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Lead isotopic composition in tree leaves as tracers of lead in an urban environment

Isidora V. Deljanin[a], Milica N. Tomašević[b,*], Mira P. Aničić Urošević[b], Davor Z. Antanasijević[a], Aleksandra A. Perić-Grujić[c], Mirjana Đ. Ristić[c]

[a] University of Belgrade, Innovation Center of the Faculty of Technology and Metallurgy, Karnegijeva 4, 11000 Belgrade, Serbia
[b] University of Belgrade, Institute of Physics, Pregrevica 118, 11080 Zemun, Serbia
[c] University of Belgrade, Faculty of Technology and Metallurgy, Karnegijeva 4, 11120 Belgrade, Serbia

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ABSTRACT

Biomonitoring has been frequently used as a tool for assessing anthropogenic influences on the environment, and trees have been recognized as efficient ecological indicators of urban air quality. Variations of stable lead isotopic ratios have been suggested as a suitable method for the Pb source identification. Substantial methodological points are yet to be resolved regarding isotopic signatures in environmental studies, and especially within plant biomonitoring. This study has been focused on an assessment of stable lead isotopic ratios ($^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$) for some common urban deciduous tree leaves (*Aesculus hippocastanum*, *Tilia cordata*, *Acer platanoides* and *Betula pendula*). The study was done in 2009, at the time when the leaded gasoline was still in use in Serbia, although during a time span when an increasing number of vehicles, using non-leaded gasoline, was evident. Thus, a decrease of the Pb emitted from leaded gasoline affected the atmospheric Pb concentrations. The measurements of the Pb isotopes were performed by ICP-MS on both, unwashed samples, and those shortly washed in bidistilled. The percentage of lead removed by applied rinsing was approximately the same in the first three above mentioned species (≈30%), while the exception was *B. pendula* (22%), indicating the highest retention for fine and coarse particulates due to thick epicuticular wax layer. It may be suggested that leaf washing is useful approach when the aim is an assessment of the isotopic composition in leaves after a certain period of exposure, assuming contribution of possible sources. However, in case of screening for possible sources in particular time sequence, leaves may remain unwashed. The obtained isotopic composition in leaf samples partially corresponded to that used in the lead additive in gasoline, implying also an influence of other Pb sources. The results imply that, among the investigated tree species, *B. pendula* could be the most efficient indicator in multiple Pb source identification. From the obtained Pb isotopic ratios, *A. hippocastanum* was confirmed as an appropriate biomonitor for the Pb atmospheric pollution, as previously shown from the Pb leaf concentration analyses.

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1. Introduction

Increasing industrialization and human activities intensify the emission of various pollutants into the environment and introduce various harmful substances into the atmosphere. Urban environment is heavily impacted by airborne particulates from traffic and industry (Adriano, 2001; Nriagu, 1990) and one of the most common pollutants is lead. The main anthropogenic sources of global lead pollution are leaded gasoline, coal combustion, industrial activities (e.g., metallurgy) and waste incineration; the initial recipient of Pb is atmosphere (Adriano, 2001; Mihaljevič, 1999). Even though leaded gasoline is not in use in most of the European countries any more, lead is still widely present in the environment (Hovmand et al., 2009). Lead enrichment in remote areas may also originate from long range transport (UN/ECE, 2006).

A wide range of activities, involving the release of this metal, often makes lead source identification very difficult, but by using stable lead isotopes, more reliable data can be provided. Ratios of stable isotopes of a particular element present in samples can be used to identify origins of the element in that sample, in a form of isotopic fingerprinting (Cheng and Hu, 2010; Komarek et al., 2008). The exploitation of Pb ore deposits and the subsequent release of Pb into the environment is of particular relevance for the use of Pb as a geochemical tracer. Lead emitted into the environment retains

* Corresponding author. Tel.: +381 11 3713145; fax: +381 11 3162190.
E-mail address: milica.tomasevic@ipb.ac.rs (M.N. Tomašević).

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the isotopic signature of the ore from which it was sourced. Radiogenic $^{206}Pb/^{207}Pb$ and $^{208}Pb/^{207}Pb$ ratios are commonly utilized as environmental tracers as they can be determined precisely and generally exhibit the greatest variability between different sources (Bollhöfer and Rosman, 2001; Komarek et al., 2006).

In general, mosses and lichens, due to absence of their root system, are considered as suitable biomonitors of trace and other elements atmospheric contamination (Aničić et al., 2007; Bargagli, 1998; Markert, 1993). In cases when mosses and lichens are not present, and due to their ability to capture and accumulate airborne trace elements, deciduous tree leaves have been used as biomonitors of urban trace and other element air pollution (Bargagli, 1998; Markert, 1993; WHO, 2000). However, depending on species, tree leaves are useful biomonitors (Ristić et al., 2013), especially in case of lead, which in leaves originates mostly from atmospheric deposition (Hovmand et al., 2009). Usually, in analysis of deciduous and evergreen tree species, tree leaves and needles have been used as bioaccumulators of trace elements in urban environments (Aničić et al., 2011; De Nicola et al., 2013; Rucandio et al., 2011; Suchara et al., 2011). Lead isotope tracing in plants were reported in several studies in recent years (LeGalley et al., 2013; Saunier et al., 2013; Sucharova et al., 2011; Tomašević et al., 2013).

