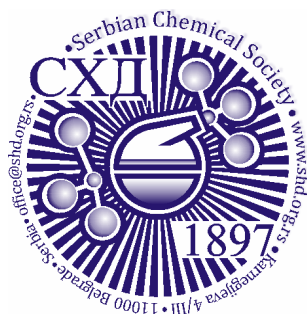




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Geochemical assessment of chemical pollutants in soil of Belgrade, Serbia: A case study of geogenic and anthropogenic influences

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Abstract: This study provides an integrated geochemical assessment of chemical pollutants in urban soil profiles (0–10 cm and 40–50 cm) collected across Belgrade, Serbia, during 2023. Ninety six samples were obtained by the Environmental Protection Agency of the Republic of Serbia (SEPA) from nine municipalities—including Stari Grad, Palilula, Zemun, Vračar, Čukarica, Novi Beograd, Rakovica, Voždovac and Surčin—covering recreation areas, major road corridors, industrial zones, agricultural plots and sites near landfills and water supply facilities. Concentrations of eight heavy metals (Cu, Zn, Pb, Cd, Ni, Cr, As, Hg) and selected organic pollutants (BTEX, PAHs, PCBs, mineral oil fractions) were evaluated relative to national limit values. Most analytes remained below regulatory thresholds, yet notable exceedances occurred: Pb reached 333 mg kg⁻¹ at Zemun and Cd 5.46 mg kg⁻¹ at Surčin (both at 50 cm), while mineral oils peaked at 458 mg kg⁻¹ in Stari Grad and PCBs surpassed limits at a Palilula water supply site. Depth dependent patterns indicated limited mobility for most metals, with Cd and Hg showing enrichment at 50 cm. Multivariate analysis distinguished an anthropogenic, traffic linked group (Cu, Zn, Pb, As, Hg; BTEX, mineral oils) from a geogenic Ni–Cr group associated with Serbia's serpentinite derived soils.

Keywords: soil geochemistry; heavy metals; organic contaminants; anthropogenic sources.

INTRODUCTION

Soil contamination by trace metals and chemical pollutants is a growing environmental concern in urban areas worldwide. Urban soils act as long-term sinks for both inorganic and organic contaminants derived from natural and anthropogenic sources. Traffic emissions, industrial activities, construction materials and historical land use contribute to the accumulation of heavy metals

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and organic pollutants in surface horizons, while soil properties control their retention and vertical migration. Understanding the depth distribution of contaminants is therefore essential for assessing exposure pathways relevant to human health and groundwater protection.

This study focuses on Belgrade, the capital of Serbia, which is constantly facing environmental problems caused by intense urbanization and heavy vehicle traffic, aiming to discriminate geogenic background content from anthropogenic enrichment using multivariate statistics and standard geochemical methods.

Soil is a finite and indispensable resource that forms the foundation of terrestrial ecosystems, providing a multitude of vital functions and ecosystem services. It is the primary medium for food and biomass production, a critical component of the water cycle responsible for filtration and purification, and a significant reservoir for carbon and nitrogen.¹

Globally, however, this essential resource is under increasing threat from a wide range of degradation processes, including erosion, loss of organic matter, compaction, and most critically, pollution.

Soil contamination, stemming from both point source and diffuse pollution, represents a major challenge to human health, food security, and environmental stability. Anthropogenic activities such as industrial production, intensive agriculture, transportation, and waste disposal introduce a diverse array of chemical pollutants into the soil matrix. These contaminants, which include heavy metals and persistent organic pollutants, can compromise soil fertility, enter the food chain, and leach into groundwater, posing long-term risks to both human and ecological systems.² Addressing this complex issue requires a holistic and multi-faceted approach, encompassing comprehensive monitoring, robust regulatory frameworks, and targeted remediation strategies.

In Serbia, soil quality is regulated through limit and remediation values defined for heavy metals and selected organic substances. However, integrated studies simultaneously addressing inorganic and organic contaminants, particularly with respect to depth-dependent behaviour, remain limited.

The aim of this study is to: (i) characterise the content of heavy metals and organic contaminants in urban soils, (ii) compare surface and subsurface distributions, (iii) assess compliance with Serbian regulatory values, and (iv) discuss environmental implications with emphasis on human health and groundwater pathways. Beyond this descriptive characterisation, the study further aims to (v) apply multivariate statistical methods to jointly interpret heavy metal and organic pollutant relationships and quantitatively discriminate geogenic from anthropogenic sources.

The underlying dataset of pollutant content was collected as part of Serbia's national soil monitoring programme, conducted by the Environmental Protection Agency of the Republic of Serbia (SEPA); the original scientific contribution of

this manuscript lies not in this dataset itself, but in how it is interpreted. Three elements distinguish this study from a standard monitoring report. First, heavy metals and organic pollutants are analysed jointly rather than separately, using Principal Component Analysis and metal–organic cross-correlation, applied here for the first time to this dataset in order to discriminate geogenic from anthropogenic sources statistically rather than qualitatively. Second, the depth-dependent behaviour of each pollutant is not only reported but mechanistically explained, linking surface/subsurface content patterns to specific physicochemical drivers — soil pH, clay content and organic matter for metals, and volatility and lipophilicity for organic pollutants — rather than treating depth as a purely descriptive variable. Third, the results are systematically benchmarked against recently published (2021–2026) research from other Serbian cities and comparable European urban centres, allowing the Belgrade findings to be assessed for regional consistency rather than interpreted in isolation. These three elements — joint statistical source apportionment, mechanistic depth interpretation, and systematic international benchmarking — constitute the novelty of this study relative to the underlying national monitoring data.

RESEARCH AREA

Belgrade is one of the oldest cities in Southeastern Europe, on the Balkan Peninsula. It lies at the confluence of the Sava River with the Danube, on the hairline between the alluvial plains. It is surrounded by water on three sides and is therefore called the "gate" of the Balkans and the "gate" of Central Europe. Its coordinates are: 44°49'14" north latitude 20°27'44" east longitude and altitude of 116.75 m. It covers 3.6 % of the territory of the Republic of Serbia and is divided into 17 municipalities. According to the 2022 census, about 1.6 million people live in the capital of the Republic of Serbia. Urbanization, thermal power plants using coal, individual fireplaces, dense traffic, developed industry, significantly contribute to the pollution of Belgrade, which is a serious problem for the health of the population and preservation of the environment.³

