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# Distribution of Heavy Metals and Arsenic in Soils of Belgrade (Serbia and Montenegro)[1]

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*Soils of the urban and suburban area of Belgrade have been hardly studied, especially concerning their concentrations of potentially toxic metals. The present paper is aimed at determining the possible pollution in soils. The total acid soluble concentrations of heavy metals and As in the samples were determined. It was found that they were arranged in the order Zn > Ni > Pb > Cr > Cu > As > Hg > Cd in samples collected in the examined area (the order of the elements is based on their arithmetic mean concentrations). In all the samples collected at 0–10 and 40–50 cm depths from 46 selected sites, the contents of Pb and Zn were lower at the depth 40-50 cm. Using target values given by the Dutch Ministry of Housing, Spatial Planning and Environment, it may be concluded that Belgrade soil can, for the most part, be regarded as unpolluted. Traffic seems to be one of the main sources of these metals, but the influence of other factors cannot be excluded.*

**Keywords** Soil pollution, heavy metals, anthropogenic

## Introduction

Urban soils act as a sink for heavy metals and other pollutants and the possible sources of pollution include vehicle emissions (Sutherland *et al.*, 2000), industrial waste (Schuhmacher *et al.*, 1997), the atmospheric deposition of dust and aerosols (Simpson, 1996; Azimi *et al.*, 2005) and others (Thornton, 1991; Tiller, 1992). As urban areas are densely populated, the environmental quality of urban soil is closely related to human health. Furthermore, any contamination of urban soils could in turn cause groundwater contamination because the contaminants of polluted soils tend to be more mobile than those of unpolluted ones (Steinmann and Stille, 1997; Wilcke *et al.*, 1998; Davydova, 2005).

Heavy metals in urban areas have been a subject of great concern, due to their non-biodegradable nature and long biological half-lives within the human body. Most heavy metals in high concentrations have an adverse effect on human health, especially on the health of young children, who have a higher rate of heavy metal adsorption because of their active digestion system and sensitivity to hemoglobin. Heavy metals may accumulate in the human body and affect the central nervous system, causing heavy metal poisoning and/or

[1]Serbia and Montenegro was the name of the country at the time this paper was accepted; the name was changed to Serbia as of May 21, 2006.

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act as cofactors in many other diseases (Schwartz, 1994; Bellinger, 1995; Tripathi *et al.*, 2001).

Several authors have pointed out the need for a better knowledge of urban soils (De Kimpe and Morel, 2000; Nijkamp *et al.*, 2002). In the past few years, studies of urban soils in many cities have been carried out around the world. Some examples are Spanish (Sánchez Martín *et al.*, 2000; Madrid *et al.*, 2004) and Italian cities (Manta *et al.*, 2002; Imperato *et al.*, 2003). Other examples for European cities are Aberdeen (Paterson *et al.*, 1996), Athens (Chronopoulos *et al.*, 1997) and Oslo (Tijhuis, 2002). The differences among cities concerning population, living habits, industrial activities, etc., cause significant differences in the conclusions for each case.

There has been little research of urban soils in Serbia. The present study is the result of a preliminary sampling campaign, aimed at obtaining some knowledge of the heavy metal distribution and ascertaining whether some pollution is likely to exist in some areas in Belgrade, the capital of Serbia and Montenegro. By the end of 2004, Belgrade had about 1.6 million urban inhabitants, which is 20% of the total population of Serbia. With continuing population growth and economic development, the percentage of population living in Belgrade is increasing dramatically and, consequently, the environmental quality of urban soils is becoming more and more important in regards to human health. However, very little information is available about heavy metals of urban soils in Serbia. The objective of this study was to determine the total As, Cd, Cr, Cu, Zn, Ni, Pb, Hg concentrations of urban soils and non-urban soil profiles near the urban area of Belgrade. They were analyzed in soil samples taken at two depths, 0–10 and 40–50 cm.

## Experimental

### *Collection of Samples*

Belgrade is located in southeastern Europe, on the Balkan Peninsula, between 44 and 49°of northern latitude and 20 and 27° eastern longitude, 90–120 m above sea level. Belgrade has a moderate continental climate, with four seasons (cold winter and hot, humid summers with well distributed rainfall). The average annual temperature of Belgrade is 11.9°C with a mean monthly variation ranging from 0.4°C in January to 22°C in July and August, the mean annual precipitation is 685 mm and the average annual insolation is 2096 hours.

Belgrade territory covers an area of 763.87 $km^2$ (the inner-city area covers 360.05 $km^2$). According to the 2002 census, there were 1.576 million citizens in the larger-city area, and 1.274 million citizens in the inner-city area.

