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# ECOLOGICAL AND HEALTH RISK ASSESSMENT OF HEAVY METALS POLLUTION IN SOIL AROUND THE THERMAL POWER PLANT "NIKOLA TESLA A"

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Abstract. Ecological and health hazards were assessed based on the concentrations of selected heavy metals (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) in thirty surface soil samples taken from the sites around the largest Serbian thermal power plant. Ecological risk caused by heavy metals was estimated through pollution indices whose values indicated low to moderate ecological risks. Health hazard associated with residents' exposure to heavy metals in soil was calculated applying the U.S. EPA model. The ingestion of soil was the most important exposure pathway. The risk assessment showed that exposure to Co, Fe, and Mn would result in an unacceptable non-carcinogenic risk for children. Cr and Pb were reported to significantly contribute to a carcinogenic risk, while total carcinogenic risk remained within the acceptable non-hazardous range.

**Key words**: environmental pollution, soil, heavy metal, pollution indices, Risk Assessment Information System, "Nikola Tesla A"

### 1. Introduction

Coal-fired thermal power plants (TEPP) are one of the major sources of environmental pollution. The percentage of coal in world energy production in 2012 was 41%, and it is going to rise to 31% in 2040, which means that coal will remain fossil fuel with the largest contribution to energy production on a world scale [1]. Suspended particles, oxides of carbon, sulfur and nitrogen are the main pollutants originating from coal combustion. The principal result of combustion of organic compounds in coal is the release of hydrocarbons [2]; however, in terms of inorganic substances, such as heavy metals, coal combustion contributes to their concentrations in combustion by-products [3]. Heavy metals

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Corresponding author: Milan N. Tanić CBRN Defense Center, Belgrade, Serbia E-mail: milantanic@yahoo.com are pollutants of special concern because they are persistent and do not undergo chemical or biological degradation, and consequently, their concentration will remain constant over time once they reached soil [4]. Disposal of combustion by-products and atmospheric emission of fly ash and flue gases are two main pathways for dispersion of heavy metals in the soil surrounding coal-fired TEPP.

Very often TEPPs are situated in densely populated areas where other point and non-point pollution sources are present, and the distinction of pollution coming from TEPP is a difficult task. In that respect, delineation of the spatial distribution of heavy metals in soil is of vital importance. Geostatistics in combination with geographic information systems (GIS) enables quantification of the spatial features of investigated variables and has been frequently applied in environmental studies for investigation and mapping of soil pollution [5–7]. Moreover, the analysis of heavy metal distribution in soils of populated regions is a baseline for assessment of ecological and human health risk [8].

For ecological risk estimation, various pollution indices have been used efficiently in many environmental studies for assessment of soil pollution status concerning uncontaminated soils in general [2,7,9–11].

Heavy metals may reach the human body as a result of dietary and non-dietary intake (direct inhalation, ingestion, and dermal absorption). Most heavy metals have adverse health effects, but some heavy metals such as Fe, Mn, Co, Cu, Zn, and Ni in minute amounts act as micronutrients, whereas some metals (e.g. Cd, Cr, Pb) have carcinogenic properties. Bearing that in mind, the population living near TEPPs should be considered to be exposed to soil and ash contaminated by heavy metals, and the health risk should be continuously or at least periodically estimated.

Thermoelectric power plants participate with more than two thirds in the total capacities of the electric power industry of Serbia, and all major TEPPs use lignite as a fuel [12]. Although numerous studies have dealt with soil pollution by heavy metals originating from the Serbian TEPPs, none of them have tried to evaluate associated health risk [9,13]. The objectives of the present study were to: (i) assess ecological risk by calculating different pollution indices; (ii) quantify carcinogenic and non-carcinogenic health hazard for residents associated with heavy metals in the surface soil around the largest TEPP in Serbia, and (iii) outline spatial distribution of heavy metals in soil by means of spatial analysis.

#### 2. MATERIALS AND METHODS

#### 2.1. Study area

The TEPP "Nikola Tesla A" is the largest individual TEPP in Serbian electric power industry. It is located near Obrenovac, on the right bank of the Sava River, 35 km upstream from Belgrade. The total capacity is 1.65 GW, and the average lignite consumption is 2.51 Mt per hour. TEPP produces and disposes approximately 2.4 million tons of alkaline ash per year. Ash deposits cover a total surface of 900 ha of the study area [14].