EXPERIMENTAL

The analysis of soil quality in this study focuses on two primary categories of chemical pollutants: heavy metals and organic compounds. Heavy metals, such as copper (Cu), zinc (Zn), lead (Pb), chromium (Cr), nickel (Ni), cobalt (Co), and cadmium (Cd), are naturally occurring elements that become environmental contaminants when their content is elevated by human activities. Common anthropogenic sources include industrial emissions, vehicle exhaust (historically from leaded gasoline), and agricultural inputs such as fertilizers and animal manure, which can contain high content of Cu and Zn. As we all know, heavy metals are natural ingredients of soil. The presence of microorganisms makes the soil a dynamic system, where heavy metals in interaction with natural compounds change their oxidation states and precipitate. Once deposited, the environmental fate and bioavailability of these metals are strongly influenced by the chemical forms of heavy metals, soil characteristics, particularly pH,

organic matter content and clay percentage. Generally, a higher pH and increased organic and clay content promote the sorption of heavy metals, reducing their mobility and potential for leaching or plant uptake.^{4,5}

Organic pollutants, specifically Polycyclic Aromatic Hydrocarbons (PAHs) and mineral oil hydrocarbons, are also of significant concern. PAHs are a group of chemical compounds formed during the incomplete combustion of organic materials such as coal, oil, gas, wood, and garbage. Sources include industrial emissions, vehicle exhaust, and waste incineration.⁶ Similarly, mineral oil hydrocarbons are found in crude oil and refined petroleum products, and their presence in soil is typically the result of accidental spills from industries, transportation, or unmanaged waste. These compounds are highly hydrophobic, meaning they have a strong affinity for soil organic matter. This characteristic causes them to bind tightly to soil particles, limiting their mobility but also contributing to their long-term persistence in the environment. The degradation of these compounds is primarily mediated by microbial metabolism, a process that can be supported by phytoremediation techniques. Some PAHs have endocrine disrupting properties and some have shown to affect the immune system.^{7,8}

The Us Environmental Protection Agency (EPA) has designated sixteen PAHs as priority environmental pollutants (USEPA, 2002).⁹

They are released to the environment through two sources, the anthropogenic and natural processes. Anthropogenic input was regarded as the main source for the elevated content of PAHs in the environmental matrix. There are two types of anthropogenic sources of PAHs: petrogenic and pyrogenic. Petrogenic sources include crude oil and petroleum products. Pyrogenic sources form by the incomplete combustion of organic matter in industrial operation, garbage incinerators, power plants, vehicle engines and forest fires.¹⁰

It is useful to distinguish the two broad categories of trace-element source referred to throughout this manuscript. Geogenic elements are those present in soil because they were already contained in the parent rock from which the soil formed; they are released gradually through weathering and pedogenesis, so their spatial distribution tracks the underlying geology rather than land use, and their content with depth is typically uniform or even increasing, since it reflects the parent material itself rather than a surface input. Anthropogenic elements, by contrast, are added to the soil by human activity — combustion, vehicular traffic, industrial emission, waste disposal or agrochemical application — in excess of this natural background; their distribution instead tracks human activity (roads, industrial zones, landfills), they are typically surface-concentrated unless subject to long-term downward migration, and they tend to co-vary with other pollutants released from the same activity. This distinction matters beyond source attribution: an elevated geogenic content reflects a naturally metal-rich substrate and does not by itself indicate contamination requiring remediation, whereas an elevated anthropogenic content reflects an ongoing, addressable pollution source.

A critical factor in the interpretation of soil heavy metal content in Serbia is the country's unique geological background. Numerous studies have established that certain regions, particularly in central and western Serbia, are underlain by ultramafic rocks, commonly referred to as serpentinites. These parent rocks are naturally enriched with specific trace elements, including nickel (Ni), chromium (Cr), and cobalt (Co).^{11,12} The presence of these geogenic sources means that background content of Ni and Cr can be significantly higher in these soils compared to typical values found in other parts of Europe.^{13,14,15}

The Republic of Serbia has established a legal framework for environmental protection, with a specific focus on soil quality. This regulation defines Maximum Allowed Quantities (MDK) for a list of hazardous and harmful substances.^{16,17}

The Serbian legal framework also includes a provision for systematic soil quality monitoring and emphasizes the "polluter pays" principle, placing the responsibility for remediation on the party that caused the pollution.

In nine municipalities of the city of Belgrade, 96 soil samples were taken at depths of 0-10 cm and 40-50 cm, which represents the upper layer of soil that is most susceptible to pollution. The sampling sites were chosen to reflect the specifics of land use in the region. The municipalities, sampling locations, their land-use description and geographical coordinates are presented in Table I.

TABLE I. Sampling municipalities, locations and their land-use description

Municipality	Land-use / Site description	Coordinates
Vračar	Recreation zone	44°79'75.5"N, 20°47'53.2"E
	Recreation zone	44°79'36.7"N, 20°44'39.2"E
	Traffic road	44°79'34.5"N, 20°48'33.8"E
	Traffic road	44°80'01.9"N, 20°46'49.3"E
	Water supply source	43°79'35.2"N, 20°46'94"E
Čukarica	Recreation zone	44°77'82.7"N, 20°43'37.9"E
	Near landfill	44°73'46.7"N, 20°41'86.3"E
	Near landfill	44°72'25.8"N, 20°40'13.3"E
	Agricultural land	44°73'63.1"N, 20°39'55.1"E
	Water supply source	44°77'31.9"N, 20°36'36.8"E
Voždovac	Residential zone	44°77'44.9"N, 20°39'91.9"E
	Recreation zone	44°78'52.4"N, 20°46'67.7"E
	Traffic road	44°76'69.4"N, 20°48'17.9"E
	Industrial zone	44°76'48.4"N, 20°49'20.3"E
	Water supply source	44°76'75.8"N, 20°48'81.7"E
Novi Beograd	Water supply source	44°75'40.7"N, 20°50'57.2"E
	Water supply spring	44°79'68.1"N, 20°40'20.8"E
	Water supply spring	44°79'47.7"N, 20°38'47.2"E
	Traffic road	44°80'35"N, 20°39'99"E
	Pedagogical institution	44°80'75.1"N, 20°38'55.5"E
Zemun	Near landfill	44°79'53"N, 20°36'76.8"E
	Near landfill	44°79'34.5"N, 20°48'33.8"E
	Near landfill	44°88'72.3"N, 20°31'72"E
	Near landfill	44°84'08.5"N, 20°35'25.5"E
	Near landfill	44°83'08.5"N, 20°38'77.1"E
Stari Grad	Agricultural land	44°83'93.2"N, 20°29'01.3"E
	Recreation zone	44°83'91"N, 20°41'86.7"E
	Industrial zone	44°86'26.1"N, 20°35'59.9"E
	Traffic road	44°81'93.1"N, 20°47'44.1"E
	Traffic road	44°81'98.3"N, 20°45'25.9"E
Zvezdara	Recreation zone	44°82'22.8"N, 20°44'94.3"E
	Recreation zone	44°82'65.3"N, 20°44'86.4"E
	Recreation zone	44°80'50.7"N, 20°47'86.9"E
Zvezdara	Traffic road	44°78'28.2"N, 20°50'43.3"E
	Traffic road	44°77'20.9"N, 20°51'83.3"E

