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Environmental impacts of microplastics in contaminated urban soils in Serbia: potential implications for Cu, Mn, and Sr phytoremediation

Ivana Mikavica*[,1], Dragana Ranđelović[1], Miloš Ilić[2], Dušan Milojkov[1], Aleksandar Jovanović[1], Milica Mišić[1], Jelena Mutić[2]

[1]Institute for Technology of Nuclear and other Mineral Raw Materials, Boulevard Franchet d`Esperey 86, Belgrade, Serbia
[2]University of Belgrade-Faculty of Chemistry, Studentski trg 12 - 16, P. O. Box 51, 11158, Belgrade, Serbia

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A B S T R A C T

Microplastics (MPs) are widespread environmental pollutants and have emerged as a growing global concern. In soil ecosystems, MPs frequently coexist with potentially toxic elements (PTEs), yet their combined effects on soil–plant interactions and phytoremediation processes remain insufficiently explored. This field-based study investigated the occurrence of MPs in urban soils from four Serbian cities and assessed their influence on the uptake of PTEs - copper (Cu), manganese (Mn), and strontium (Sr) - by Capsella bursa-pastoris (L.) Medik. MPs were extracted from soil using an optimized density separation method, while total (aqua regia) and phytoavailable (EDTA-extractable) fractions of PTEs were quantified in both soils and plant tissues. Maximum MPs abundance was recorded in Bor — 500 ± 100 MPs kg-1.The highest total concentrations of Cu (516.14 µg g^{-1}), Mn (553.46 µg g^{-1}), and Sr (173.69 µg g^{-1}) were detected in soils from Bor. The geoaccumulation index (Igeo) indicated moderate to heavy contamination levels. CuEDTA accounted for UP to 50.7% of CuAR, MnEDTA for 34.4% of MnAR, and SrEDTA 27.3% of SrAR. After the uptake, C. bursa-pastoris primarily translocated the elements to the aerial parts, indicating shoot accumulation as the dominant strategy. Principal component analysis (PCA) revealed distinct clustering of samples by city, while Spearman correlation analysis highlighted significant associations between MPs and PTEs mobility in the soil-plant system. Strongest correlations were found between MPs phytoavailable Cu fraction (ρ =+0.49) and Cu content in shoots (ρ =+0.56). The highest BCF values were determined for Sr, ranging from 2,40 (SM) to 5,41 (BO). PTEs were mainly transferred to the shoots. TF range for Cu was 0.54 (BO) – 1.48 (VR), 0,68 (BO) to 1,42 (VR) for Mn, and 0,76 (BO) to 1,34 (VR) for Sr. Strong correlations among MPs abundance and Cu mobility and accumulation in shoots (ρ = +0.56), and Sr bioaccumulation potential (BCF up to 5.41), highlight the role of MPs in modifying element transfer within urban soil–plant systems and consequent phytoremediation potential.

Keywords: urban soils, *Igeo* index, phytoaccumulation, correlations.

1. Introduction

Although only 9% of plastic waste is recycled, the usage of disposable plastics is still extensively expanding (Duan et al. 2024). Plastic waste enters the environment usually without proper treatment and undergoes different degradation processes. Nearly 80% of disposed plastic waste defragments into MPs, plastic particles smaller than 5 mm (Wang et al. 2024). MPs have already been recognized worldwide as an emerging pollutant and omnipresent environmental threat (UNEP, 2021). Studies have revealed that terrestrial environments are long-term sinks of MPs, highly vulnerable to MPs contamination (Saha et al. 2024). Soil ecosystems are facing significant degradation since MPs may disrupt their structure and deteriorate soil quality, posing a risk to soil biota and human health (Wang et al. 2024). Thus, further investigations on environmental effects of soil MPs are urgently needed.

MPs are not only a hazard for the soil, but also the carriers of diverse soil pollutants (Duan et al. 2024). Meanwhile, the discharge of industrial waste and parent material weathering produce an excess of PTEs in soils. As a result, MPs and PTEs co-occurrence in soils is common. Studies revealed that MPs could directly or indirectly change soil chemistry, inducing PTEs' chemical redistribution (Jiang et al. 2024). Owing to the high specific surface area, active binding sites on MPs may interact with PTEs, promoting transformation from residual

* Corresponding author.
E-mail address: i.mikavica@itnms.ac.rs (Ivana Mikavica).

to available states (Chen et al. 2024). Even though the degree of the influence depends on MPs concentration and type, the combined MPs-PTEs effects are intensified compared to either pollutant alone (Jiang et al. 2024). Moreover, improved PTEs mobility could lead to promoted uptake by plants depending on the species under consideration (Chen et al. 2024). The responses of the soil-plant systems to MPs and co-existing PTEs are irregular and still underexplored.