The climate is moderate-continental with average monthly temperatures from 0.1 °C in January to 22.1 °C in July. The mean annual precipitation is 670 dm<sup>3</sup> m<sup>-2</sup>. Prevailing winds are variable and tend to primarily blow from the south-southeast, west-northwest, southeast, and west directions with the mean annual wind speed of 2.33 m s<sup>-1</sup> [15].

#### 2.2. Soil sampling and heavy metal analysis

Surface soil samples were collected at 30 locations in the vicinity of the TEPP "Nikola Tesla A", following a systematic radial sampling grid covering the period 2011–2013. The sampling points were distant from the TEPP 1, 2, 4, 6, 8, and 10 km to the west (W), southwest (SW), south (S), southeast (SE), and east (E) direction (later shown in Figure 2). Method 3050B of the United States Environmental Protection Agency (US EPA) was used for the digestion of soil samples. Total concentrations of Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn were measured by atomic absorption spectrometry. The procedures of soil sampling, sample preparation, and analysis were described in detail in our previous paper [16].

### 2.3. Pollution indices

The level of soil pollution by selected heavy metals in the study area in comparison with mean values of their concentrations in European uncontaminated topsoils [17] were evaluated by enrichment factor (*EF*) calculated according to the adjusted formula proposed by Zoller et al. [18]:

$$EF = \left[c_{sa}(HM)/c_{sa}(Fe)\right]/\left[c_{Fur}(HM)/c_{Fur}(Fe)\right] \tag{1}$$

where c(HM) and c(Fe) are the concentrations of particular heavy metal and Fe in soil, respectively, and the subscripts "sa" and "Eur" denote those concentrations in the study area and Europe.

The geo-accumulation index,  $I_{\rm geo}$ , was used originally for comparison of the current concentration of a specific element in particular media to its level in the preindustrial era. In this work,  $I_{\rm geo}$  was calculated according to the equation below [19]:

$$I_{\text{geo}} = \log_2[c_{\text{sa}}(\text{HM})/k \times c_{\text{Eur}}(\text{HM})]$$
 (2)

where k is a factor with value 1.5 that accounts possible variations in background levels for a given metal due to lithospheric effects and minor anthropogenic influences.

Contamination factor, *CF*, represents the ratio between the measured concentration of each heavy metal in the soil sample from the study area and its concentration in unpolluted soil [20] and is given by equation (3).

$$CF = c_{sa}(HM)/c_{Fur}(HM)$$
(3)

Soil classification based on EF groups [10],  $I_{\rm geo}$  classes [19] and CF grades [20] are indicated in Figure 1.

To estimate the overall level of pollution caused by all heavy metals investigated, the pollution load index (*PLI*) was calculated for each sampling point using relation (4) based on *CF* for all elements [21].

$$PLI = \sqrt[n]{CF(HM_1) \times CF(HM_2) \times \dots \times CF(HM_n)}$$
(4)

When *PLI* > 1, it is an indication that soil of interest is polluted by heavy metals in comparison with the reference soil.

#### 2.4. Health risk assessment

The potential non-cancerogenic and carcinogenic risk for children and adults were estimated for exposures to minimal, mean and maximal total measured concentration of selected heavy metals. In the case of Cr, 6:1 ratio of Cr(III):Cr(VI) was used as a reasonable estimate of the relative abundance of the two species as recommended by the US EPA. Namely, Cr(III) is an essential dietary nutrient, while Cr(VI) is a carcinogen to humans [22].

US EPA risk assessment model, which is fully accessible through Risk Assessment Information System (RAIS) [22] was exploited for risk calculation taking into account direct ingestion of soil, inhalation of resuspended particles and dermal exposure to heavy metals in soil adhered to the skin.

The basic equation for the calculation of excess lifetime cancer risk (ELCR) is:

$$ELCR = CDI \times SF \tag{5}$$

where *ELCR* represents the unitless probability of an individual developing cancer over a lifetime; *CDI* is a chronic daily intake expressed in mg kg<sup>-1</sup> day<sup>-1</sup>, and *SF* is a cancer slope factor in mg<sup>-1</sup> kg day.