In the study areas, there are no specific point-sources of heavy metals and, therefore, heavy-metal contamination of the soils is derived from continuous urbanization and development, which can adversely affect human health and food crops grown in the contaminated area.

Forty-six urban soil profiles, distributed in different districts including the central part (4), the urban area (6), residential areas (6), campuses and vegetable gardens within the urban area (10), agricultural land (10) and non-urban soil profiles near the urban area (10), were randomly selected. Due to urbanization, intense urban activity and large traffic densities are now widespread throughout the city and less limited to the center, and about 60% of the samples were collected from sites near roadways. Samples were not collected from known contaminated sites and polluting industries are located apart from the examined area. The location of the sampling points is shown in Figure 1. The samples were collected

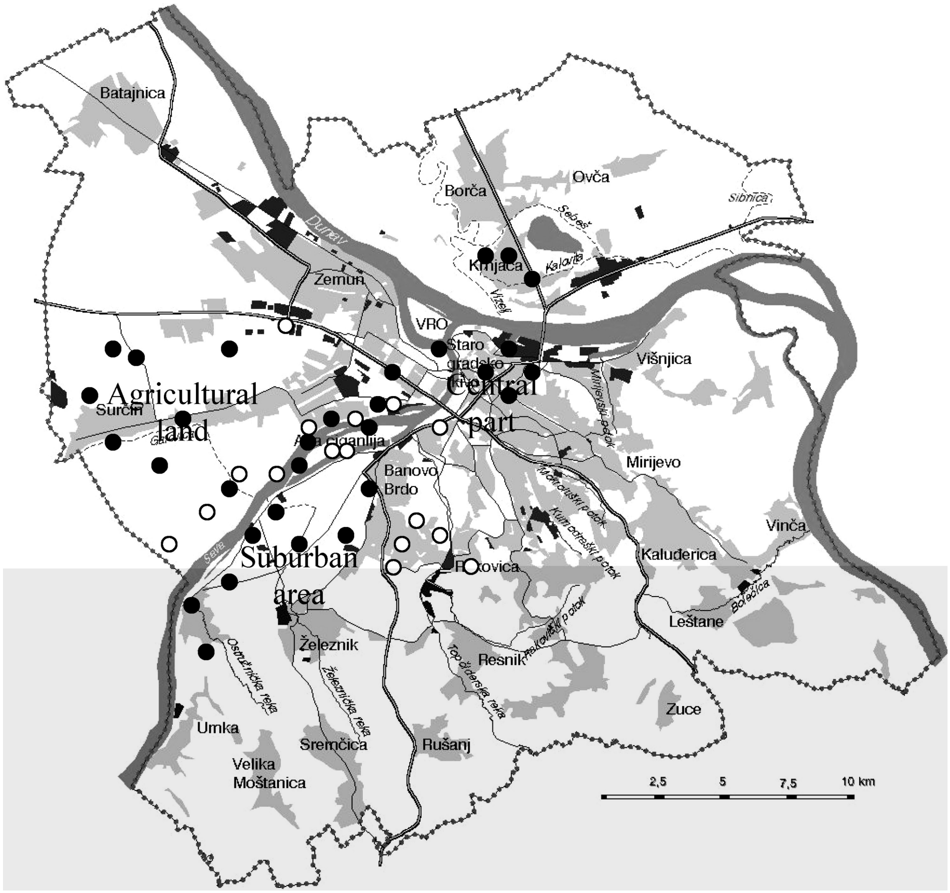


**Figure 1.** Map of Belgrade city with the location of the sampling sites: 2003 (solid circles), and 2004 (open circles).

in April and December 2003 and July and October 2004 at depths of 0–10 and 40–50 cm (US EPA, 1995) using an Auger device. They were stored in plastic bags for subsequent sample preparation and analysis.

## Methods and Instrumentation

The soil chemical properties were determined by the following standard procedures: pH in aqueous (1:2.5) extract, total P by $HClO_4$ digestion (Olsen and Sommers, 1982), total nitrogen by the Kjeldahl procedure (Radojević and Bashkin, 1999) and sulfate using EPA Method 375.4.

