumulate in the body and in the food chain. Therefore, a common characteristic of toxic metals is the chronic nature of their toxicity. This is particularly notable with radioactive heavy metals such as thorium, which imitates calcium to the point of being incorporated into human bone, although similar health implications are found in lead or mercury poisoning. The exceptions to this are barium and aluminium, which can be removed efficiently by the kidneys **(Hogan, 2010)**.

Acknowledgments: This contribution is the result of the project implementation: VEGA no. 1/4435/07. This contribution is the result of the project implementation: Centre of excellence forwhite-green biotechnology, ITMS 26220120054, supported by the Research & Development Operational Programme funded by the ERDF.

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