Non-carcinogenic risk is expressed as the hazard quotient (HQ), a unitless number that does not have the meaning of probability. Hazard quotient for single heavy metal is calculated using equation (6):

$$HQ = CDI \times RfD^{-1} \tag{6}$$

where RfD is chronic reference dose (mg kg<sup>-1</sup> day<sup>-1</sup>). The value of HQ should be less than unity to consider risk from systemic toxicity negligible.

The total cancer risk,  $ELCR_{total}$ , is calculated as a sum of all ELCR for all heavy metals investigated and exposure routes considered. The overall non-carcinogenic risk is expressed as hazard index, HI, a unitless number equal to the sum of all HQ for all heavy metals and all exposure routes accounted for. If HI > 1 significant non-cancerogenic toxicity could be expected from a mixture of heavy metals.

Chronic daily intakes for carcinogenic risk for ingestion of soil,  $CDI_{ing,cancer}$  (mg kg<sup>-1</sup> day<sup>-1</sup>), non-carcinogenic risk for soil ingestion,  $CDI_{ing,nc}$  (mg kg<sup>-1</sup> day<sup>-1</sup>), inhalation of particulates emitted from soil for carcinogenic and non-carcinogenic risk,  $CDI_{inh}$  (mg m<sup>-3</sup>), and cancerogenic and non-carcinogenic risk due to dermal contact with soil,  $CDI_{derm}$  (mg kg<sup>-1</sup> day<sup>-1</sup>), were obtained using formulae (7–10), respectively:

$$\begin{split} CDI_{\rm ing, cancer} &= c({\rm HM}) \times EF \times IF \times AT^{-1}; \\ IF &= IR_{\rm adult} \times ED_{\rm adult} \times BW_{\rm adult}^{-1} + IR_{\rm child} \times ED_{\rm child} \times BW_{\rm child}^{-1} \end{split} \tag{7}$$

$$CDI_{\text{inc.nc}} = c(\text{HM}) \times EF \times ED \times IR \times AT^{-1} \times BW^{-1}$$
(8)

$$CDI_{\text{inh}} = c(\text{HM}) \times EF \times ED \times (PEF^{-1} + VF^{-1}) \times (ET_{\text{out}} + ET_{\text{in}} + DF_{\text{in}}) \times AT^{-1}$$
(9)

$$CDI_{\text{derm}} = c(\text{HM}) \times EF \times ED \times SA \times AF \times ABS \times BW^{-1} \times AT^{-1}$$
(10)

where c(HM) is the concentration of particular heavy metal in soil (mg kg<sup>-1</sup>); EF is exposure frequency (350 day year<sup>-1</sup>); IF is intake factor; AT is averaging time for

cancerogenesis (365 day year<sup>-1</sup> × 70 years) and non-cancerogenesis (365 day year<sup>-1</sup> × ED);  $IR_{\text{adult}}$  and  $IR_{\text{child}}$  are soil ingestion rates for adults (100 mg day<sup>-1</sup>) and children (200 mg day<sup>-1</sup>);  $ED_{\text{adult}}$  and  $ED_{\text{child}}$  are exposure duration for adults (70 years) and children (6 years);  $BW_{\text{adult}}$  and  $BW_{\text{child}}$  are body weights for adults (70 kg) and children (15 kg); PEF is a particulate emission factor (site-specific, m<sup>3</sup> kg<sup>-1</sup>); VF is volatilization factor (contaminant specific, m<sup>3</sup> kg<sup>-1</sup>);  $ET_{\text{out}}$  and  $ET_{\text{in}}$  are exposure times outdoor (0.073) and indoor (0.683);  $DF_{\text{in}}$  is dilution factor indoor (0.4); SA is available surface area in m<sup>2</sup> day<sup>-1</sup> (0.28 for children, 0.57 for adults); AF is resident soil adherence factor in mg cm<sup>-2</sup> (0.2 for children, 0.07 for adults); and ABS is absorption factor (0.001). Slope factors and reference doses considered here were taken from the RAIS compilation where toxicological profiles for all heavy metals investigated can be found [22].

