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Human health risk assessment of PTEs in soil originating from urban parks in Serbia

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Abstract

Soil pollution with potentially toxic elements (PTEs) and their impact on human health has become an increasingly serious worldwide concern. The content of ten PTEs (As, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sr and Zn) in soil samples from urban parks in Pančevo, Smederevo, Obrenovac and Belgrade (Serbia) were measured in order to evaluate their possible health risk in this study. The concentration of As, Co, Cr, Fe, Mn and Sr were within values described for upper continental crust, unlike Cu and Pb content at all sampling sites, Ni in Pančevo, Smederevo and Obrenovac and Zn in Belgradewhich exceeded values described for upper continental crust. Their increased content is conditioned by the nature of the geological substrate, industrial activity in the environment, as well as traffic.Analysis of the health risks showed that children are more susceptible to non-carcinogenic and carcinogenic health effects of PTEs compared to adults, and that oral ingestion has the highest potential riskboth for adults and children. Lead poses a potential non-carcinogenic risk to children through ingestion pathway. The carcinogenic risk was within the acceptable limits. The results of this study could be useful in providing basic information about the PTEs content in selected urban parks and the health risk status of people inhabiting these areas.

Keywords: urban parks; soil;potentially toxic elements; health risk assessment

Introduction

Soil pollution by potentially toxic elements (PTEs) is one of the greatest issues worldwide.Although naturally present in the environment, the relative abundance of PTEs can beseriously impaired due to different anthropogenic activities, among which traffic emissions, industrial activity and municipal waste disposal have been recognized as main pollution sources in urban environment [1-6]. The highest content of PTEs occur generally in the topsoils, given the fact that surface layers, especially organic horizons, have the greatest ability to bond PTEs [2,7-9].PTEs can cause serious potential risks for humans and the environmentdue to their long term persistence in the environment, bioaccumulation, as well as acute and chronic toxicity effects, which isparticularly noticeable in the urban soils [1].Urban soils, especially in public park areas and children playgrounds may have an immediate impact on public health, mainly due to the direct contact with humans [7,8,10,11].
Health risks from exposure to PTEs in soils should not be ignored. Humans are exposed to PTEs in three ways - through ingestion, inhalation and dermal contact [1,12,13] and any high levels of PTEs in soil entering the body tissues will threaten human health and result in severe health risks [12]. In particular, the ingestion of soil has been widely regarded as one of the key pathways by which children are exposed to the PTEs from different sources [7].Previous studies reported that long-term exposure to PTEs in soils can cause serious adverse effects on human. For instance, chronic exposure to As can lead to dermal lesions, lung, skin and internal organ cancers in humans [10,14], while excessive intake of Co can lead to problems with lung, polycythemia and thyroid artery malfunction [15]. Toxic exposure to Pb can cause a severe damage to the nervous, skeletal, immune and circulatory system [15].

The main objectivesof this study were to determine content of ten PTEs (As, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sr and Zn) in the soil of urban parks in Serbian cities exposed to different sources of pollution and to assess non-carcinogenic and carcinogenic health risks of PTEs for children and adults via different exposure pathways.

Materials and methods

Sampling site description

Soil samples were collected in urban parks exposed to various sources of pollution in Pančevo, Smederevo, Obrenovac and Belgrade. The park where the sampling was performed in Pančevo is 1.3 km away from the city center, and 2.5-4.5 km from the nitrogen fertilizer factory, petrochemical industry and Pančevo Oil Refinery. The park in Smederevo is located in the central city center and is 7 km away from the iron smelter. The park in Obrenovac is also located in the central city zone, which is about 4 km away from the thermal power plants "Nikola Tesla-A" and "Nikola Tesla-B" and two ash and slag deposits caused by coal combustion. The park in Belgrade is about 2 km away from the city center and is located near several major roads (Figure 1).

