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# EVALUATION OF THE HEAVY METALS CONTENT IN SOIL AND PLANT MATERIAL AT DIFFERENT DISTANCES FROM THE MOTORWAY E75 IN THE SECTION BELGRADE-PRESEVO (SERBIA)

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## Abstract

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The study included the screening of soil and plant material for the contents of some elements along the motorway E75 through Serbia, the section from Belgrade to Preљevo (a length of 400 km). Samples of soil and aerial parts of plant material were taken from both sides of lanes at a distance of about 8 km and at 10, 30, 50 and 400 m perpendicular to the direction of the highway.

In the soil samples it were determined pH in 1MKCl and content of total forms of Pb, Cu, As and Hg. The plant material was analyzed for Pb, Cu, As and Hg.

It can be concluded that, besides the anthropogenic pollution, which is reflected in the excessive use of plant protection products and fertilizers, and also the impact of air pollution from motor vehicles in certain sections of the examined area, the dominant influence on concentration of some examined elements comes from geochemical composition of parent material from which the soils were developed.

*Key words*: soil, plant, heavy metals, highway

## Introduction

The highway is the highest traffic class of roads. It is exclusively designed for fast motor traffic, which is operating in physically separated carriageway, usually width of 27.5 m, with at least two running and one stopping lane.

Observations presented in this paper were performed on the section of the highway E75, which is very frequent throughout the year, thus an impact of emissions from motor vehicles on soil and plant is especially emphasized. Since the soil along the highway mainly belongs to an agricultural area, examinations were aimed to determine whether there is and what is the level of the pollution of the soil in the examined area. In addition, soil, as an essential natural element, represents a very complex system, sensitive to different influences. It responds to small changes and this can cause a degradation of its main characteristics. Therefore, the relations arising from the different spheres of influence on soil also define the whole question of the relationship between the highway and the environment.

Human activities have dramatically changed the balance, biochemical, and geological cycles of many heavy metals. An assessment of the environmental risk caused by soil contamination is especially important for agricultural as well as non-cultivated areas due the fact that metals potentially harmful to human health persist in soils for a relatively long time and may transfer into the food chain in considerable amounts (Szynkowska et al., 2009). The content of heavy metals in soil and their impact on ecosystems can be influenced by many natural factors, such as parent material, climate, soil processes and anthropogenic activities such as industry, agriculture and transportation (Wei et al., 2007). Urban roadside soils are the ‘‘recipients’’ of large amounts of heavy metals from a variety of sources including vehicle emissions, coal burning waste and other activities (Jose Acosta et al., 2009; Saeedi et al., 2009). Automobile traffic pollutes roadside environments

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with a range of contaminants. Heavy metals are found in fuels, in the walls of fuel tanks, engines and other vehicle components, in catalytic converters, tires and brake pads, as well as in road surface materials (Zehetner et al., 2009; Deska et al., 2011). Contamination of the soil over the natural level by Pb, Zn, Cr and Co could be one of the indicators of anthropogenic environmental pollution. Fast development of industry, continuously increasing population, and intensification of road traffic are regarded as the foremost causes of ecosystem pollution in urban areas (Jankiewicz et al., 2010). Averages of Cu and Pb are compared with other cities around the world are significantly lower (Birke et al., 2000; Manta et al., 2002; Imperato et al., 2003; Ruiz-Cortes et al., 2005; Ljung et al., 2006; Bretzel et al., 2006), meaning the anthropic activities have a low impact on the soil heavy metal concentrations in the study area.

Soil responds to small changes and this can cause a degradation of its main characteristics. Therefore, the relations arising from the different spheres of influence on soil also define the whole question of the relationship between the route and the environment.

Numerous studies on roadside soil pollution have focused on total emission loads of heavy metals into open grassland and agricultural areas (Wheeler and Rolfe, 1979; Harrison, 1981; Ward et al., 1990; Viard et al., 2004; Hjortenkrans et al., 2006; Nabulo et al., 2006). Generally, total heavy metal contents in roadside soils were found strongly dependent on traffic density and showed an exponential decrease with distance from the road, reaching background levels 10-100 m. The natural soil concentration of heavy metals depends primarily on the parent material composition (De Temmerman et al., 2003). Recently, roadside soils have been an increasingly important sampling medium for assessing anthropogenic metal concentrations. A variety of heavy metals have been measured in roadside soils and reported by many researchers (Manta et al., 2002; Turer et al., 2003; Wang et al., 2005; Ljung et al., 2006; Wang et al., 2008; Petrotou et al., 2010).