The samples were analyzed for 8 elements of which, apart from As (metalloid), the following 7 elements are heavy metals: Cr, Cu, Hg, Zn, Ni, Cd and Pb. The soil samples were extracted according to EPA Method 3050b. Briefly, 2 g (wet weight) of soil were treated with 10 $cm^3$ of 1:1 $HNO_3$. The sample was heated and refluxed for 10 to 15 minutes without boiling. After cooling, 5 $cm^3$ of concentrated $HNO_3$ were added and refluxed for 30 minutes. If brown fumes were generated, indicating oxidation of the sample by $HNO_3$, the addition of 5 $cm^3$ of conc. $HNO_3$ was repeated over and over until no brown fumes were given off by the sample indicating the complete reaction with $HNO_3$. The solution was allowed to evaporate to approximately 5 $cm^3$. The sample was cooled and 2 $cm^3$ of water and 3 $cm^3$ of 30% $H_2O_2$ were added. The vessel was covered with a watch glass and returned to the heat source for warming and to start the peroxide reaction. Care was taken to ensure that losses did not occur due to excessively vigorous effervescence. The sample was heated until effervescence subsided. After cooling, $H_2O_2$ (30%) was added in 1 $cm^3$ aliquots with warming until the effervescence was minimal. The sample was then heated at 50°C ± 5°C for 2 hours. After cooling, 10 $cm^3$ conc. HCl were added to the sample and covered with a watch glass. This mixture was refluxed at 95°C ± 5°C for 15 minutes or until the volume was 5 $cm^3$. This strong acid digestion dissolved almost all the elements that could become environmentally available. By design, elements bound in silicate structures are not normally dissolved by this procedure as they are not usually mobile in the environment.

The Cd, Cr, Cu, Ni, Pb and Zn concentrations of all the solutions were determined by flame atomic absorption spectrometry (FAAS) using a Varian Spectra AA-200 instrument. Hydride generation was used to analyse As (VGA-77, Spectra AA20+, Varian, Australia). The analysis of mercury was carried out by cold vapour atomic absorption spectrometry (Varian, Spectra AA-475). In the stage of digestion, three quality-assurance samples (a blank, a duplicate and a spike) were included with every 12 soil samples. Quality-control samples (analytical blanks and calibration standards) were included with every 10 samples in the determination of elemental concentrations in the digestion solution. The percentage recoveries of the elements from spiked soil samples ranged from 93.6 to 98.7%.

## Results and Discussion

Some physico-chemical properties of the soils used in the study are given in Table 1. All the samples were neutral to slightly alkaline regardless of the depth, with pH 6.8–9.3.

Summary statistics for the analysed elements in all the studied samples are presented in Table 2. The results reported in Tables 1 and 2 include both the 0–10 cm and 40–50 cm samples. The concentrations of Cd, Cr, Cu, Ni, Pb, As, Zn and Hg in Belgrade soils have a wide range of values. Cadmium was only detected in twelve samples and Hg was detected in thirteen samples. Consequently, the data for these elements are not presented in Table 2. Other elements were detected in all the samples. It was found that acid soluble elements were arranged in the order Zn > Ni > Pb > Cr > Cu > As > Hg > Cd in the samples collected in the examined area (the order of the elements is based on their arithmetic mean concentrations). Copper and Cr have arithmetic mean contents of around 30 mg $kg^{-1}$, Ni and Pb of around 60 mg $kg^{-1}$. Arsenic has the lowest mean concentration (7.2 mg $kg^{-1}$), while Zn has the highest mean concentration (118 mg $kg^{-1}$). The coefficients of variation

**Table 1**
Means, medians, standard deviations and ranges for the general properties of the soil samples

| | Mean | Median | SD | Range |
|---|---|---|---|---|
| pH | 8.36 | 8.45 | 0.40 | 6.8–9.3 |
| P (%) | 0.05 | 0.02 | 0.07 | 0.01–0.30 |
| N (%) | 0.18 | 0.15 | 0.14 | 0.01–0.8 |
| Sulfate (mg $kg^{-1}$) | 13.98 | 11.61 | 7.52 | 5–31.6 |

**Table 2**

Means, medians, coefficients of variation (mean/standard deviation), ranges and skewness coefficients for the metal contents (mg kg$^{-1}$) of the 46 samples