Savski Venac	Traffic road	44°80'67.4"N, 20°50'95.9"E
	Near landfill	44°76'53.1"N, 20°53'84.7"E
	Agricultural land	44°79'69.1"N, 20°53'88"E
	Recreation zone	44°79'07.1"N, 20°45'24.9"E
	Recreation zone	44°79'94.9"N, 20°45'03.2"E
	Traffic road	44°78'96.3"N, 20°45'57.2"E
	Traffic road	44°77'53.6"N, 20°43'92.4"E
	Water supply source	44°78'35.8"N, 20°44'28.6"E
	Industrial zone	44°85'24.83"N, 20°53'62.51"E
	Agricultural land	44°88'93"N, 20°47'28.1"E
Palilula	Near landfill	44°81'34.7"N, 20°52'81.4"E
	Water supply source	44°82'11.94"N, 20°53'37.87"E
	Water supply source	44°82'76.37"N, 20°54'43.86"E

The soil samples were analyzed for a comprehensive suite of physical, chemical, and pollutant parameters. These parameters can be grouped into the following categories:

- **Physical/Chemical Properties:** Organic matter content, Clay, Moisture content, pH in H₂O.
- **Heavy Metals and Other Elements:** Copper (Cu), Zinc (Zn), Lead (Pb), Cadmium (Cd), Nickel (Ni), Chromium (Cr), Arsenic (As), Mercury (Hg).
- **Organic Pollutants:** Toluene, Xylene, Benzene, total Polychlorinated Biphenyls (PCBs), total Polycyclic Aromatic Hydrocarbons (PAHs), along with the sum of C10-C40 mineral oil hydrocarbons.

The selection of this analyte suite reflects the convergence of three considerations. First, all of the measured metals and organic pollutants are explicitly regulated under the current Serbian national soil-quality framework, for which Maximum Allowed Quantities have been prescribed,^{16,17} ensuring that the results are directly comparable with legally binding national thresholds; the same substances also fall within the scope of pollutants addressed by the proposed EU Soil Monitoring Law.¹ Second, most of the selected substances rank among the highest-priority hazardous substances identified internationally: arsenic, lead, mercury, PCBs, benzene, cadmium, PAHs and chromium are all listed among the top twenty entries of the ATSDR/CERCLA Substance Priority List, which ranks hazardous substances according to their toxicity, frequency of occurrence at contaminated sites, and potential for human exposure,¹⁸ while the PAHs analyzed correspond to the priority PAH list established by the US EPA.⁹ Third, the specific combination was chosen to capture the dominant pollution pathways expected in a large, traffic-dense, historically industrialized European capital such as Belgrade: Pb, Zn and Cu are established tracers of vehicular traffic (exhaust, tire and brake wear), Ni, Cr and Co additionally carry a strong geogenic signal linked to Serbia's serpentinite geology, Cd and As are sensitive indicators of diffuse industrial and agrochemical inputs, and BTEX, PAHs, PCBs and mineral oil hydrocarbons together represent the principal classes of petroleum-derived and legacy industrial organic contamination. This combination therefore allows both regulatory-compliance assessment and geogenic/anthropogenic source apportionment to be carried out within a single, internationally comparable dataset.

Therefore, soil samples were collected from representative urban locations. At each site, composite samples were taken from two depths: surface soil (0-10 cm) and subsurface soil (40-50 cm). Samples were air-dried, homogenised and sieved prior to analysis.

All analytical determinations were carried out by the National Laboratory of the Environmental Protection Agency of the Republic of Serbia (SEPA), accredited according to the international quality management standard SRPS ISO/IEC 17025:2017.¹⁹ Prior to instrumental analysis, solid soil samples were prepared by acid digestion. Heavy metals (Al, As, Co, Cd, Cu, Ni, Zn, Pb, Sb, Fe, Mn, Cr, B and Hg) were determined by the Section for Inorganic Residual Analysis using ultraviolet–visible (UV/VIS) spectrophotometry, flame atomic absorption spectrometry (FAAS), electrothermal atomic absorption spectrometry (ETAAS), direct mercury analysis (DMA) for Hg, and inductively coupled plasma mass spectrometry (ICP-MS) and optical emission spectrometry (ICP-OES); of these, Cu, Zn, Pb, Cd, Ni, Cr, As and Hg are reported in the present study. Organic pollutants were determined by the Section for Organic Residual Analysis using gas and liquid chromatography coupled with mass spectrometric, fluorescence, tandem mass spectrometric and time-of-flight mass spectrometric detectors; of these, the polycyclic aromatic hydrocarbons, polychlorinated biphenyls, BTEX compounds and mineral oil hydrocarbons are reported in the present study. Organic matter content was measured by loss-on-ignition at 550 °C. All applied methods are accredited according to SRPS ISO/IEC 17025:2017, and the laboratory regularly participates in external proficiency testing (PT) schemes for all accredited parameters to verify measurement accuracy, precision and traceability.¹⁹

The measured content was compared with Serbian regulatory limit values (Official Gazette RS 30/2018 and 64/2019).¹⁷ Depth-dependent trends were evaluated to infer potential sources of pollution and mobility of pollutants.

RESULTS AND DISCUSSION

The analysis of 96 soil samples from nine Belgrade municipalities provides detailed information on their physical, chemical characteristics and pollutant content. The results are presented as a series of tables for easier direct comparison and evaluation.

Descriptive statistics for heavy metal content in surface (10 cm) and subsurface (50 cm) soils are summarised in **Table 2** and **Table 3**, respectively.

TABLE II. Descriptive statistics of heavy metals at 10 cm depth

Element	Min (mg kg ⁻¹)	Median (mg kg ⁻¹)	Mean (mg kg ⁻¹)	Max (mg kg ⁻¹)	SD	LV (mg kg ⁻¹)
Cu	5.76	24.00	27.15	98.80	13.95	100
Zn	35.80	67.19	72.44	158.06	25.17	300
Pb	11.96	24.35	33.44	317.49	44.54	100
Cd	0.10	0.18	0.22	1.32	0.20	3.0
Ni	24.00	44.55	50.09	105.00	18.44	50
Cr	23.33	47.40	48.79	88.10	15.54	100
As	6.24	9.80	10.39	22.30	2.95	25
Hg	0.035	0.035	0.052	0.40	0.068	1.0