So far, different crop species have been investigated as model plants for estimating MPs' influence on PTEs mobility (Wang et al. 2024), while species used for ecological remediation have been neglected. Chen et al. (2024) explored MPs' influence on phytoremediation of heavy metal–contaminated soils by hyper-accumulator plant ramie (*Boehmeria nivea* L.). They revealed that polystyrene (PS) MPs could improve Pb availability and promote the efficiency of plant uptake. There is a lack of studies concerning the interaction among MPs and species capable of accumulating high amounts of PTEs and indicating environmental pollution. *Capsella bursa-pastoris* (L.) Medik is a terrestrial plant widely distributed in areas with high PTEs loads (Aksoy et al. 1999). On the other side, MPs are prevalent in sites enriched with PTEs. Therefore, the high accumulation capacity suggests *C. bursa-pastoris* as a promising species for assessing the interconnection among environmental MPs and PTEs mobility in soil-plant systems.

The present study aimed to address knowledge gap about how soil MPs affects remediation of contaminated sites by assessing Cu, Mn and Sr availability in soils from four urban areas in Serbia and uptake by *C. bursa-pastoris* in the presence of environmental MPs. The main objectives were to 1) analyze Cu, Mn and Sr total and bioavailable content in urban soils, 2) determine their concentrations in roots and shoots of *C. bursa-pastoris*, and 3) examine correlations among soil MPs and PTEs mobility in soil-plant system. These findings will enhance our understanding of MPs-PTEs interference in soil and provide insights into MPs potential effects on PTEs phytoremediation from contaminated soils.

2. Materials and methods

2.1. Sampling sites

Sampling encompassed suburban/urban areas in four cities in Serbia (Figure 1): Belgrade (44°49'14"N, 20°27'44"E), as the most populated city with heavy traffic and industrial activities, Sremska Mitrovica (44°58′20″N, 19°36′33″E), located close to the Zasavica nature reserve, Vršac (45°07′00″N, 21°18′08″E), known for pharmaceutical production complex and Bor (44°04′25″N, 22°05′26″E), recognized by extensive copper mining activities.

2.2. Sample preparation and PTEs determination

Soils were sampled from the top soil layer (20 cm of depth), nearby the roots of *C. bursa-pastoris*. Samples were air-dried at room temperature for two weeks. Soil fraction <2 mm was further analyzed for the pseudo-total and phytoavailable PTEs content. Pseudo-total concentrations in soil were determined via *aqua regia* digestion, following USEPA method 3050. Phytoavailable soil fraction was assessed by single extraction with 0.05 M EDTA solution. Collected plant material was thoroughly washed with tap and distilled water, air-dried and separated to roots and shoots for the elemental analysis. Ground plant samples were subjected to microwave digestion using concentrated HNO_3 and 30% H_2O_2. Microwave oven used for digestion was Ethos 1, Advanced Microwave Digestion System; Milestone, Italy. PTEs content in soil and plant samples was analyzed in three replicates and determined by inductively coupled plasma optical emission spectroscopy (ICP-OES) – model iCAP 6500 (Thermo Fischer Scientific, USA).

2.3. MPs isolation from soils

MPs in soils was isolated using an optimized density separation method. A polarizing microscope (Carl Zeiss Jena Pol-U) was used for MPs quantification. Isolated particles were characterized by Fourier-transform infrared (FTIR) spectroscopy using a Thermo Scientific Nicolet iS50 spectrometer (Thermo Fisher Scientific, Waltham, MA, USA).

2.4. **Data analysis**

Bioconcentration factor (BCF) was expressed as the ratio between PTEs contents in underground parts and the corresponding pseudo-total concentration in soil. The translocation factor (TF) was calculated as the ratio among the PTEs content in aboveground and underground parts. The geo-accumulation index (I_{geo}) (Müller, 1969) was defined as:

$$I_{geo} = log_2\left(\frac{C_n}{1.5\ x\ BC_n}\right) \quad (1)$$

where C_n is the PTEs measured concentration "n" in the soil, while BC_n is the corresponding PTEs background concentration (Alloway, 1995). Kruskal Wallis test, correlation analysis and principal component analysis (PCA) were performed in Statistica Statsoft package (2007).