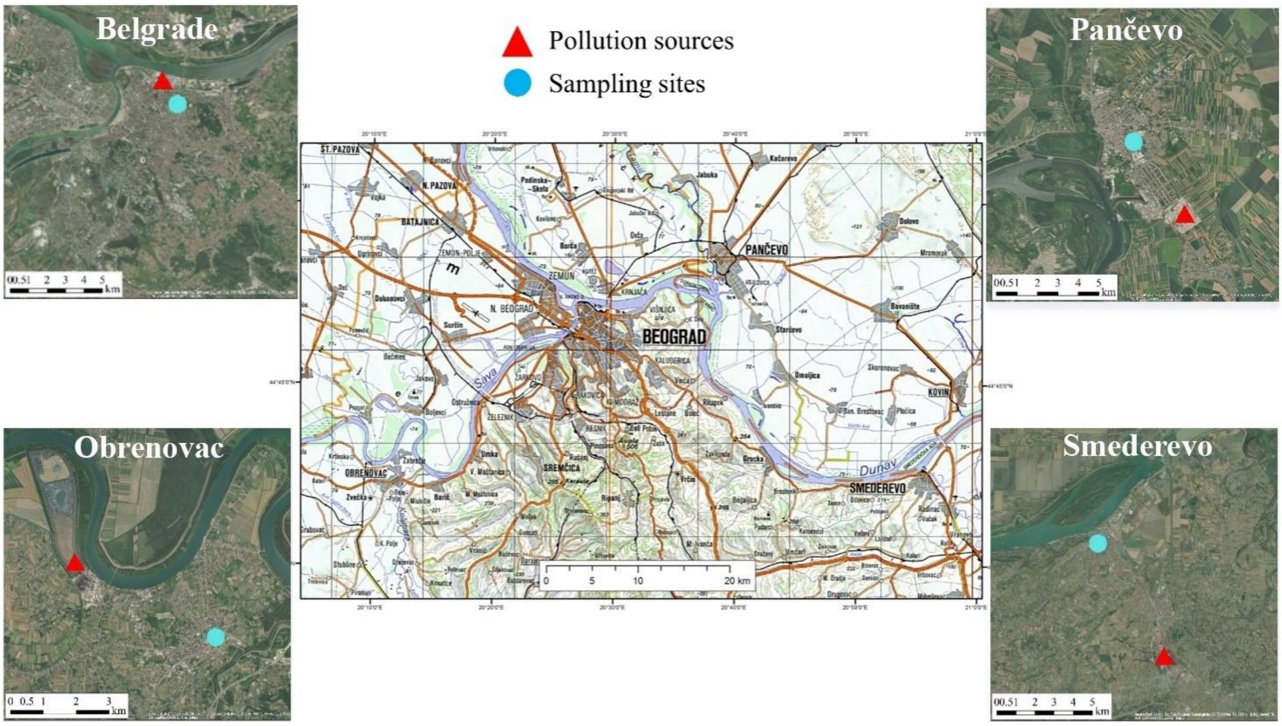

Figure 1. *Map of the selected urban parks and their distance from the pollution sources*

Soil sampling and analysis

In each park, soil sampling was performed using a pedological soil auger made of stainless steel, from a depth of 0-10 cm.In each of the examined park, samples were collected from five points, covering the whole surface of the park, from which one composite sample per park was formed.Soil samples were packed in marked polyethylene bags and delivered to the laboratory where they were dried to constant weight, grounded, and then sieved through a 2 mm sieve.Soil samples prepared in this way were used to determine the content of As, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sr and Zn. These elements were selected based on their significant contribution to soil contamination and health risk [2].

Soil samples were digested in Teflon iPrep vessels including a mixture of concentrated HNO_3 and HCl (1:3) using a microwave digestion system (CEM, 39 MDS-2000). The solutions were then diluted with ultrapure water to a final volume of 50 ml. Concentrations of the extracted elements were measured using optical emission spectrometry for simultaneous multi-element analysis (ICP - OES, Spectro Genesis). Limits of detection for the elements were: 0.006 (As), 0.001 (Co), 0.011 (Cr), 0.007 (Cu), 0.011 (Fe), 0.001 (Mn), 0.029 (Ni), 0.001 (Pb), 0.001 (Sr), 0.004 (Zn), all mg kg^{-1}. Quality assurance and quality control were performed usingmethod blanks and certified reference material (Loam soil - ERM - CC141). Standard solutions obtained by Merck(Darmstadt, Germany) were used for calibration curves. The ultrapurewater was used to prepare all solutions. The analyticalprecisionwas within ±15%.The content of chemical elements was expressed in mg kg^{-1} of the dry sample.