Although different sample preparations can affect the leaf element concentrations, published data usually lack details concerning the washing procedure, including duration and type of washing media. By brief rinsing, the effects of prior precipitation events, which may wash off the atmospheric deposition, are avoided (Luyssaert, 2002; Tomašević et al., 2011). Thus, application of standardized sample preparation procedures is important for comparison of relevant data between different biomonitoring studies.

The aim of this study was to investigate whether it is possible to distinguish different sources of atmospheric lead emission in the Belgrade urban area by measuring lead isotopic ratios in leaves of different deciduous trees, common in this area. Also, selection of more appropriate bioindicator among the investigated tree species was considered. In addition, at the methodological level, the influence of brief washing the leaf surfaces as a part of sample preparation on lead isotopic ratios was analyzed.

2. Experimental

2.1. Study area

The sampling sites were situated in the urban area of Belgrade (φ = 44°49N, λ = 20°27E, Hs = 117 m), the capital of Serbia, with a population of about 1.7 million inhabitants. The samples were collected from three urban parks situated in heavy traffic area: Studentski park, Karađorđev park and Botanička bašta (Fig. 1). All parks are located in the center of Belgrade with intense and slow-moving traffic (>50 000 vehicles per day). Studentski park is a small park in the very city center, next to the bus terminal with high traffic density. Karađorđev park is situated close to Slavija, the busiest roundabout in Belgrade and with a highway in the vicinity. At Botanička bašta, samples were collected from the traffic-exposed streets bordering it, which are surrounded by many high buildings. In the studied period, leaded gasoline was still widely used in Serbia, since the official ban was put into effect at the beginning of 2011. Vehicles older than 10 years in year 2008 in Serbia added up to the total number with 67.5%, while only 17% of vehicles was less than 5 years old (Prvulović et al., 2008).

2.2. Sampling and sample preparation

Four tree species: horse chestnut (*Aesculus hippocastanum* L.), linden (*Tilia cordata* Mill.), Norway maple (*Acer platanoides* L.) and birch (*Betula pendula* Roth) were chosen since they are common in Belgrade city parks. These species had been used for biomonitoring in air quality studies (Bargagli, 1998; Baycu et al., 2006; Kim and Fergusson, 1994), using mainly leaves, since foliage has been proved as very efficient in trapping atmospheric particles (Nowak et al., 2006; Peachey et al., 2009). Also, some of the selected tree species had been previously used for biomonitoring of trace elements in Belgrade urban area (Aničić et al., 2011; Šućur et al., 2010; Tomašević et al., 2008, 2011). As described by Tomašević et al. (2011), the leaf samples were collected in the middle of the vegetation cycle, in July 2009. Leaves were cut with stainless scissors at about 2 m height above ground and taken by hand, wearing polyethylene gloves. Five

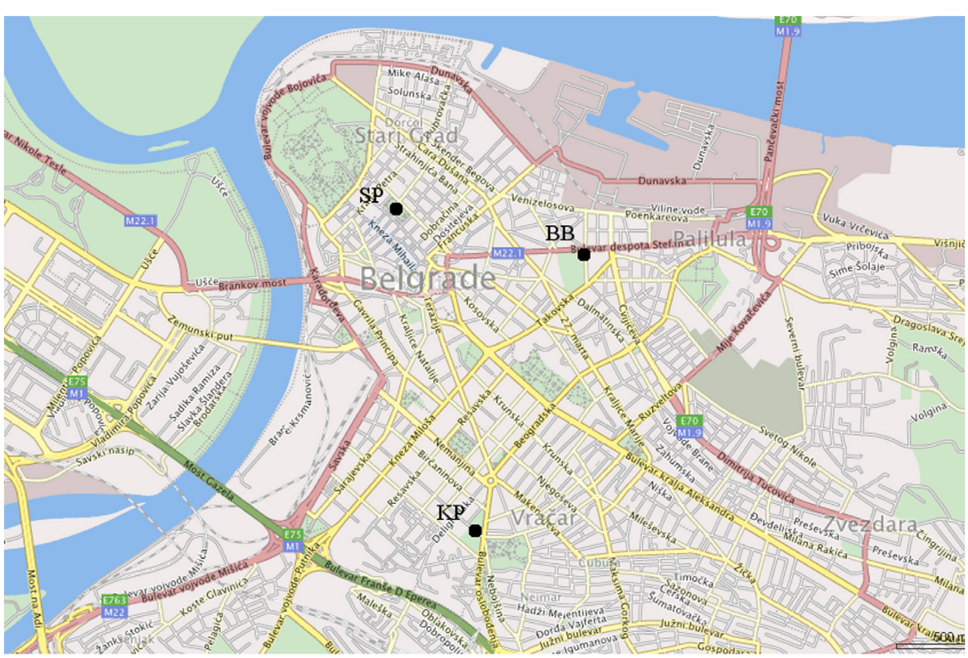

Fig. 1. Map of the studied sites in the Belgrade urban area: Studentski park (SP), Karađorđev park (KP) and Botanička bašta (BB).

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