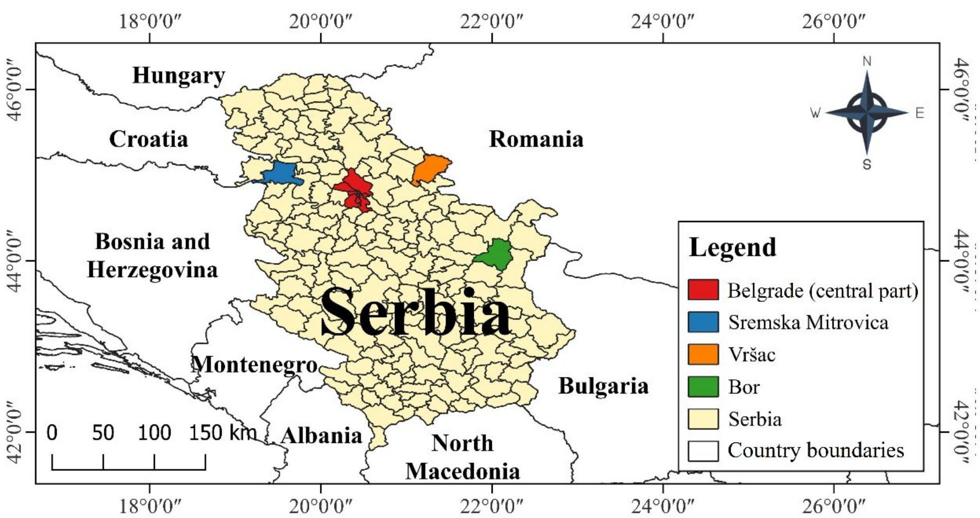


Fig. 1. Sampling sites in Serbia: Belgrade (BG), Sremska Mitrovica (SM), Vršac (VR), and Bor (BO)

3. Results and discussion

3.1. Cu, Mn and Sr content in soils

The soil from Bor exhibited the highest levels of Cu (516,14 µg g^{-1}), Mn (553,46 µg g^{-1}) and Sr (173,69 µg g^{-1}) (Figure 2), exceeding the average values for world soils (Kabata Pendias, 2011). Beside natural enrichment, extreme Cu concentrations in samples from Bor also have an anthropogenic origin, such as atmospheric deposition from the former city smelter, industrial emissions, and surrounding mine waste deposits. Total Cu content (Cu_{AR}) in the other three sites was multiple times lower, ranging between 17,08 µg g^{-1} (BG) and 27,34 µg g^{-1} (SM). In locations other than Bor, Mn abundance in soils varied from 306,41 µg g^{-1} (BG) to 485,53 µg g^{-1} (VR), while the Sr level range was 22,94 (VR) – 117,92 (SR). Overall, PTEs contents varied significantly between sampling sites (Kruskal Wallis test, $p<0.05$). The phytoavailable fraction followed the same trend. Around 50% of the total Cu content in soil was EDTA extractable (Cu_{EDTA}), i.e., phytoavailable. Although Mn is the Earth's crust's second most abundant element and considered relatively immobile, nearly 35% of the total content was available. On the other side, Sr can be easily released during soil weathering (Kabata Pendias, 2011), while around 27% of Sr was determined in the available pool (Sr_{EDTA}). I_{geo} index for Cu in soils from city of Bor ranged from 4,25 to 5,26, (the fifth and sixth categories) signifying strong to extreme soil contamination, according to Müller (1969) (Figure 1). Moreover, I_{geo} for Sr was between 2,80 to 3,06, classifying this area as contamination grade 2 and 3.

Pollution index mirrored an exceedance of remediation values noticed in PTEs concentrations in soil (Official Gazette of Republic of Serbia, 2018). Obtained values indicate that basic soil functions are disrupted and soil recovery and remediation are required.