Health risk assessment

Non-carcinogenic risks (HQs) and carcinogenic risks (CRs) of PTEs for residential adults and children through ingestion, dermal absorption and inhalation pathways are calculated in accordance with USEPA [16,17] recommendations using the following equations:

Non-carcinogenic risk

$$HQing = [(C \times IRS \times RBA \times EF \times ED)/(BW \times AT \times RfDo)] \times 10^{-6} \quad (1)$$

$$HQder = [(C \times SA \times AF \times ABSd \times EF \times ED)/\ (BW \times AT \times RfDo \times GIABS)] \times 10^{-6} \quad (2)$$

$$HQinh = [(C \times EF \times ED)/(AT \times RfC \times PEF)] \quad (3)$$

Carcinogenic risk

$$CRing = (C * IFS * RBA * CSFo)/AT) \times 10^{-6} \quad (4)$$

$$IFS = (EF \times EDa \times IRSa/BWa) + (EF \times EDc \times IRSc/BWc) \quad (5)$$

$$CRder = [(C \times DFS \times ABSd \times CSFo)/(AT \times GIABS)] \times 10^{-6} \quad (6)$$

$$DFS = (EF \times EDa \times SAa \times AFa/BWa) + (EF \times EDc \times SAc \times AFc/BWc) \quad (7)$$

$$CRinh = C \times EF \times ED \times IUR \times 1000/AT \times PEF \quad (8)$$

The total non-carcinogenic risk for each for the three exposure pathways were assessed by hazard index (HI), which represents the sum of the HQs for all exposure pathways. The total carcinogenic risk (TCR) is calculated following the same principle as the non-carcinogenic risk. Following equations were used for calculation:

$$HI = HQing + HQder + HQinh \quad (9)$$

$$TCR = CRing + CRder + CRinh \quad (10)$$

However, as HQs, HI, CRs and TCR refer to the risk for a specific PTE, their cumulative values were calculated in order to obtain a more complete picture; this represents the total impact of all the tested elements for each exposure route (CHQs and CCRs) and the impact of all the elements through all the exposure routes together (CHI and CTCR)[2,7].

Assessments for non-carcinogenic risk are performed in relation to the value of HQ and HI, in a manner that negative health effects are not expected if their values are less than 1, while the increase in their values increases the possibility of negative non-carcinogenic effects.Thecarcinogenic risks that range between 10^{-4} and $10^{-6}$are considered to be acceptable [1,2].

The values and units associated with these equations are given in detail inPavlović et al. [7].

Results and discussion

The content of ten PTEs in examined soil samples are presented in Figure 2.

The concentrations of As, Co, Cr, Fe, Mn and Sr were within values described for upper continental crust [18], while Cu and Pb exceeded these values. Nickel content inPančevo, Smederevoand Obrenovac, as well as Zn in Belgrade were also higher than those found in the upper continental crust [18].However, the content of all tested PTEs was lower than the maximum allowable concentrations [19], except for Pb in Smederevo and Ni at all sampling sites.The increased content of PTEs that was determined in this research is on one side the consequence of the nature of the geological substrate [20], and on the other the result of the industrial activity in the environment. The impact of traffic should not be neglected as well. Namely, Cu, Pb, Ni and Zn in urban soils can originate from various anthropogenic activities, where non-exhaust vehicle emission is often mentioned as the dominant source of various PTEs in urban environment [21].For example, significant amounts of Cd, Co, Cr, Cu, Hg, Mo, Ni, Pb and Zn were associated with dust from tire wear.Zinc was the most abundant PTE from tire wear, due to the fact that ZnO and ZnS are used for tire vulcanization [21]. Also asphalt represents significant source of Ni and As in road dust [22].In addition, another source of PTEs in urban environment can be wear from braking systems and exhaust emissions.Adachia and Tainoshob [23] and Hjortenkrans et al. [24] have reported that brake dust contained significant amounts of Fe, Cu, Sb, Ba, Al, Si, S, Ti, Zn, Ni, Cr, and Pb.