TABLE III. Descriptive statistics of heavy metals at 50 cm depth

Element	Min (mg kg ⁻¹)	Median (mg kg ⁻¹)	Mean (mg kg ⁻¹)	Max (mg kg ⁻¹)	SD	LV (mg kg ⁻¹)
Cu	2.66	22.86	26.15	61.85	11.21	100
Zn	27.76	60.25	71.62	252.84	39.60	300
Pb	7.31	19.03	31.00	333.00	46.91	100
Cd	0.10	0.18	0.30	5.46	0.77	3.0
Ni	13.46	42.00	47.59	93.73	16.77	50
Cr	12.47	46.06	44.58	81.47	13.47	100
As	3.49	9.75	10.21	16.30	2.57	25
Hg	0.035	0.035	0.070	0.40	0.085	1.0

Descriptive statistics for heavy metals at 10 cm depth are presented in Table II, while corresponding values at 50 cm depth are shown in Table III. At both depths, Zn, Pb and Ni exhibit the highest mean content among the analysed elements, whereas Cd and Hg occur at the lowest content levels. At 10 cm depth, mean content decreases in the order: Zn (72.44 mg kg⁻¹) > Ni (50.09 mg kg⁻¹) ≈ Cr (48.79 mg kg⁻¹) > Pb (33.44 mg kg⁻¹) > Cu (27.15 mg kg⁻¹) > As (10.39 mg kg⁻¹) > Cd (0.22 mg kg⁻¹) > Hg (0.052 mg kg⁻¹). The highest maximum value is observed for Pb (317.49 mg kg⁻¹), followed by Zn (158.06 mg kg⁻¹). Standard deviation values indicate pronounced variability for Pb and Zn. At 50 cm depth, mean content follows a similar pattern: Zn (71.62 mg kg⁻¹) > Ni (47.59 mg kg⁻¹) > Cr (44.58 mg kg⁻¹) > Pb (31.00 mg kg⁻¹) > Cu (26.15 mg kg⁻¹) > As (10.21 mg kg⁻¹) > Cd (0.30 mg kg⁻¹) > Hg (0.070 mg kg⁻¹). Lead again shows the highest maximum value (333.00 mg kg⁻¹), while Cd displays a markedly elevated maximum (5.46 mg kg⁻¹) compared to surface soil. Variability, expressed as standard deviation, is highest for Pb and Zn at this depth as well. For most elements, mean values are higher than median values at both depths, particularly for Pb and Cd, whereas Ni and Cr show similarity between mean and median values. Comparison between depths indicates broadly similar median content for most elements, with slightly lower medians at 50 cm for Pb and Zn and comparable values for Ni and Cr. Mean Cd content is higher at 50 cm than at 10 cm, reflecting the influence of elevated maximum values in subsurface soils. All median content remains below the Serbian limit values at both depths. However, maximum content of Pb exceeds the limit value of 100 mg kg⁻¹ at both 10 and 50 cm, and Cd exceeds its limit value (3.0 mg kg⁻¹) at 50 cm depth. Nickel mean content approaches the limit value (50 mg kg⁻¹) at both depths. Since it occurs evenly and in deeper layers, this indicates that the elevated nickel content is primarily of natural (geogenic) origin, and not solely due to pollution. Thus, the high nickel content in central Serbia and Belgrade is a natural feature of serpentinite soils.²⁰ Contrary to expectations, recreational zones (like Čuburski Park) show higher copper and zinc pollution in deeper layers (50 cm) than on the surface. This is evidence of historical pollution – the use of

untested embankment material and rubble during the urbanization of Belgrade in the last century.²¹ In traffic zones (Zvezdara, bus stop Lasta), pollution is of a surface character (10 cm). A moderate positive correlation between lead (Pb) and zinc (Zn) across all 96 samples which is markedly stronger at the surface ($r = 0.64$ at 10 cm) than at depth ($r = 0.37$ at 50 cm), is consistent with exhaust gases and the fraying of car tires and brakes acting as shared, surface-concentrated sources of roadside contamination.^{22,23} Arsenic (As) is the highest in Čukarica, but its fate is directly related to the composition of the soil. A weak-to-moderate correlation between arsenic and clay content ($r = 0.25$ across all 96 samples) suggests that clay may contribute modestly to binding this element, though it is clearly not the dominant control on its distribution. Locations with higher clay content (ex. Čuburski park in Vračar, 24.6 % at the surface) still tend to show elevated metal levels, but this local pattern should be read as one contributing factor among several rather than a strong, standalone relationship, and does not by itself explain the higher accumulations observed at these sites. These metals have accumulated in urban soils over decades of continuous input.^{24,25} The average pH value of the soil is 8.17, which tells us that the soil is classified as alkaline. We know that in the alkaline environment, the mobility of heavy metals is reduced because they tend to form insoluble compounds (carbonates and hydroxides). In doing so, they lose their bioavailability, which means they find it harder to penetrate plants or groundwater despite their elevated content.^{26,27} Although the values of Nickel (Ni) and Chromium (Cr) are about or slightly higher than stricter environmental standards, it is important to note that lands in Serbia often have naturally elevated content of these metals due to geological substrates. However, the zinc and lead values in Čuburski Park at a depth of 50 cm deserve attention because they are significantly above the average of other sample. By analyzing the data obtained for organic-type pollutants, we come to the knowledge that urban soils are characterized by a moderate content of organic matter, which helps to contain organic pollutants. While BTEX compounds and PAHs occurred at low content indicative of diffuse urban sources, elevated C10-C40 hydrocarbons at both depths, particularly in subsurface soils, suggest legacy petroleum-derived contamination. The absence of detectable PCB contamination further indicates limited industrial impact, highlighting traffic-related and historical urban activities as the dominant sources of organic pollutants.^{28, 29, 30} The following table gives an analysis of organic pollutants by municipalities.

TABLE IV. Analysis of organic pollutants by municipalities

Municipality	Mineral oils (average mg kg ⁻¹)	Toluene (average mg kg ⁻¹)	Organic matter (%)
Stari grad	123.14	4.96	5.09
Vračar	80.70	2.68	5.05
Palilula	64.72	0.005	4.66
Savski venac	53.46	0.005	5.01
Novi Beograd	40.34	0.005	5.39
Zemun	33.92	0.005	7.01

The municipalities of Stari grad and Vračar are designated as "hot spots". The two central municipalities have by far the highest average content of both mineral oils and toluene. This was expected given traffic density, age of infrastructure, and historical urban load. Although the municipality of Zemun has relatively low mineral oil pollution, it has the highest percentage of organic matter in the soil (over 7 %). This suggests higher quality, "oily" soils, but also a potentially greater presence of humus in the zones sampled.³¹ Also, it is noted that the presence of toluene is practically negligible (at the level of the detection limit) in all municipalities except Stari Grad and Vračar. The municipality of Palilula occupies the third place in terms of mineral oil pollution, which may be related to industrial zones and specific locations along the Danube that this municipality encompasses.