### 2.5. Statistical analysis

The statistical software SPSS 20.0 was used for statistical analysis. Geostatistical data were processed and spatial modeling was carried out by Golden Software's Surfer 12<sup>©</sup> application. Inverse distance weighting was used as the interpolation method. This method presumes that the influence of the variable used for calculation is decreasing with the increase of distance from the point where the value of the variable is known [5].

#### 3. RESULTS AND DISCUSSION

#### 3.1. Heavy metal concentration in analyzed soils

A statistical summary for concentrations of analyzed heavy metals is given in Table 1. Compared to maximal limit values (LV) for concentrations of heavy metals in soil valid for European Union [23], higher content was measured only for Ni in 21 samples.

Parameter				c(F	IM)/mg kg	1			
	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Mean	0.89	17.1	66.6	36.0	38040	455	90.4	58.1	108
Median	0.74	17.7	63.3	33.6	37610	458	98.2	60.3	87.0
Std. dev.	0.49	3.7	20.9	13.1	6290	125	32.2	16.4	45.4
Min	0.41	10.9	21.8	11.5	22930	206	31.7	16.9	59.2
Max	2.45	23.5	110	64.6	51270	827	147.1	91.5	252
LV [23]	3	_	_	140	_	_	75	300	300
TV [24]	0.8	9	100	36	_	_	35	85	140
RV [24]	13	190	380	190	_	_	100	530	720
Europe [17]	0.284	8.91	32.6	16.4	21700	524	30.7	23.9	60.9

**Table 1** Descriptive statistics of heavy metal concentrations c(HM) in the soil of the study area

In environmental studies, the target (TV) and remediation (RV) values defined by Dutch soil quality guidelines [24] are widely recognized as an indication for serious deterioration of soil and as such have been incorporated in Serbian legislation for soil not used for agricultural production. Concentrations of Cd, Cr, Cu, Ni, Pb, and Zn exceeded TVs by 43%, 7%, 47%, 93%, 7%, and 20%, respectively. More importantly, none of the heavy metal concentrations is higher than corresponding RVs.

Heavy metal	Greece [25]	Turkey [3]	Slovakia [26]	Romania [27]	Croatia [28]
Cd	0.5	6.29	0.5	0.30	0.42
Co	21.9	-	_	11.6	_
Cr	346	70.5	21.2	_	184.8
Cu	36.9	59.5	18.9	29.2	215.7
Fe	52300	14630	_	_	_
Mn	1000	-	_	525	_
Ni	287	89.8	19.3	45.2	_
Pb	17.2	26.7	29.1	70.2	137.4
Zn	80	89.2	84.8	100	415.7

Table 2 Average heavy metal contents (mg kg<sup>-1</sup>) in the soil around TEPPs in Europe

Table 2 summarizes mean heavy metal contents in soils around coal-fired power plants in different European counties close to Serbia [3, 25–28]. Heavy metal concentrations reported in this work were comparable with those concentrations, with certain differences mostly related to the soil parent material, type of the coal used in TEPP, or influence of other anthropogenic sources of pollution.

## 3.2. Ecological risk assessment

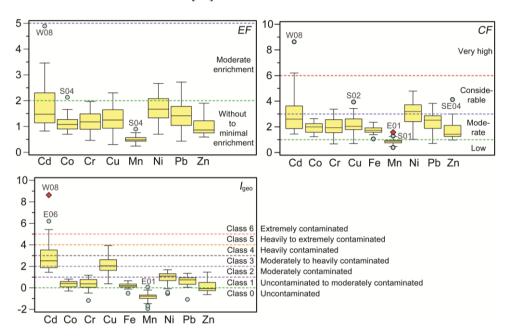
The pollution indices presented in Table 3 and Fig. 1, were calculated using the mean concentrations of heavy metals derived from aqua regia digested samples of European uncontaminated soil (0–25 cm) listed in Table 1 [17].