The most frequently reported heavy metals of concern have been Pb, Zn and Cu. These heavy metals in roadside soils are principally derived from vehicle emissions, wear and tear on automobile parts (Wang et al., 2007).

Plants are the intermediaries through which elements from the soil and partly from the air and water are transferred to the human body by consumption. Some of the elements are necessary for growth and development of crops and without them they cannot survive, some of them have stimulating effect on plant growth, while a group of elements at high concentrations affects very toxically on the plants. An assessment of the environmental risk caused by soil contamination is especially important for agricultural as well as non-cultivated areas due the fact that metals potentially harmful to human health persist in soils for a relatively long time and may transfer into the food chain in considerable amounts (Szynkowska et al., 2009).

Concentrations of metals in plants vary with plant species (Alloway, 1994). Plant uptake of heavy metals from soil either occurs passively with the mass flow of water into the roots, or through active transport crosses the plasma membrane of root epidermal cells (Kim et al., 2003). Common source of soil and plant contamination with heavy metals is traffic (Gworek et al., 2011). Heavy metals are naturally present in the environment, however, the dynamic development of industry and motorization, as well as the continuing over-intensive use of various chemical compounds in agriculture, causes constantly increase of toxic heavy metals in the environment (Blagojević et al., 2009).

## Material and Methods

***Study area***

The area of study included the route of the E75 in the section from Belgrade, capital city of Serbia, to Preљevo, near the border with FYROM (**Former Yugoslav Republic of Macedonia)**, a distance of about 400 km. Composite soil samples were taken from each side of the lane at the distance of 8 km and at 10, 30, 50 and 400 m perpendicular to the direction of the highway from a depth of 0 do 30 cm. Sampling was conducted during August and September, 2010.

***Soil analysis***

In the laboratory, composite soil samples were dried and passed through a 2-mm sieve. Soil pH in water and in 1M KCl was analyzed potentiometrically, using glass electrode (SRPS ISO 10390:2007). Microelements and heavy metals were determined using inductively coupled plasma optical emission spectrometer ICAP 630 (ICP-OES), after the soils were digested with concentrated $HNO_3$ for extraction of total forms (Soltanpour et al., 1996).

The concentration of the trace element Hg was determined using the flame atomic adsorption spectrophotometer (AAS, GBC, SENSA DUAL HG), by the method of hydration, after the so-called “wet” combustion of samples, i.e. boiled in the mixture of concentrated acids: $HNO_3$ and $H_2O_2$, with filtration and the necessary dilution (Krishanamurty et al., 1976).

Reference soil NCS ZC 73005 (Soil Certificate of Certified Reference Materials approved by China National Analysis Center Beijing China) and reagent blanks were used as the quality assurance and quality control (QA/QC) samples during the analysis.

***Plant analysis***

Analyzed aerial parts of the study plant species were dried for 2 hours at 105$^{o}$C, using gravimetric method for determination of dry matter content of plant tissue. The dry matter determination is used to correct the sample element concentration to an absolute dry matter basis (Miller, 1998).

The content of heavy metals (Pb, Cu) and metalloid (As) in selected plants was determined with an inductively coupled plasma optical emission spectrometer ICAP 630 (ICP-OES), after the samples were digested with concentrated $HNO_3$/$H_2O_2$ for total form extraction.

The concentration of the trace element Hg in plant material was determined using AAS method by hydration after the "wet "combustion of the plant samples, meaning, boiling in the mixture of concentrated acids: $HNO_3$ and $HClO_4$, with filtration and the necessary dilution.

***Statistics and cartographic data processing***

Statistical analyses were performed with SPSS version 16 software (SPSS Inc, SYSTAT version 16.0, 2007). Results are presented in Tables 1 and 2. Cartographic data processing was performed by using ESRI' ArcView GIS 8.3.

