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Honeybees as sentinels of lead pollution: Spatio-temporal variations and source appointment using stable isotopes and Kohonen self-organizing maps

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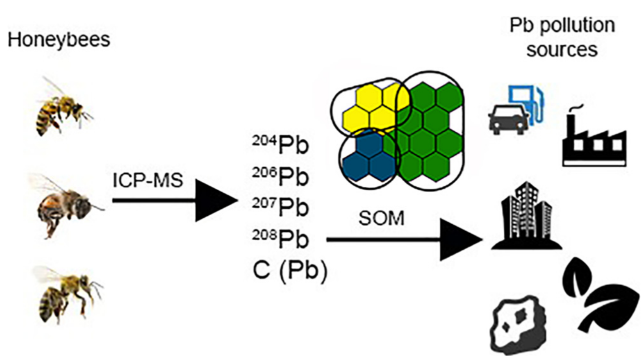
HIGHLIGHTS

- Honeybees are used to determine variations and source appointment of Pb pollution
- ANOVA, three isotope plots and SOM are recommended statistical methods
- SOM, used for the first time in combination with honeybees, showed separate clusters
- Determination of spatio-temporal variations and sources of Pb was successful

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, honeybees were used to determine spatio-temporal variations and origin sources of Pb. Lead concentrations and isotopic composition were used in combination with selected statistical methods. The sampling was carried out at five different locations in Serbia: urban region (BG), petrochemical industry (PA), suburban region (PV), rural region (MS) and thermal power plant region (TPP) during 2014. At PA and PV locations, samples were taken during multiple years. This is the first use of Kohonen self-organizing map (SOM) in combination with honeybees as bioindicators to determine spatio-temporal variations and origin of Pb pollution. It was observed that during the years Pb concentrations were in decline. Anthropogenic sources are most dominant in BG and TPP, in PA there are mixed sources of natural and anthropogenic origin and in PV Pb is of natural origin. It can be concluded that honeybees in combination with SOM can be used to differentiate between slight changes in spatio-temporal variations of Pb, as well as for source appointment.

1. Introduction

Industrialization has led to the increase in concentration of heavy metals in the environment (Diels et al., 2002; Rodriguez-Iruretagoiena et al., 2015; Sun et al., 2010). Human exposure to heavy metals can have negative effect on health (Li et al., 2016). Some of these elements,

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including Pb, can have carcinogenic effects even at very low levels (Itoh et al., 2014; Zhuang et al., 2009). It is important to track spatial and temporal variation in concentrations and sources of these metals to be able to characterize the paths and extent of human exposure. For differentiating spatio-temporal variations and sources of metal pollution most commonly used methods included analysis of different matrixes (PM, road dust, soil, water, etc.) in combination with different multivariate statistical methods (Bi et al., 2018; Taiwo et al., 2014).

For the determination of metal concentration in the environment several species were proposed as suitable biomonitors (Findik and Çiçek, 2011; García-Hernández et al., 2015; Jovičić et al., 2016; Prussia and Killingbeck, 1991). Most of these species are representative for pollution which is present only in one particular sphere of the environment: lithosphere, hydrosphere or atmosphere. Therefore, honeybees (*Apis mellifera* L.) and their products have been proposed as one of the most convenient bioindicators (Gutiérrez et al., 2015; Jones, 1987; Leita et al., 1996; Porrini et al., 2002; Rashed et al., 2009; Van Der Steen et al., 2012, 2016; Zarić et al., 2016, 2018). Metal concentrations measured in bodies of honeybees represent pollution from multiple spheres of the environment. Pollution can originate from atmosphere by direct deposition on hairy bodies of honeybees (Zarić et al., 2016); from lithosphere through plant absorption and transfer to flowers (pollen and nectar) visited by bees, or by erosion of topsoil and its deposition on flowers (Zarić et al., 2017); from hydrosphere by drinking water (Hladun et al., 2015; Silici et al., 2013). Honeybees have been used to determine differences in metal concentrations in environments with different anthropological pollution. They were used to distinguish pollution levels between industrial, urban, suburban and unpolluted regions (Conti and Botrè, 2001; Giglio et al., 2017; Perugini et al., 2011). Although both honeybees and their products (pollen, wax, honey, propolis) can be used as indicators of metal pollution, it was suggested that honeybees are more suitable indicator for the monitoring of Pb (Lambert et al., 2012). Numerous studies on the use of honeybees for the determination of lead pollution have been conducted in the past (Conti and Botrè, 2001; Gutiérrez et al., 2015; Lambert et al., 2012; Perugini et al., 2011; Porrini et al., 2002; Roman, 2005, 2010; Van Der Steen et al., 2015), however, honeybees were not used for the study of lead isotopes.

Stable Pb isotopes are used as tracers of Pb sources in the environment (Cheng and Hu, 2010), since their isotopic composition is not affected during industrial or environmental processes (Komárek et al., 2008). Some Pb sources have similar isotopic composition which depends on geochemical composition of Pb in parental material (Farmer et al., 2000). Therefore, sources of Pb pollution cannot always be determined using Pb isotopic ratios (Deljanin et al., 2015b).