Comparison with the legal framework of the Republic of Serbia reveals that the situation with soil pollution in the central parts of Belgrade is quite serious. The analysis was carried out according to the regulation on limit values of polluting, harmful and hazardous substances in soil (official gazette of RS, no. 30/2018 and 64/2019).¹⁷ The limit values (LV) are adjusted for each sample individually based on the measured percentage of organic matter (as required by law). Out of a total of 96 samples, as many as 81 samples (84 %) have a value of at least one parameter above the limit value (LV).

The law defines a limit value (LV) as a level indicating contamination. Below are examples where the values are many times greater than the allowed.

Although the excesses of toluene are numerically enormous (a thousand times greater than LV), the values are still far below the remedial value (RV) of about 130 mg kg⁻¹. This means that the soil is contaminated, but it is not at a level that requires immediate remediation, but intensified monitoring. Mineral oil is the most important parameter. Almost every sample in the center of Belgrade exceeds the limit considered "clean" land. Recreational zones, such as Čuburski park and Neimarski park, show systemic pollution from both petroleum products and volatile organic compounds (BTEX), which is likely due to decades of accumulation of exhaust gases and traffic surrounding these parks.

TABLE V. Municipalities with the highest deviations from the limit values for mineral oils and toluene

Location	Depth (cm)	Matter	Measured (mg kg ⁻¹)	Allowed (LV)	Exceedance index
Stari grad (Poenkareva)	50	Mineral oil	458.2	20.40	22 x more
Vračar (Čuburski park)	50	Mineral oil	172.2	36.95	4.6 x more
Vračar (Neimarski park)	50	Toluene	5.96	0.006	990 x more
Stari grad (Pristanište)	10	Toluene	10.20	0.005	2000 x more

Below are the analysis for the second group of parameters, which includes PCBs (polychlorinated biphenyls) and PAHs (polycyclic aromatic hydrocarbons - naphthalene and fluorene). This group of substances is specific because they are persistent organic pollutants that break down slowly and can be toxic even in small doses.

TABLE VI Average and maximum values for PCBs, naphthalene and fluorene given by municipalities

Municipality	PCB (mean, mg kg ⁻¹)	PCB (max, mg kg ⁻¹)	Naphthalene (mean, µg kg ⁻¹)	Naphthalene (max, µg kg ⁻¹)	Fluorene (mean, µg kg ⁻¹)	Fluorene (max, µg kg ⁻¹)
Palilula	0.011	0.024	0.011	0.020	0.010	0.010
Novi Beograd	0.010	0.010	0.014	0.060	0.018	0.110
Stari grad	0.010	0.010	0.030	0.050	0.010	0.010
Vračar	0.010	0.010	0.013	0.030	0.010	0.010
Zemun	0.010	0.010	0.011	0.020	0.013	0.040
Zvezdara	0.010	0.010	0.013	0.040	0.012	0.030

As for PCBs (polychlorinated biphenyls), a total of 5 limit values has been registered. The most critical location is Palilula (public fountain of Višnjica) at a depth of 50 cm, where 0.024 mg kg⁻¹ was measured (the limit is lower due to the lower percentage of organic matter at that depth). PCBs are lipophilic and more difficult to bind to organic matter. In soils with less humus (as is the case here) they migrate more easily to deeper layers and accumulate there for decades. It is interesting that mild overflows also occur in the zones of water supply sources (Makiš, Reni bunar).^{32,33} Although PAHs (naphthalene and fluorene) are detected in most samples, their values are generally below legal limits. Nevertheless, the Stari grad leads with an average of 0.030 µg kg⁻¹ of naphthalene, which is almost 3-4 times more than in other municipalities. In Novi Beograd, the maximum value of fluorene (0.110 µg kg⁻¹) was recorded, which is an isolated case of pollution, probably related to the immediate vicinity of a road or industrial outlet.³⁴ Due to the presence of specific toxicants, the following locations require attention and: 1. Palilula-Višnjica (public fountain) because it has the highest content of PCBs.; 2. The Stari grad-Kalemegdan (Topolivnica) has an elevated level of naphthalene.;

3. Novi Beograd - Reni bunar (block 70a) has been recorded the presence of PCBs in the deeper layer of soil (50 cm).; 4. Čukarica-Makiš (deposition) PCBs were detected at both depths (10 cm and 50 cm). While mineral oils and toluene are a "chronic disease" of Belgrade land caused by traffic, PCBs appear localized, often along riverbanks or specific technical facilities (such as deposits and wells). Fortunately, the levels of PAHs (naphthalene, fluorene) are currently within safe limits for most of the city.²⁹ In the entire data set, only at one location was clearly detected a value above the sensitivity limit (0.01 mg kg^{-1}). This is the municipality of Palilula, measuring place of public fountain Višnjica, where 0.024 mg kg^{-1} PCB was measured at a depth of 50 cm. This value is significant because it exceeds the standard limit of 0.02 mg kg^{-1} , which at this depth and type of site (headwaters) requires added attention. For the other 95 samples, the result was $< 0.01 \text{ mg kg}^{-1}$ (below the detection limit). However, the law in Serbia stipulates that the limit value (LV) is reduced if the soil has little organic matter. At the following locations the allowed limit is so low (below 0.01 mg kg^{-1}) that a laboratory finding "less than 0.01" does not actually guarantee that the land is completely clean under the law: * Čukarica-Makiš (depository) (10 cm and 50 cm) * Novi Beograd-Reni bunar (block 70a) (50 cm) * Stari grad-Kalemegdan (Topolivnica) (10 cm).³⁵ Analysis of benzene pollution (a volatile organic compound that is often an indicator of pollution from petroleum products or fuel combustion) brings good news compared to other parameters we have analyzed so far.³⁶

TABLE VII Average levels and maximum values for benzene given by municipalities

Municipality	Average level of benzene ($\mu\text{g kg}^{-1}$)	Maximum value of benzene ($\mu\text{g kg}^{-1}$)
Vračar	0.124	0.48
Stari grad	0.109	0.29
Palilula	0.103	0.38
Zemun	0.055	0.54
Zvezdara	0.054	0.33
Savski venac	0.033	0.11

Thus, central metropolitan municipalities (Vračar, Stari grad) again show slightly higher levels, which is a direct correlation with traffic density and the "canyon" effect of streets in the center of Belgrade where exhaust gases are retained for longer.³⁶

Looking at the above data, we note the correlation between benzene and mineral oils. In locations where mineral oils are elevated (such as busy roads and recreational areas in the city center), these small but measurable amounts of benzene have also been detected. Fortunately, benzene is highly volatile and subject to biodegradation, so it does not accumulate in soils in such alarming amounts as heavier mineral oils or PCBs.

The data shows a clear difference in the " behavior " of different pollutants depending on the depth.