| | Cr | Cu | Zn | Ni | As | Pb |
|---|---|---|---|---|---|---|
| Minimum value | 8.2 | 2.6 | 29.4 | 7.8 | 1.95 | 5.2 |
| Maximum value | 65 | 136.1 | 783.9 | 117.7 | 58.8 | 306.9 |
| Mean | 32.1 | 28.3 | 118 | 68 | 7.2 | 55.5 |
| Median | 29.8 | 24.5 | 81 | 66 | 6 | 29.4 |
| Standard deviation | 14.9 | 17.1 | 120 | 28.9 | 8.4 | 46.0 |
| Coeff. of variation (%) | 45 | 60 | 100 | 43 | 84 | 150 |
| Skewness coefficient | 1.3 | 3.4 | 3.8 | 0.07 | 3.2 | 3.5 |
| SD of log-transformed | 0.20 | 0.23 | 0.27 | 0.13 | 0.25 | 0.19 |
| Distribution type | Log-normal | Log-normal | Log-normal | Normal | Log-normal | Log-normal |
| National regulation (mg kg$^{-1}$) (Pravilnik, 1994) | 100 | 100 | 300 | 50 | 25 | 100 |
| Percentage of polluted samples (according to national regulations) | — | 2 | 7 | 60 | 2 | 2 |

vary from 43% (Ni) to 150% (Pb). In most cases, the ranges of the examined metals show a rather large dispersion of the data, although somewhat smaller in the case of Cr, Cu and Ni. The high concentrations of Zn and Pb found at several sampling points, together with the large ranges observed in the data, are the reason for the large standard deviations for these metals. All the elements, except Ni, have positively skewed distributions, and they were log-transformed. The SD of the log-transformed data are also given in Table 2.

The total soil Pb content ranged between 5.2 and 306.9 mg kg$^{-1}$ with a median value of 55.5. Approximately 75% of the analyzed soils contained less than 50 mg kg$^{-1}$ of Pb.

Cd was in the range 0.60–1.3 mg kg$^{-1}$, while Hg was in the range 0.50–1.5 mg kg$^{-1}$ and all the values were below the maximum concentration according to national regulations, Pravilnik, Službeni list Republike Srbije, 1994 (3 mg kg$^{-1}$ for Cd and 2 mg kg$^{-1}$ for Hg).

Namely, heavy metal concentrations also varied to a great extent at different sampling points. The largest contents of Zn and Pb were found within the city center and urban part where traffic is very intense. Since coal combustion is still a source of heat in many houses in Belgrade, it can also contribute to Zn input through the deposition of atmospheric emission. There is no evidence of recent contamination due to the significant use of fertilizers and other amendments, since the amounts of the examined metals were low in agricultural soil.

The possible pollution can also be evaluated according to Dutch guidelines due to the fact that national legislation was established ten years ago. We chose to use the latest (2000) Dutch Ministry of Housing, Spatial Planning and Environment target and intervention values for soil remediation (Table 3) on the basis that it is a long established (first introduced in the early 1980s), tried and tested scheme, where the intervention values are based on extensive studies of both the human and eco-toxicological effects of soil contaminants.

The Dutch intervention values for soil/sediment remediation are considered to be numeric manifestations of the concentrations above which it may be said that there is a case of serious contamination. These values indicate the concentration levels of metals above which the functionality of the soil for human, plant and/or animal life may be seriously

**Table 3**
Target values and soil remediation intervention values for selected metals in soils from the Dutch Ministry of Housing, Spatial Planning and Environment

| | Hg | As | Ni | Zn | Cu | Cr | Pb | Cd |
|---|---|---|---|---|---|---|---|---|
| Target value (mg $kg^{-1}$) | 0.3 | 29 | 35 | 140 | 36 | 100 | 85 | 0.8 |
| Intervention value (mg $kg^{-1}$) | 10 | 55 | 210 | 720 | 190 | 380 | 530 | 12 |

compromised or impaired. Target values indicate the level at which there is a sustainable soil quality and gives an indication of the benchmark for environmental quality in the longterm on the assumption of negligible risk to the ecosystem.

It may be concluded that Belgrade soils, for the most part, can be regarded as unpolluted since the mean metal concentrations in the soils did not exceed the target values. The maximum values for Cu, Pb, Hg and Cd were higher than the target values indicating contamination not sufficiently high to require remediation/intervention, but not excluding risk to the ecosystem. The mean concentration of Ni exceeded the Dutch Ministry target value, indicating soil pollution. However, except for a few abnormally high values that are an expression of the influence of pollutant sources, relatively high levels of nickel in the investigated soils were interpreted to reflect a natural enrichment by weathering and pedogenesis processes. However, the maximum values for Zn and As exceeded intervention values, indicating serious contamination at some locations.

The heavy metal average contents for 0–10 cm depth are compared in Table 4 to those found in some cities in the world. Compared to average concentrations in urban soils in the world, the Ni mean concentration is 3–5 times higher. The mean values of other heavy metals in the analyzed soils are generally lower than those reported for samples from some large and/or industrialized cities (i.e. Madrid, London), but are similar to those measured in small cities such as Rostock.