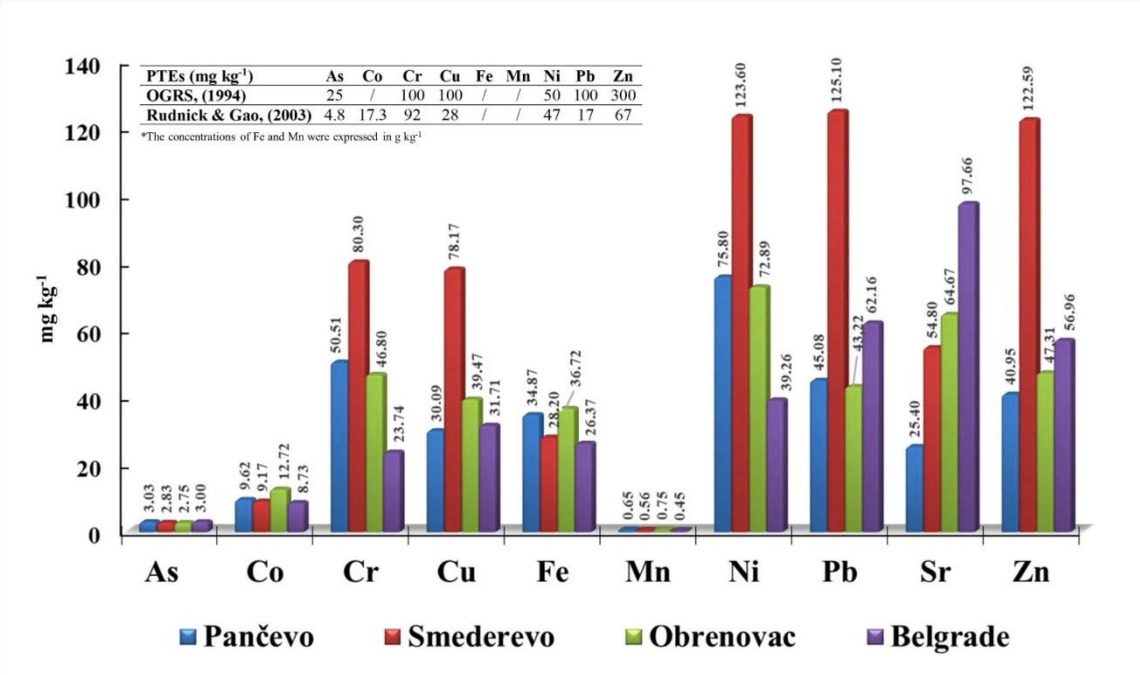

Figure 2. *Content of PTEs in the examined soils*

HQs, CHQs, HI and CHI values for children and adults are given in Table 1 and Table 2, while CRs, CCRs, TCR and CTCR values are given in Table 3.

The non-carcinogenic risks and carcinogenic risks were determined for each respective PTEs for both adults and children via various pathways, while carcinogenic risk was only quantified for those elements with defined slope factor. It was established that HQs and HI values for both children and adults were lower than 1 (Tables 1,2), suggesting that there was no significant non-carcinogenic health risk to children or adults through the ingestion, inhalation and dermal pathway. HQs and HI values lower than 1 were also reported by Han et al. [12], who studied dust and soils from urban parks and schools and by Čakmak et al. [10] who studied dust at children playgrounds. However, it was also found that HQing and HI values for Pb in Smederevo were higher than 1, indicating that Pb presents a significantly higher non-carcinogenic risk for children than other tested PTEs (Table 1).Jia et al.[25] and Pavlović et al.[7] also found an increased non-cancer risk (HQing, HI) for Pb in urban soils, which they linked to elevated Pb concentrations, as well as by its high intoxication and respective RfD value. In this research, among examined PTEs, Pb and Fe for ingestion and Mn for dermal and inhalation had highest HQ values for both adults and children. On the other side Sr for ingestion and dermal contact, and Zn for inhalation had lowest HQ values for both adults and children (Tables 1,2). Non-carcinogenic risk values of three exposure pathways for children and adults decreased in the following order: HQing - Pb $\geq$ Fe > Co > Mn > Cr > As > Ni > Cu > Zn > Sr; HQder

- Mn > Cr > As > Ni > Fe > Pb > Co > Cu > Zn > Sr, and HQinh - Mn > Co > Ni > Cr > As > Pb > Cu > Zn. Cumulative hazard quotient (CHQs) values through the three pathways mentioned above for both children and adults followed the order of CHQing > CHQinh > CHQderm, indicating that ingestion has significantly higher potential risk for children and adults, which is in accordance with previous research by Huang et al. [26], Jadoon et al. [15] and Jia et al. [25]. However, although the individual HI values for children were less than 1 (except for Pb in Smederevo), the CHI values for children were above the maximum acceptable values at all sampling sites and ranged from 1.93 in Belgrade to 2.95 in Smederevo (Table 1), while CHI values for adults were within acceptable level and ranged from 1.91×10^{-1} to 2.91×10^{-1} in Belgrade (Table 2). High CHI values for children were reported by many studies and they have also highlighted the increased sensitivity of children to the effects of PTEs [1,2,7,21,26].