## Results and Discussion

***Soil properties***

Based on the examination, (a total of 398 soil samples), the following results were obtained: In the examined area it is represented 40 types of soils, with 12 separate zones with different plant cover. Fields are dominating with 43% of examined area, abandoned production areas (neglected land-parlog) with of 23% of areas and meadows with 20%. The rest of the area is occupied by orchards, vineyards, gardens, vegetable gardens, forests, industrial crops and swamp surface. Based on recorded results, 40% of soil samples have limitations, such as strong acid reaction. In Table 1, statistical description of pH in 1MKCl and heavy metals in soils samples in the study area is shown.

Total concentrations of As above the MAC (maximum allowable concentration), were recorded in 1.51% of all samples and are equally spread out along the area of research (Official Gazette of Republic of Serbia, 1994).

The concentration of Cu above the MAC was recorded in 0.25% of the studied samples, half were located in the vineyards which are a consequence of the century – old practice of using copper- sulphate (Bordeaux mixture) and other copper containing fungicides to control vine downy mildew (Zovko and Romić, 2011).

The concentration of Pb above the MAC was recorded in 5.28% of the samples, except one sample in which was registered an extremely high concentration of this element of 215.45 mg $kg^{-1}$ and the position of the sample was from the area with 10 m distance from the road.

The concentration of Hg above the MAC was recorded in 0.75% of samples in the zone of 10-50 m, from the traffic lanes.

It can be concluded that in addition of anthropogenic pollution (excessive use of plant protection products and fer-

**Table 1**
**Statistical description of pH in 1M KCl and heavy metals end As in soils samples in the study area**

| | Statistical parameters | Soil pH (1M KCl) | Total Pb | Total Cu | Total As | Total Hg |
|---|---|---|---|---|---|---|
| | | | mg·kg$^{-1}$ | | | |
| Surface layer (0-30 cm) | Total N$^{o}$ of soils samples | 398 | 398 | 398 | 398 | 398 |
| | Min | 3.60 | 8.70 | 4.64 | 0.69 | 0.00 |
| | Max | 7.50 | 215.45 | 223.82 | 61.45 | 4.84 |
| | Mean | 6.02 | 40.94 | 25.25 | 7.69 | 0.13 |
| | SD | 0.93 | 27.19 | 19.53 | 6.85 | 0.39 |
| | VC | 0.86 | 665.67 | 283.95 | 39.88 | 0.10 |
| | Median | 6.00 | 32.24 | 22.12 | 6.02 | 0.08 |
| | Modus | 7.30 | 27.53 | 17.73 | 4.92 | 0.05 |
| Limits | Usual | | <50 | <50 | 0-10 | >0.1 |
| | Higher | | 50-100 | 50-100 | 10-25 | 1-2 |
| | MAC* | | 100 | 100 | 25 | 2 |
| | Extreme | | >150 | >200 | >100 | |

SD- standard deviation; VC- variation coefficient
*Official Gazette of Republic of Serbia, 23/94

tilizers), as well as the impact of air pollution from motor vehicles, in the Morava valley the dominant is geochemical pollution. The geological parent materials are river sediments and loess, and the geochemical background concentrations (in topsoil) range from, 8.7 to 17.5 mg $kg^{-1}$ for Cu, 26 mg $kg^{-1}$ for Ni, 18 to 23mg $kg^{-1}$ for Pb (Geochemical Atlas of Europe). The origin of increased concentration of Pb and As is also linked with ultra basic rocks, but the causes of pollution should be linked also to anthropogenic influence.

***Plant chemical composition***

Along with the sampling of soil material it was sampled also a plant material (vegetative mass) on corresponding locations (394), in order to determine contents of Pb, Cu, As, and Hg.

Plant metal uptake is influenced by soil factors including pH, organic matter, and cat ion exchange capacity as well as plant species, cultivars and age. The mobility and availability of heavy metals in the soil are generally low, especially when the soil is high in pH, clay and organic matter (Jung and Thornton, 1996; Rosselli et al., 2003).

It is important in which part of the plant the elements are collected. It is well known that Cu is more present in the root then in the stem, and Pb also has predominant presence in the root, while in the stem are in very low concentrations.