TABLE VIII: Behavior of different pollutants at different research depths

Parameter	Surface (10 cm)	Depth (50 cm)	Trend
Organic matter (%)	5.58%	4.66%	Expected higher at the surface (humus)
Mineral oils (mg kg ⁻¹)	46.57	59.40	Grow with depth
Toluene (mg kg ⁻¹)	0.84	0.56	Decreases with depth
Benzene (µg kg ⁻¹)	0.055	0.057	Almost identical (high mobility)

Analyzing aforementioned data, we come to the conclusion that: a) mineral oils accumulate in deeper layers (50 cm), which may mean that the pollution is of an older date and has had time to penetrate deeper or that it is more easily washed down from the surface, and b) that toluene is more present on the surface (10 cm), indicating fresher pollution, probably directly from the atmosphere or from vehicle exhaust deposited on the surface of the soil.

Based on a comprehensive analysis of all parameters (mineral oils, BTEX compounds, PCB and PAH), I have singled out the 5 most critical points in Belgrade. The ranking was carried out by combining the level of toxicity of substances and the degree of exceeding the legal limit values (LV).³⁷

In the next table there is a list of 5 municipalities with the highest content of organic pollutants.

TABLE IX: The most critical points with the highest content of organic pollutants

Location	Main pollutant	Pollution intensity
Stari grad- Poenkareva/V.Dobrnjca (50 cm)	Mineral oils	458.2 mg kg⁻¹ (22 times above the limit)
Stari grad - Pristanište (10 cm)	Toluene	10.20 mg kg⁻¹ (maximum measured value)
Palilula - Višnjica (public fountain 50 cm)	PCB	0.024 mg kg⁻¹ (only confirmed PCB pollution)
Vračar - Čuburski park (50 cm)	Mixed (Mineral oils+Toluene)	Mineral oils: 172.2 mg kg⁻¹ + high content of toluene
Vračar - Neimarski park (50 cm)	Toluene + Mineral oils	Toluene: 5.96 mg kg⁻¹ + Oils: 122.3 mg kg⁻¹

We see that the Stari grad and Vračar occupy 4 out of 5 places on the list. This confirms that the urban zone is the most endangered part of Belgrade when it comes to organic pollutants in the soil. In 4 of the 5 most important samples, environmental pollution is significantly higher by 50 cm than on the surface. This suggests that pollution is not cleaned naturally but penetrates deep and accumulates there for decades. Although mineral oils have higher numbers, the location in Višnjica is alarming because it is a source of water supply and extremely persistent poison (PCB).³⁸

Multivariate statistical analysis (Principal Component Analysis)

To explore the joint structure of the contaminant dataset, Principal Component Analysis (PCA) was performed in two complementary steps using Python (scikit-learn). All variables were z-score standardized prior to analysis given their very different measurement scales and units (mg kg^{-1} vs. $\mu\text{g kg}^{-1}$ vs. %). The first analysis used the full-resolution dataset of all 96 individual soil samples, including the eight heavy metals (Cu, Zn, Pb, Cd, Ni, Cr, As, Hg) together with organic matter, clay content, moisture and pH. The second analysis addressed the relationship between heavy metals and the organic pollutants (mineral oil, toluene, PCB, naphthalene, fluorene and benzene) specifically.

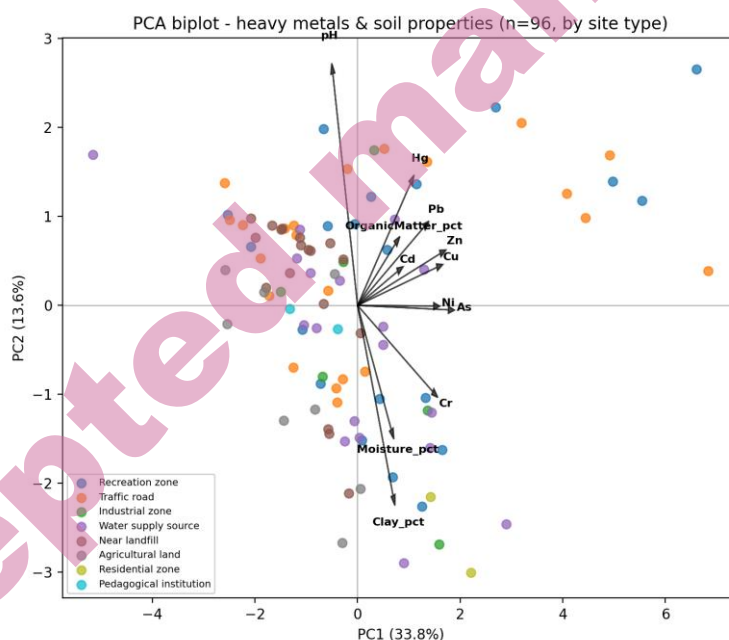


Figure 1. PCA biplot of heavy metals and soil physicochemical properties across all 96 individual soil samples, colored by site type. PC1 (33.8% of variance) reflects overall metal enrichment; PC2 (13.6%) reflects soil pH/texture.

It is confirmed directly by the metal–organic cross-correlation matrix (Table X): mineral oil and toluene correlate strongly with Zn ($r = 0.82$ and 0.89), Cu ($r = 0.81$ and 0.78), As ($r = 0.57$ and 0.63) and Hg ($r = 0.51$ and 0.61), while Ni, Cd and Cr — the elements most strongly tied to geogenic serpentinite sources^{11,12,14}, show weak or near-zero correlation with every organic pollutant ($|r| \leq 0.3$ in all cases). PCB behaves differently again, correlating weakly and even negatively with most metals (e.g. $r = -0.20$ with Zn, $r = -0.54$ with Cr), consistent with its

previously noted localized, riverbank/facility-linked occurrence^{32,33} rather than a shared traffic source.

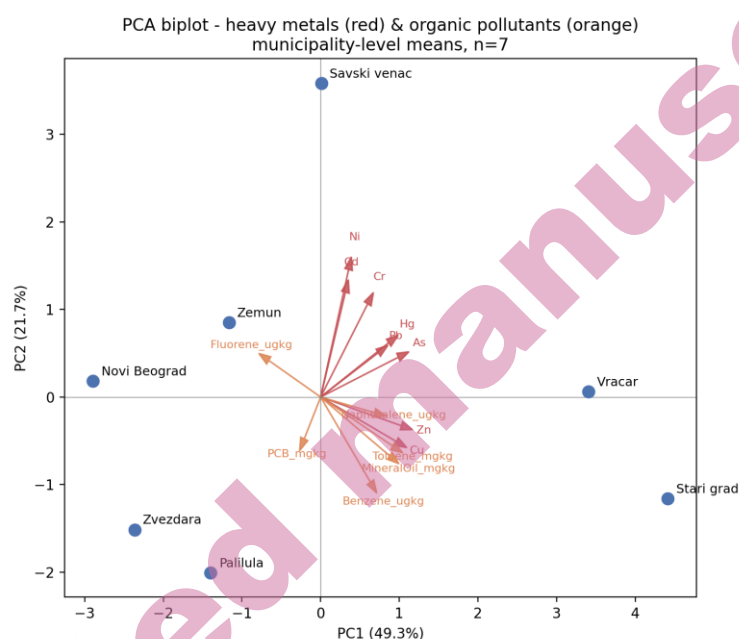


Figure 2. PCA biplot of heavy metals (red) and organic pollutants (orange) using municipality-level mean values ($n = 7$). PC1 (49.3%) captures the shared traffic-derived contamination gradient; PC2 (21.7%) separates PCB/fluorene behavior from the traffic-linked variables.