The carcinogenic risk (CRs) values of As, Co, Cr, Ni and Pb through ingestion, dermal contact and inhalation pathways and total carcinogenic risk (TCR) values were within acceptable risk range of 1×10^{-4} and 1×10^{-6} (Table 3), indicating that carcinogenic riskfor residents were not expected. The CCR values for all three exposure pathways decreased in the following order: CCRing> CCRder > CCRinh. In our research, among PTEs, Cr for three types of soil exposure pathways had highest CR and TCR values, whereas the lowest values for CR and TCR had Pb and Ni respectively (Table 3). CCRing accounts for the largest share of the total cumulative cancer risk (CTCR), being as much as 10 times higher than CCRder and CCRinh. Results of this research were in line with findings of Varol et al. [2].When comparing the sampling sites, it is evident that the residents of Smederevo have the highest potential risk of developing carcinogenic illness, while the residents of Belgrade have the lowest.

Table 1. Non-carcinogenic (HQs, CHQs, HI, and CHI) risk for children through ingestion, inhalation, and dermal contact exposure pathways

HQing	As	Co	Cr[a]	Cu	Fe	Mn	Ni	Pb	Sr	Zn
Pančevo	7.74E-02	4.10E-01	2.15E-01	9.62E-03	6.37E-01	3.44E-01	4.85E-02	4.12E-01	5.41E-04	1.75E-03
Smederevo	7.24E-02	3.91E-01	3.42E-01	2.50E-02	5.15E-01	2.96E-01	7.90E-02	**1.14E+00**	1.17E-03	5.22E-03
Obrenovac	7.04E-02	5.42E-01	1.99E-01	1.26E-02	6.71E-01	3.99E-01	4.66E-02	3.95E-01	1.38E-03	2.02E-03
Belgrade	7.66E-02	3.72E-01	1.01E-01	1.01E-02	4.82E-01	2.41E-01	2.51E-02	5.68E-01	2.08E-03	2.43E-03
HQder	As	Co	Cr[a]	Cu	Fe	Mn	Ni	Pb	Sr	Zn
Pančevo	9.19E-03	9.73E-04	2.04E-02	2.28E-05	1.51E-03	2.04E-02	2.87E-03	9.77E-04	1.28E-06	4.14E-06
Smederevo	8.59E-03	9.27E-04	3.25E-02	5.93E-05	1.22E-03	1.76E-02	4.69E-03	2.71E-03	2.77E-06	1.24E-05
Obrenovac	8.35E-03	1.29E-03	1.89E-02	2.99E-05	1.59E-03	2.37E-02	2.76E-03	9.37E-04	3.27E-06	4.78E-06
Belgrade	9.09E-03	8.83E-04	9.61E-03	2.41E-05	1.14E-03	1.43E-02	1.49E-03	1.35E-03	4.94E-06	5.76E-06
HQinh	As	Co	Cr[a]	Cu	Fe	Mn	Ni	Pb	Sr	Zn
Pančevo	1.42E-04	1.13E-03	3.56E-04	8.84E-06		9.11E-03	5.94E-04	2.12E-05		8.25E-07
Smederevo	1.33E-04	1.08E-03	5.66E-04	2.30E-05		7.84E-03	9.68E-04	5.88E-05		2.47E-06
Obrenovac	1.29E-04	1.49E-03	3.30E-04	1.16E-05		1.06E-02	5.71E-04	2.03E-05		9.53E-07
Belgrade	1.41E-04	1.03E-03	1.67E-04	9.32E-06		6.38E-03	3.08E-04	2.92E-05		1.15E-06
HI	As	Co	Cr[a]	Cu	Fe	Mn	Ni	Pb	Sr	Zn
Pančevo	8.68E-02	4.12E-01	2.36E-01	9.65E-03	6.38E-01	3.74E-01	5.19E-02	4.13E-01	5.42E-04	1.75E-03
Smederevo	8.11E-02	3.93E-01	3.75E-01	2.51E-02	5.16E-01	3.22E-01	8.47E-02	**1.15E+00**	1.17E-03	5.24E-03
Obrenovac	7.89E-02	5.45E-01	2.19E-01	1.27E-02	6.72E-01	4.33E-01	4.99E-02	3.96E-01	1.38E-03	2.02E-03
Belgrade	8.59E-02	3.74E-01	1.11E-01	1.02E-02	4.83E-01	2.62E-01	2.69E-02	5.69E-01	2.09E-03	2.43E-03