Table 3	Mean,	minimal	and	maximal	values	of	enrichment	factor	(EF),	contamination	l
	factor (	(CF) and $g$	geoac	cumulation	n index	$(I_{\text{geo}})$	) for heavy n	netals ir	n inves	tigated soil	

Heavy		EF			CF			$I_{ m geo}$	
metal	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max
Cd	1.78	0.82	4.89	3.14	1.44	8.63	3.01	1.44	8.63
Co	1.13	0.70	2.13	1.92	1.23	2.64	0.32	-0.29	0.81
Cr	1.19	0.46	1.97	2.04	0.67	3.38	0.37	-1.17	1.17
Cu	1.30	0.30	2.30	2.19	0.70	3.94	2.14	0.38	3.94
Fe	_	_	_	1.75	1.06	2.36	0.21	-0.51	0.66
Mn	0.50	0.24	0.90	0.87	0.39	1.58	-0.84	-1.93	0.07
Ni	1.69	0.70	2.67	2.95	1.03	4.79	0.86	-0.54	1.68
Pb	1.43	0.43	2.72	2.43	0.71	3.83	0.63	-1.08	1.35
Zn	1.01	0.59	1.90	1.78	0.97	4.13	0.14	-0.63	1.46

According to the mean EF values (0.50 for Mn to 1.78 for Cd) investigated soils are soils without minimal enrichment in comparison with reference soils. Nonetheless, Fig. 1 shows that with respect to Cd, Cu, Ni, and Pb a significant number of samples (10, 4, 9 and 4, respectively) were moderately enriched, which may imply their anthropogenic origin. Zacháry et al. reported  $EF \ge 2$  for Ni and Cu, whereas Zn, Pb, and Cd had EF between 1.2 and 1.8 for urban soil in the Ajka, town in Hungary affected by large lignite-based TEPP and lignite opencast mines [7]. The EF values obtained in the study on heavy metals in the soil around three coal-fired power plants in South Africa revealed no soil

enrichment in Mn and Fe, moderate enrichment in Cd, Cr, Cu, and Zn, and significant enrichment in Ni and Pb content [11].



**Fig. 1** Box-plot diagram of pollution indices (*EF*-enrichment factor, *CF*-contamination factor,  $I_{geo}$ -geoacumulation index) for analyzed heavy metals

Based on the mean values of *CF*, the soil of the study area was depleted in terms of Mn, considerably contaminated by Cd, and moderately contaminated by other heavy metals. However, a certain number of sampling sites were considerably endangered by the high content of Cd, Cr, Cu, Ni, Pb, and Zn, but it was significant only for Cd (8 samples) and Ni (17 samples). A very high *CF* value for Cd was recorded only for three locations (E01, E06, and W08). The mean *CF* values for Pb, Cd, Cu, Mn, Zn, Fe, and Cr of 122, 20, 8.2, 4.4, 3.4, 1.7, and 1.1, respectively were reported for the Korba city (India) near the richest coal deposits in India and four TEPPs. Such high values of *CF* for these heavy metals (especially Pb and Cd) were attributed to continuous coal exploitation, including burning and mining [29].

The  $I_{\rm geo}$  values show that soil samples were uncontaminated to moderately contaminated (classes 0–2), considering Co, Cr, Fe, Mn, Ni, Pb, and Zn together with 27% and 43% of the samples regarding Cd and Cu content, respectively. Related to Cu, the rest of the samples were classified as moderate to heavily contaminated (class 3 and 4). Among metals, Cd showed the highest accumulation in soil with 53% of the samples belonging to classes 3 and 4, while one and five samples were in class 5 and 6, respectively.

The PLI varied from 1.41 to 2.66, and the average value of 1.92 demonstrates that all sampling sites have suffered from pollution from multiple heavy metals to some extent. Furthermore, all PLI > 2 matched sampling sites taken from the eastern and south-eastern transects which correspond to downwind directions of prevalent winds.