Table 2 presents average critical and toxic concentration of heavy metals in plats according to Kloke et al. (1984)* and Kastori et al. (1997)**.

Table 3 shows the percentages of Cu, As, Hg and Pb in the aerial parts of the plant material, tested depending on the distance from the road, and values of desirable, critical and toxic value of the tested elements.

Figure 1 presents the distribution of plant material in which the toxic concentrations of Cu were determined.

**Table 2**
**Average and toxic concentration of heavy metals in plants**

| Element | Normal content in plants Kloke et al. * | Critical contents for plant food Kloke et al. * | Critical concentration Kastori et al.** | Toxical concentration Kastori et al.** |
|---|---|---|---|---|
| | mg·$kg^{-1}$ | | μg·$g^{-1}$ | |
| Cu | 3-15 | 15-20 | 15 | 20 |
| As | <0.1-5 | 10-20 | | |
| Pb | 1-5 | 10-20 | 10 | 20 |
| Hg | <0.1-0.5 | 0.5-1 | 2 | 5 |

**Table 3**
**Percentages of Cu, As, Pb i Hg in the aboveground plant material tested depending on the distance from the road**

| Element | Limit values | Distance 10m | Distance 30m | Distance 50m | Distance 400m | Total for 394 samples |
|---|---|---|---|---|---|---|
| | | | **Cu,** mg·$kg^{-1}$ | | | |
| <15 | Desirable | 96.97% | 99.00% | 93.00% | 91.58% | 95.18% |
| 15-20 | Critical | 3.03% | 1.00% | 5.00% | 5.26% | 3.55% |
| >20 | Toxic | 0.00% | 0.00% | 2.00% | 3.16% | 1.27% |
| | | | **As,** mg·$kg^{-1}$ | | | |
| <10 | Desirable | 100.00% | 100.00% | 100.00% | 100.00% | 100.00% |
| 10-20 | Critical | 0.00% | 0.00% | 0.00% | 0.00% | 0.00% |
| >20 | Toxic | 0.00% | 0.00% | 0.00% | 0.00% | 0.00% |
| | | | **Pb,** mg·$kg^{-1}$ | | | |
| <10 | Desirable | 89.90% | 90.00% | 90.00% | 93.68% | 90.86% |
| 10-20 | Critical | 7.07% | 6.00% | 6.00% | 4.21% | 5.84% |
| >20 | Toxic | 3.03% | 4.00% | 4.00% | 2.11% | 3.30% |
| | | | **Hg,** mg·$kg^{-1}$ | | | |
| <2 | Desirable | 100.00% | 100.00% | 100.00% | 100.00% | 100.00% |
| 2-5 | Critical | 0.00% | 0.00% | 0.00% | 0.00% | 0.00% |
| >5 | Toxic | 0.00% | 0.00% | 0.00% | 0.00% | 0.00% |

The increased concentrations of this element were present on locations with vineyards and cultivated soil where it was noted that the values of this element in the soil is above the MAC, probably due to an excessive use of pesticides. Next, the results of the examinations show that the concentrations of As in plant material were within the desirable limits.

Figure 2 presents the distribution of collected plant material in which the toxic level of Pb was determined. It is evident that the points where the samples of plant material with increased concentrations of this element are matching with the locations where the total content of Pb was above MAC level. In the zone of examination up to 50 m from the road lane, the number of samples where the concentrations of Pb in plants were toxic is constant.

Based on the performed examinations, it was determined that in all plant samples the content of Hg was in values that correspond to desirable limits and that there were no samples with critical or toxic values.

## Conclusions

By the analysis of the results obtained it can be concluded that, besides the anthropogenic pollution, which is reflected in the excessive use of plant protection products and fertilizers, and also the impact of air pollution from motor vehicles in certain sections of the examination (including the available literary sources), the dominant contamination from examined elements comes from geochemical composition of bedrock.

The content of total forms of Pb above the MAC (maximum allowable concentration) was found in 5.28%, Cu in 0.25%, As in 1.51%, while Hg in 0.75% of the studied soil samples.

In the examined plant material, there were not detected toxic concentrations of arsenic and mercury.