CHQs	CHQing	CHQder	CHQinh	CHI
Pančevo	**2.16E+00**	5.64E-02	1.14E-02	**2.22E+00**
Smederevo	**2.87E+00**	6.83E-02	1.07E-02	**2.95E+00**
Obrenovac	**2.34E+00**	5.76E-02	1.31E-02	**2.41E+00**
Belgrade	**1.88E+00**	3.79E-02	8.06E-03	**1.93E+00**

[a]Cr(VI)

Table 2. Non-carcinogenic (HQs, CHQs, HI, and CHI) risk for adults through ingestion, inhalation, and dermal contact exposure pathways

HQing	As	Co	Cr[a]	Cu	Fe	Mn	Ni	Pb	Sr	Zn
Pančevo	7.26E-03	3.84E-02	2.02E-02	9.02E-04	5.97E-02	3.23E-02	4.54E-03	3.86E-02	5.07E-05	1.64E-04
Smederevo	6.79E-03	3.66E-02	3.21E-02	2.34E-03	4.83E-02	2.78E-02	7.41E-03	1.07E-01	1.09E-04	4.90E-04
Obrenovac	6.60E-03	5.08E-02	1.87E-02	1.18E-03	6.29E-02	3.74E-02	4.37E-03	3.70E-02	1.29E-04	1.89E-04
Belgrade	7.18E-03	3.49E-02	9.49E-03	9.50E-04	4.52E-02	2.26E-02	2.35E-03	5.32E-02	1.95E-04	2.28E-04
HQder	As	Co	Cr[a]	Cu	Fe	Mn	Ni	Pb	Sr	Zn
Pančevo	1.53E-03	1.62E-04	3.41E-03	3.81E-06	2.52E-04	3.41E-03	4.80E-04	1.63E-04	2.14E-07	6.91E-07
Smederevo	1.43E-03	1.55E-04	5.42E-03	9.89E-06	2.04E-04	2.93E-03	7.82E-04	4.52E-04	4.62E-07	2.07E-06
Obrenovac	1.39E-03	2.15E-04	3.16E-03	4.99E-06	2.66E-04	3.95E-03	4.61E-04	1.56E-04	5.46E-07	7.98E-07
Belgrade	1.52E-03	1.47E-04	1.60E-03	4.01E-06	1.91E-04	2.39E-03	2.48E-04	2.25E-04	8.24E-07	9.61E-07
HQinh	As	Co	Cr[a]	Cu	Fe	Mn	Ni	Pb	Sr	Zn
Pančevo	1.42E-04	1.13E-03	3.56E-04	8.84E-06		9.11E-03	5.94E-04	2.12E-05		8.25E-07
Smederevo	1.33E-04	1.08E-03	5.66E-04	2.30E-05		7.84E-03	9.68E-04	5.88E-05		2.47E-06
Obrenovac	1.29E-04	1.49E-03	3.30E-04	1.16E-05		1.06E-02	5.71E-04	2.03E-05		9.53E-07
Belgrade	1.41E-04	1.03E-03	1.67E-04	9.32E-06		6.38E-03	3.08E-04	2.92E-05		1.15E-06
HI	As	Co	Cr[a]	Cu	Fe	Mn	Ni	Pb	Sr	Zn
Pančevo	8.94E-03	3.97E-02	2.39E-02	9.14E-04	6.00E-02	4.48E-02	5.62E-03	3.88E-02	5.09E-05	1.65E-04
Smederevo	8.35E-03	3.79E-02	3.81E-02	2.38E-03	4.85E-02	3.86E-02	9.16E-03	1.08E-01	1.10E-04	4.94E-04
Obrenovac	8.12E-03	5.25E-02	2.22E-02	1.20E-03	6.31E-02	5.19E-02	5.40E-03	3.72E-02	1.30E-04	1.91E-04
Belgrade	8.84E-03	3.60E-02	1.13E-02	9.64E-04	4.53E-02	3.14E-02	2.91E-03	5.35E-02	1.96E-04	2.30E-04