TABLE X. Pearson correlation coefficients (r) between heavy metals and organic pollutants, municipality-level means ($n = 7$)

Metal	Mineral oil	Toluene	PCB	Naphthalene	Fluorene	Benzene
Cu	0.81	0.78	0.09	0.46	-0.80	0.79
Zn	0.82	0.89	-0.20	0.66	-0.57	0.75
Pb	0.21	0.33	-0.21	0.01	-0.49	0.36
Cd	0.08	-0.05	0.09	0.15	-0.28	-0.34
Ni	-0.03	-0.08	-0.09	0.13	-0.06	-0.28
Cr	0.10	0.27	-0.54	0.33	0.16	0.12
As	0.57	0.63	-0.23	0.41	-0.60	0.44
Hg	0.51	0.61	-0.40	0.67	-0.41	0.09

Taken together, the two analyses converge on the same conclusion reached qualitatively earlier in this manuscript: Cu, Zn, As and Hg pattern together with the petroleum-derived organic pollutants (mineral oil, toluene, benzene, naphthalene) and are best explained by a common traffic/urban source, whereas Ni

and Cr remain statistically distinct, supporting their geogenic origin.^{11,12,14} These results should be read as exploratory rather than confirmatory: the municipality-level analysis ($n = 7$ municipalities, 14 variables) has more variables than observations, it was included because it is the only resolution at which heavy metals and organic pollutants can currently be compared directly in this dataset. The full-resolution analysis ($n = 96$) is statistically more robust and is offered as the primary, confirmatory result. Future sampling campaigns that measure organic pollutants at the individual-sample level, matched to the existing heavy-metal dataset, would allow this correlation structure to be tested with full statistical power.

Geochemical processes controlling metal distribution

To identify which geochemical mechanisms govern the distribution of individual metals, Pearson correlations were calculated between each of the eight heavy metals and the four measured soil properties (pH, clay content, organic matter content and moisture) across all 96 samples.

The geochemical basis for treating Ni and Cr as geogenic rather than anthropogenic in this dataset rests on well-established mineralogical mechanisms governing the formation and weathering of ultramafic (serpentine) parent rock. Fresh ultramafic rock is composed principally of olivine and pyroxene, in which Ni^{2+} substitutes readily for Mg^{2+} and Fe^{2+} within the crystal lattice owing to their similar ionic radii; upon hydrothermal alteration (serpentinization), these primary minerals are converted to serpentine-group phyllosilicates (lizardite, chrysotile, antigorite) that retain Ni within their octahedral sheet and, together with associated chlorite-group minerals, host the majority of Ni present in serpentine soils.³⁹ Chromium follows a related but distinct pathway, being concentrated chiefly in chromite and in serpentine formed from pyroxene rather than olivine. This mineralogical partitioning explains why Ni and Cr in serpentine-derived soils occur predominantly in the residual, structurally-bound fraction rather than in exchangeable or mobile forms, consistent with the sequential-extraction evidence already reported for serpentine soils^{11,12} in Serbia and for Belgrade urban soils specifically.²¹ The average Ni content of the Earth's upper continental crust is approximately 34 mg kg^{-1} , whereas ultramafic rocks such as serpentine and peridotite can contain several thousand mg kg^{-1} . The sampled soils have no comparable geological reservoir for Cu, Zn, Pb, As or Hg: the Belgrade area is predominantly underlain by Quaternary alluvial, proluvial and loess-like sediments overlying Pannonian marls and silts,^{40,41} none of which are naturally enriched in these elements, reinforcing why their correlation with traffic-derived organic pollutants rather than with local geology is the more parsimonious explanation for their distribution.

Chromium and arsenic are the only metals showing a clear pH dependence, and in both cases the relationship is negative ($r = -0.30$ and -0.28 , respectively):

content tends to be higher under more acidic conditions, opposite to the pattern expected from simple carbonate/hydroxide precipitation.^{26,27} This is consistent with the amphoteric, oxyanion-forming behavior of arsenic, whose sorption onto mineral surfaces typically decreases as pH rises, rather than increases as it does for simple cationic metals. Chromium also shows the strongest clay association in the dataset ($r = 0.38$), a real but moderate effect rather than a dominant control. Organic matter exerts a mild, fairly uniform positive pull on copper, zinc and lead ($r = 0.27$ – 0.31), consistent with organic-matter complexation contributing to, but not dominating, their retention.^{4,5} Nickel and mercury show essentially no relationship with any of the four soil properties measured ($|r| \leq 0.17$), indicating that their spatial variability is controlled mainly by source strength and location rather than by local soil chemistry — consistent with the geogenic origin already established for nickel.^{11,12}

The depth-accumulation pattern shows that only cadmium and mercury genuinely accumulate at depth, with 50 cm/10 cm content ratios of 1.40 and 1.36 respectively; every other metal is essentially flat with depth or slightly enriched at the surface (ratios of 0.91–0.99 for Cr, Pb, Ni, Cu, As and Zn). This is a more limited pattern of vertical migration than observed for the organic pollutants, where mineral oils showed pronounced accumulation at 50 cm,^{28,29,30} suggesting that most of the analyzed metals are retained close to their point of surface deposition under the current soil conditions, while cadmium and mercury — both capable of forming more mobile organo-metallic or chloride complexes — show greater potential for downward migration over time.

Vertical distribution of pollutants

The heavy metals and organic pollutants examined in this study show two distinct types of vertical behavior. Ni and Cr, inherited from the serpentinite parent material, are essentially uniform with depth or slightly lower at 50 cm, while Pb, Cu, Zn and As — despite their anthropogenic, traffic-linked origin — are likewise retained close to the surface once deposited, rather than leaching downward. This limited mobility is consistent with international findings that under neutral-to-alkaline soil conditions, trace metals are progressively converted to residual and oxide-bound fractions that resist vertical transport,^{15,26,27} including the comparative Salzburg–Thessaloniki–Belgrade study, which found PTEs in urban park soils predominantly in the geochemically stable residual fraction rather than the exchangeable, mobile fraction.¹⁵ Cadmium and mercury are the exception, accumulating at depth by 36–40%, consistent with their known capacity to form more mobile chloride and organic complexes even where other metals remain immobile.