Toxic content of Pb above the maximum value was recorded in 3.3% of plant samples in zones at the distance of 10-50 m from the road. The element Cu in the plant material

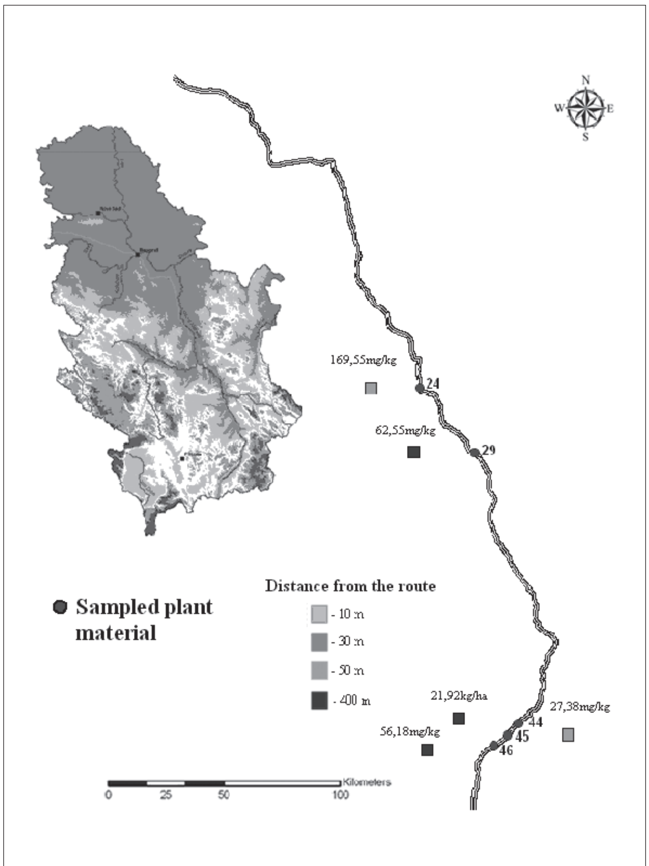


**Fig. 1. Locations of plant material in which the toxic concentrations of Cu was determined**

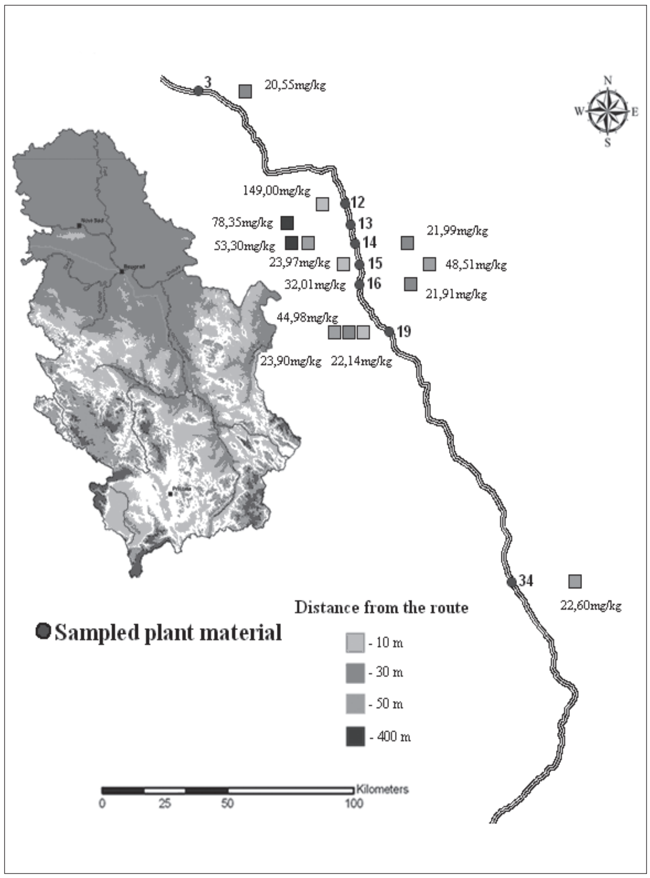


**Fig. 2. Locations of plant material in which it was determined toxic level of Pb**

is present in toxic concentrations in 1.27% samples of which two samples were from the vineyards at a distance of 50 m from the motorway, while three samples were from the zone at a distance of 400 m.

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