CHQs	CHQing	CHQder	CHQinh	CHI
Pančevo	2.02E-01	9.41E-03	1.14E-02	2.23E-01
Smederevo	2.69E-01	1.14E-02	1.07E-02	2.91E-01
Obrenovac	2.19E-01	9.60E-03	1.31E-02	2.42E-01
Belgrade	1.76E-01	6.32E-03	8.06E-03	1.91E-01

[a]Cr(VI)

Table 3. Carcinogenic (CRs, CCRs, TCR and CTCR) risk for residents through ingestion, inhalation and dermal contact exposure pathways

CRing	As	Co	Cr[a]	Ni	Pb	CCRing
Pančevo	3.61E-06		3.35E-05		5.08E-07	3.76E-05
Smederevo	3.38E-06		5.32E-05		1.41E-06	5.80E-05
Obrenovac	3.28E-06		3.10E-05		4.87E-07	3.48E-05
Belgrade	3.57E-06		1.57E-05		7.00E-07	2.00E-05
CRder	As	Co	Cr[a]	Ni	Pb	CCRder
Pančevo	5.08E-07		3.76E-06		8.40E-08	4.36E-06
Smederevo	4.75E-07		5.99E-06		2.33E-07	6.69E-06
Obrenovac	4.62E-07		3.49E-06		8.05E-08	4.03E-06
Belgrade	5.03E-07		1.77E-06		1.16E-07	2.39E-06
CRinh	As	Co	Cr[a]	Ni	Pb	CCRinh
Pančevo	3.14E-09	2.09E-08	1.02E-06	5.49E-09	1.30E-10	1.05E-06
Smederevo	2.94E-09	1.99E-08	1.63E-06	8.94E-09	3.62E-10	1.66E-06
Obrenovac	2.86E-09	2.76E-08	9.48E-07	5.27E-09	1.25E-10	9.84E-07
Belgrade	3.11E-09	1.89E-08	4.81E-07	2.84E-09	1.80E-10	5.06E-07
TCR	As	Co	Cr[a]	Ni	Pb	CTCR
Pančevo	4.12E-06	2.09E-08	3.82E-05	5.49E-09	5.92E-07	4.30E-05
Smederevo	3.85E-06	1.99E-08	6.08E-05	8.94E-09	1.64E-06	6.63E-05
Obrenovac	3.75E-06	2.76E-08	3.54E-05	5.27E-09	5.67E-07	3.98E-05
Belgrade	4.08E-06	1.89E-08	1.80E-05	2.84E-09	8.16E-07	2.29E-05

[a]Cr(VI)

Conclusion

Content of potentially toxic elements As, Co, Cr, Cu, Fe, Mn, Ni, Pb, Sr and Zn in soil samples collected from urban parks in Pančevo, Smederevo, Obrenovac and Belgrade (Serbia) were measured

using optical emission spectrometry for simultaneous multi-element analysis (ICP - OES, Spectro Genesis). Furthermore, the risk model developed by USEPA was used to assess the possible health risks of these PTEs to people.
As, Co, Cr, Fe, Mn and Sr content at all sampling sites was within upper continental crust values reported by Rudnick and Gao (2003),while the concentrations of Cu and Pb were greater than the values of upper continental crust. Nickel content in Pančevo, Smederevo and Obrenovac, as well as Zn in Belgrade was also higher than the same found in the upper continental crust (Rudnick and Gao, 2003). Their increased content is conditioned by the nature of the geological substrate, industrial activity in the environment, as well as traffic.
The non-carcinogenic risk for children in the study area was in acceptable range, except in case of Pb where a significant non-carcinogenic risk via ingestion exists. Total potential carcinogenic health risks for adults were in acceptable range. The calculated results show that children are more sensitive to the adverse health effects of PTEs in relation to adults, because children are most likely to have oral intake by hand and mouth. Finally, the results of this study could be useful as basic information about the PTEs content in selected urban parks and the health risk status of people living in these cities.

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