The organic pollutants show a sharper, and mechanistically distinct, depth split that maps directly onto volatility and lipophilicity rather than onto soil pH. Mineral oils (C10–C40) increase substantially with depth, matching soil-column

evidence that heavier, less volatile and less biodegradable petroleum fractions migrate downward over time while lighter fractions attenuate near the surface.^{28,29,30} Toluene shows the opposite trend, decreasing with depth as expected for a volatile, readily biodegradable compound that is lost from the surface before it can migrate, while benzene — more soluble and mobile still — shows almost no depth gradient at all, consistent with international soil guideline assessments of its high leaching potential.³⁶ PCBs follow a third pattern: although strongly lipophilic, they migrate into deeper layers specifically where soil organic matter is insufficient to retain them, in agreement with recent findings that soil organic carbon content, rather than depth itself, is the dominant control on PCB vertical mobility.^{32,33}

Taken together, the comparison indicates that vertical mobility in this dataset is governed less by whether a pollutant is geogenic or anthropogenic in origin, and more by its physicochemical behavior under local soil conditions: alkaline pH and clay content restrict metal leaching regardless of source, while for organic pollutants volatility, biodegradability and the availability of organic matter for sorption determine whether a compound stays near the surface, migrates to depth, or equilibrates throughout the profile. This is consistent with the wider international literature on urban soils.

In the study Jelić *et al* (2025), while studying concentrations of heavy metals in the city soil of Novi Sad, was found a distinct clustering of Pb and Zn separate from the other measured metals, interpreted as evidence of a predominantly anthropogenic, traffic-linked origin⁴² — essentially the same Pb–Zn grouping identified in the Belgrade dataset. A comparable dual-origin pattern was reported for Debrecen, Hungary, where Cr and Ni were attributed to natural geogenic sources while As, Co, Pb and Zn showed strong anthropogenic signatures linked to traffic, urban waste and construction materials, with Cd reflecting a mixed lithogenic and point-source origin⁴³ — a split that maps almost exactly onto the Ni/Cr versus Cu/Zn/Pb/As/Hg division established for Belgrade in this manuscript. The three-city sequential-extraction comparison of Salzburg, Thessaloniki and Belgrade reached the same conclusion by a different method, finding Al, Cr and Ni predominantly in the geochemically stable residual fraction (lithogenic) across all three cities, while Cu, Pb and Zn were consistently associated with the more mobile, anthropogenically influenced fractions.¹⁵

Elsewhere in Serbia the picture is broadly consistent but shows useful local variation. In Vojvodina, Ni and Co were again found to be predominantly natural, but Cr showed a more mixed natural/anthropogenic signature depending on proximity to industrial and agricultural activity¹⁴, and a 2026 study of Kragujevac similarly found that while As, Co and Mn reflected the geochemical background, Cr — treated as fully geogenic in the present Belgrade dataset — appeared partly anthropogenically influenced there, alongside Cu.⁴⁴ This is a useful reminder that the geogenic/anthropogenic split for a given element is not universal but depends

on the specific mix of local geology and industrial history. The study Miletić *et al* (2025), an assessment of agricultural and urban soils near Serbia's largest oil refinery found pronounced anthropogenic hotspots for Pb, As, Hg, Cd and Zn attributable to industrial emissions⁴⁵, and chemical fractionation across several industrialised Serbian cities confirmed elevated but still low-risk Cu, Pb and Zn from traffic and industry alongside geogenic Ni.⁴⁶

CONCLUSION

This study provides an integrated geochemical evaluation of heavy metals and selected organic pollutants in urban soils of Belgrade, based on 96 samples collected from representative land use categories at two depths. Multivariate statistical analysis enabled clear separation between geogenic influences (Ni, Cr), linked to Serbia's serpentinite geology, and anthropogenic inputs associated with traffic and industrial activity (Cu, Zn, Pb, As, Hg; BTEX, PAHs, PCBs, mineral oils). Zn, Pb and Ni exhibited the highest mean content, while Cd and Hg occurred at the lowest levels. Pb and Cd exceeded Serbian limit values at several locations, whereas elevated Ni and Cr primarily reflected natural geological enrichment. The alkaline soil conditions (mean pH $\approx$ 8.2) suggest reduced mobility of most metals, consistent with limited depth variation. Organic pollutants showed surface enrichment and co-variation with traffic related metals. Although most median values remained below regulatory thresholds, localized exceedances indicate the need for continued monitoring. Any assessment of potential implications for groundwater or human exposure requires dedicated risk assessment studies beyond the scope of this work.

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ИЗВОД

ГЕОХЕМИЈСКА ПРОЦЕНА ХЕМИЈСКИХ ЗАГАЂИВАЧА У ЗЕМЉИШТУ БЕОГРАДА, СРБИЈА: СТУДИЈА ГЕОГЕНИХ И АНТРОПОГЕНИХ УТИЦАЈА

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Ова студија пружа интегрисану геохемијску процену хемијских загађивача у профилима урбаног земљишта (0–10 cm и 40–50 cm) прикупљеним широм Београда, у Србији, током 2023. године. Агенција за заштиту животне средине Републике Србије (СЕРА) прикупила је деведесет шест узорака из девет општина – укључујући Стари град, Палилулу, Земун, Врачар, Чукарицу, Нови Београд, Раковицу, Вождовац и Сурчин – који покривају рекреативне зоне, главне путне коридоре, индустријске зоне, пољопривредне парцеле и локације у близини депонија и водоводних објеката. Концентрације осам тешких метала

(Cu, Zn, Pb, Cd, Ni, Cr, As, Hg) и одабраних органских загађивача (BTEX, PAH, PCB, фракције минералних уља) процењене су у односу на националне граничне вредности. Већина анализата је остала испод регулаторних прагова, али је дошло до значајних прекорачења: Pb је достигао 333 mg kg⁻¹ у Земуну, а Cd 5,46 mg kg⁻¹ у Сурчину (оба на 50 cm), док су минерална уља достигла врхунац од 458 mg kg⁻¹ у Старом граду, а PCB-ови су прешли границе на водоводу Палилула. Обрасци зависни од дубине указивали су на ограничену мобилност за већину метала, при чему Cd и Hg показују обогаћење на 50 cm. Мултиваријантна анализа је разликовала антропогену, саобраћајно повезану групу (Cu, Zn, Pb, As, Hg; BTEX, минерална уља) од геогене Ni-Cr групе повезане са земљиштима изведеним из серпентинита у Србији.

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