Kohonen self-organizing map (SOM) is an unsupervised multidimensional data reduction method (Kohonen, 1982a, 1982b). In the past SOM was used for the studies of air, water or sediment pollution (Barron-Adame et al., 2007; Veses et al., 2014; Yang et al., 2012). SOM was also used in different biomonitoring studies (Deljanin et al., 2015a, 2015b; Samecka-Cymerman et al., 2009a, 2009b). So far SOM has not been used to study heavy metal concentrations in combination with honeybees as bioindicators.

The aim of this study was to analyze Pb concentrations and its isotopic composition in bodies of honeybees and to determine sources, spatial and temporal differences, using multivariate statistical analysis, including Kohonen self-organizing map (SOM). To the best of our knowledge this is the first time that Pb isotopes were determined in honeybees, and also the first time SOM is used for differentiation between samples of bees.

2. Materials and methods

2.1. Sampling locations and sampling

The study was conducted at five different locations in Serbia (Fig. 1) during three years. All samples were taken during summer (June and July) months. Between 50 and 100 honeybees (around 5–10 g) were

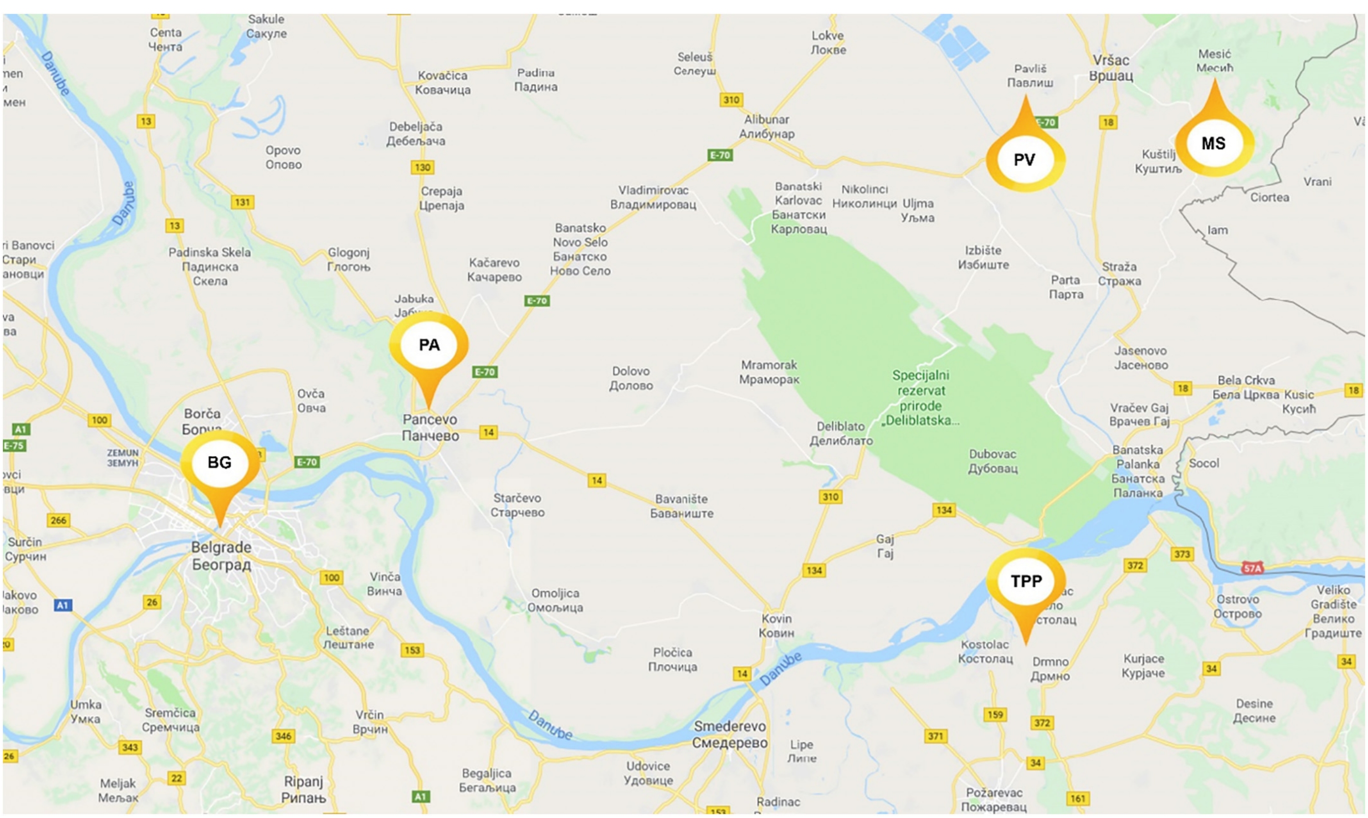

Fig. 1. Sampling site map.