**Table 4**
Average heavy metal concentrations in urban soils from different cities across the world mg $kg^{-1}$

| City | Cr | Cu | Pb | Zn | Ni | Reference |
|---|---|---|---|---|---|---|
| London | n.a. | 73 | 294 | 183 | n.a. | Thornton (1991) |
| Madrid | 75 | 72 | 161 | 210 | 14 | De Miguel *et al.* (1998) |
| Rostock | 48 | 35 | 83 | 100 | 30 | Kahle (2000) |
| Hong Kong | n.a. | 24.8 | 93.4 | 168 | n.a. | Li *et al.* (2004) |
| Palermo | 39 | 77 | 253 | 151 | 19.1 | Manta *et al.* (2002) |
| Naples city | 74 | 11 | 262 | 251 | n.a. | Imperato *et al.* (2003) |
| Nanjing city | 84.7 | 66.1 | 107.3 | 162.6 | n.a. | Lu *et al.* (2003) |
| Sevilla | 42.8 | 64.6 | 161 | 107 | 23.5 | Madrid *et al.* (2004) |
| Belgrade* | 33.2 | 29 | 53.2 | 129.1 | 67.4 | |

n.a.: not available.
*The Belgrade results are the means of the 0–10 cm samples.

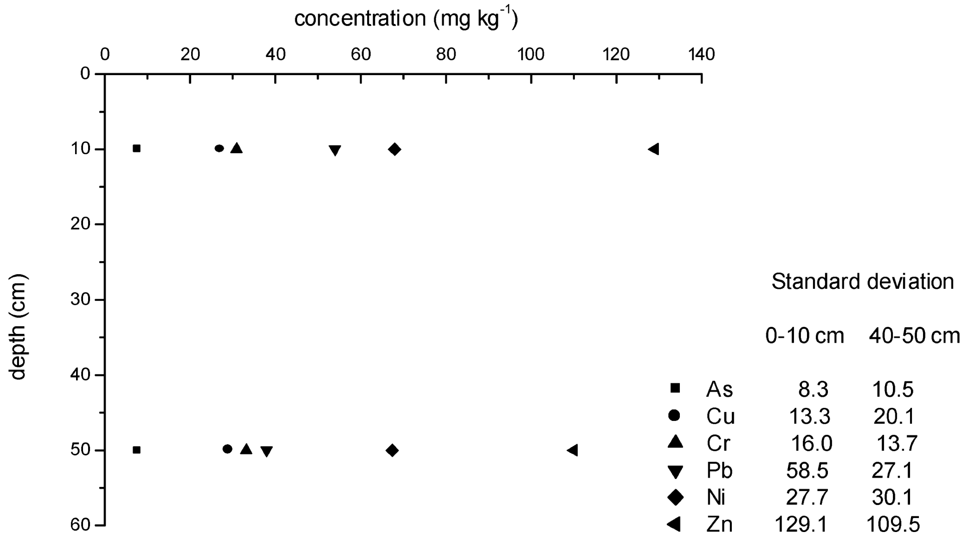


**Figure 2.** Vertical distribution of As and the examined heavy metals in soils from Belgrade (the values shown are mean concentrations).

In all the samples collected at two depths from 46 selected sites, the contents of Pb and Zn are lower at 40–50 cm depth (Figure 2). The mean concentration of the other elements is almost the same at these two depths.

The enrichment of Pb in these soils could be attributed to increased metal inputs deriving from the expanded utilization of leaded fuels, which are still in use in Serbia. It is probable that automobile exhausts are also a source of Zn, since higher concentrations of Pb and Zn were found in same samples, those taken from the city center and the urban part. As Zn is indicated as a vulcanization agent in vehicle tires (Alloway, 1990), this source may contribute to the high Zn content in the soil.

## Conclusions

It may be concluded that there is no significant degree of metal pollution in the examined soils within the urban and suburban area of Belgrade. However, the Belgrade area has been affected by human activity leading to the accumulation of Pb and Zn. The topsoil enrichment clearly revealed an anthropogenic origin of the pollution.

The source of this moderate pollution is not likely to be industrial, considering that most of the polluting industries are located apart from the examined areas. Traffic seems to be one of the main sources of these metals, but the influence of other factors cannot be excluded.

This study presents the basis for defining the requirements for further research in order to characterize the heavy metals and As contents in the soils of Belgrade as a function of their mineralogical and chemical characteristics, as well as to identify areas of potential toxicity due to heavy metal participation in biochemical cycles.

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