3.2. Cu, Mn and Sr content in C. bursa-pastoris

PTEs concentrations in plant mainly followed the general patterns noticed in soils. BO samples contained the highest levels of Cu (24,85 µg g^{-1} and 13,37 µg g^{-1} for roots and shoots, respectively) and Sr (67,12 µg g^{-1} in roots and 50,65 µg g^{-1} in shoots). Mn concentrations in under and aboveground parts were the highest in samples from Belgrade, where the Mn was the highest in the phytoavailable pool. These results suggest that greater plant contamination was recorded in larger, industrially more developed sites with increased urbanization. *C. bursa-pastoris* biomonitoring capabilities were previously tested in the city of Bradford, UK, where around 16 µg g^{-1} of Cu was found in washed leaves, which is accordant to our results (Aksoy et al. 1999). In another study, *C. bursa-pastoris* accumulated 95,58 µg g^{-1} of Cu, and showed the best bioaccumulation capacity among other examined species from Brassicaceae family (Tuna et al. 2022). However, the highest BCF values were determined for Sr. BCF ranged from 2,40 (SM) to 5,41 (BO). After the absorption from soil, PTEs were mainly transferred to the shoots. TF range for Cu was 0.54 (BO) – 1.48 (VR), 0,68 (BO) to 1,42 (VR) for Mn, and 0,76 (BO) to 1,34 (VR) for Sr.

3.3. MPs-PTEs associated effects on the soil-plant system

Spearman correlation analysis signified the relationship between soil MPs and PTEs content in the soil-plant system. Positive correlations were identified among MPs and Cu phytoavailable ($\rho=0.49$) and total content ($\rho=0.56$). Earlier studies have already revealed that MPs have promoting effects on PTE's mobility. For instance, polystyrene (PS) MPs could cause an increase in Cu mobility in soil by "dilution effects". MPs may block the active sites on soil and stimulate the desorption of PTEs, thus increasing their leachability (Peng et al. 2024). Moreover, in our study MPs abundance positively correlated with Cu content in shoots of *C. bursa-pastoris* (0,51), which, on the other side, showed positive correlations with Cu_{EDTA} (0,55).

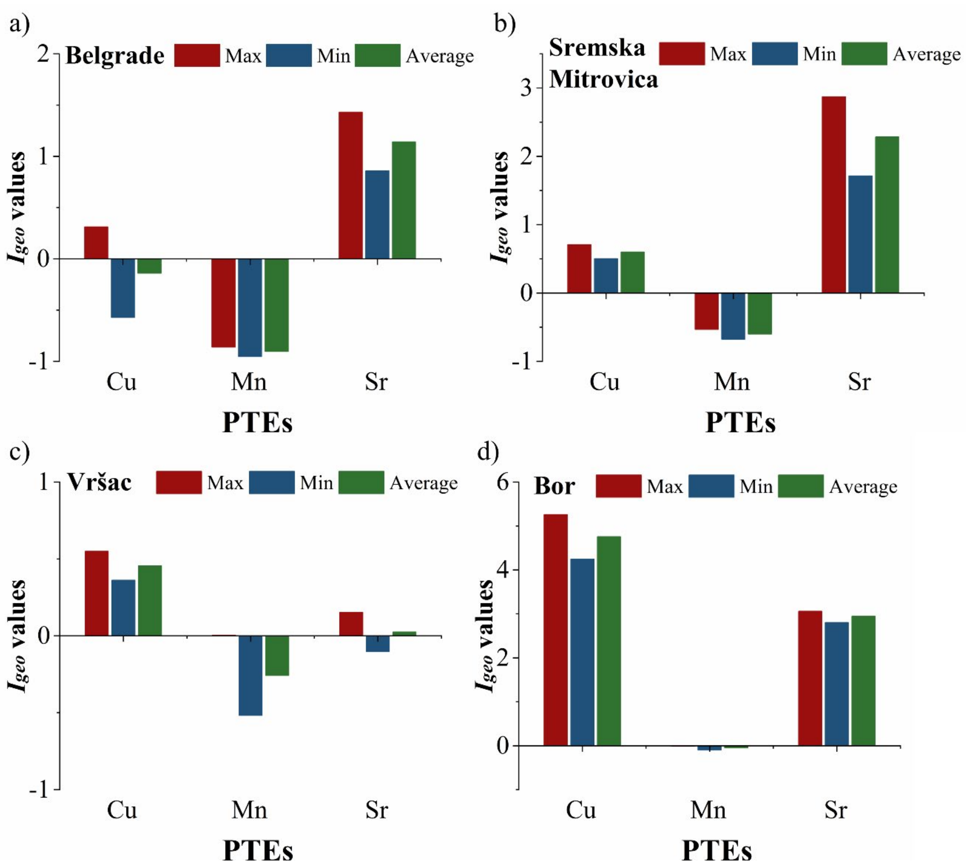
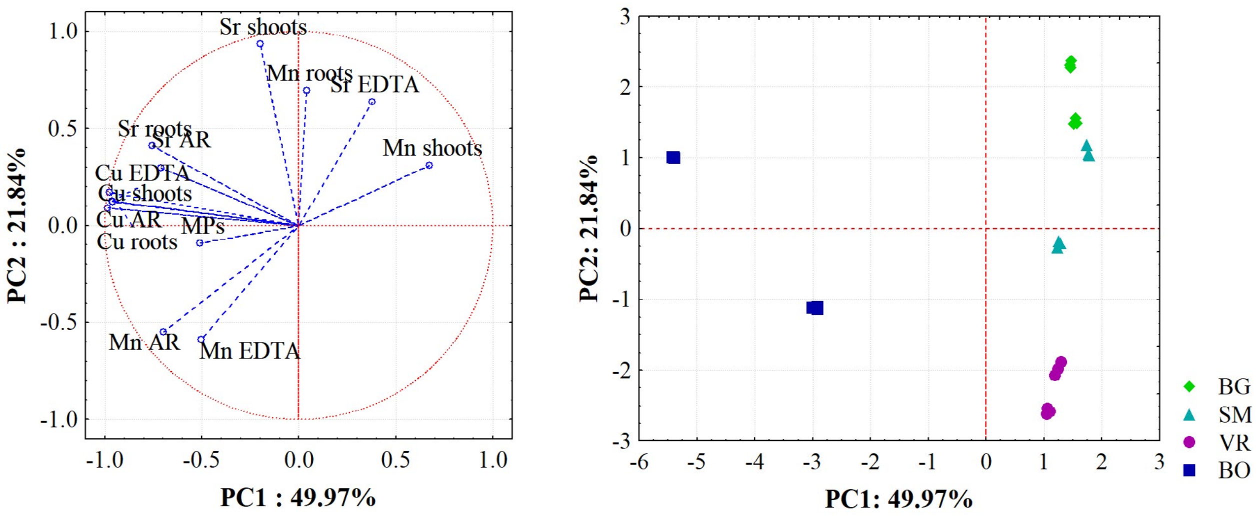


Fig. 2. Geoaccumulation index (I_{geo}) for analyzed soils from a) Belgrade, b) Sremska Mitrovica, c) Vršac, and d) Bor



a) b)

Fig. 3. Principal Component Analysis a) loading plot, b) score plot

PCA analysis revealed the distinction between samples from different sites based on analyzed parameters. MPs, Cu_{total}, Cu_{EDTA}, Cu_{roots}, and Cu_{shoots} influenced the separation of BO scores from other sites along PC1 axes (Figure 2). Meanwhile, Mn content in roots and shoots of *C. bursa-pastoris* affected the differentiation of the BG cluster in the positive side of the PC2 component. The formation of separate clusters representing different investigated cities suggests the presence of different MPs and PTEs pollution types and distinct responses of *C. bursa-pastoris* plants inhabiting these areas. These results suggest a potential interconnection among soil MPs, PTEs mobility in the soil, the accumulation capacity of *C. bursa-pastoris* and its consequent potential in phytoremediation of such sites. Further research is required to clarify the intricate relations between MPs and processes in the soil-plant systems.

4. Conclusions

The present study investigated the potential influence of environmental MPs in PTEs-contaminated soils on the accumulation by *C. bursa-pastoris*. Among the four studied cities, industrial city Bor exhibited the highest Cu, Mn, and Sr contamination levels, whereby 27-50% of their total content was phytoavailable. I_{geo} values indicated moderate to heavy contamination with Cu (4,76) and Sr (2,94). *C. bursa-pastors* mainly accumulated PTEs in the shoots, whereby samples from Vršac demonstrated the highest transfer ability. Spearman correlation analysis revealed positive correlations between soil MPs with Cu_{AR}, Cu_{EDTA}, and Cu_{shoots}, signifying the potential MPs-PTEs associated effects on the plant accumulation ability. The findings of this study contribute to a better understanding of how environmental MPs may potentially affect phytoremediation of PTEs pollution in soil.

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