#### 3.3. Spatial distribution of heavy metals in soil

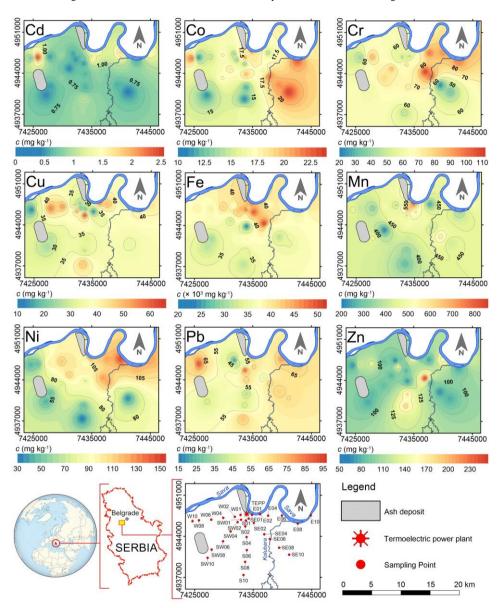
The isoconcentration contour maps for all heavy metals of interest are illustrated in Fig. 2. No regular trends can be observed, but generally, it can be noted that the most contaminated sites were located along eastern and south-eastern sampling directions. Those directions correspond to the downwind area of western and north-western winds with the strongest influence on the aerial distribution of fly ash from ash deposit, as well as flue gases from the TEPP. A considerable influence of predominant wind direction on heavy metal deposition was reported by other similar studies [3, 6, 8, 9, 26]. The discrepancy from this pattern was noticed only for Mn whose content was quite evenly distributed over the investigated area, suggesting that parent material was a dominating factor that influenced Mn spatial variability. Despite this, elevated Mn levels can be noticed in the soil close to the TEPP and ash dump. Research of Popovic et al. demonstrated that Mn was continuously leached during and after transport of ash pointing out that elevated Mn concentrations in this part of the study area could be a consequence of transport of ash/water suspension [13].

A detailed inspection of Fig. 2 shows that concentrations of most heavy metals (except Cu) were relatively higher in the closest proximity of the TEPP. Hence, it can be deduced that there has been an input of these elements from the TEPP or ash deposit. Raja et al. found higher content of fly ash-fugitive dust in close proximity of TEPPs which had led to a pH increase and higher accumulation of heavy metals [6].

Apart from soil samples collected from the vicinity of the TEPP, the highest Co concentrations were determined in sites positioned eastward from the Kolubara River where *Eutric Cambisols* are present, contrary to the eastern part of the study area consisting of alluvial soil. According to Kabata-Pendias, Co concentrations in *Cambisols* are higher than in other soil types rich in organic matter like alluvial deposits are, pointing out the pedogenic origin of Co [30].

Certain similarities can be spotted for Cd and Zn distribution which is particularly evident in the eastern part of the study area. Petrotou et al. ascribed the high concentration of these elements to the deposition of large fly ash particles in the vicinity of the TEPPs in northwestern Greece [25]. Airborne Cd emission from coal-fired electricity generation has been recognized as the most important atmospheric pollution source in the European Union, with the average annual release of 53 kg Cd per single TEPP [31]. All together suggests that Cd and Zn concentrations in the soil are likely to be related to the atmospheric deposition of fly ash from the stacks.

The concentration of Fe did not differ significantly at sites more distant from the TEPP reflecting its natural origin. Nonetheless, it is known that soil response to the deposition of fly ash from the TEPPs is the increase of soil pH through acidity neutralization [6], and that alkaline conditions promote precipitation of Fe [30]. These lands support the assumption that higher concentrations in soil adjoining the TEPP reflect airborne deposition of fly ash particles that has changed the geogenic content of Fe.



**Fig. 2** Spatial distribution of analyzed heavy metals in topsoil from the study area (Gauss–Krüger coordinate system. 7. meridian zone)

The concentration of Cr and Ni seems to be strongly influenced by the geology of the study area due to their similar distribution patterns. The highest concentrations were found very close to the Sava and Kolubara River. Alluvial sediments of these rivers were developed on ultramafic rocks, and higher geochemical content of Cr and Ni has been already reported [32]. On the other hand, chemical analysis of lignite used in TEPP "Nikola

Tesla A" showed high content of Ni, Cr and Cu and their enrichment in fly ash [33]. That may imply that geochemical concentrations of Cr and Ni could be elevated due to the TEPP activity.

The existence of certain hotspots in the western part of the study area common for Cu and Pb, and partially for Cd, Co, and Zn might indicate the impact of other anthropogenic sources. Since this part of the investigated area is an agricultural region based on fruit and vegetable growing, the application of phosphate and other fertilizers, as well as pesticides and foliage sprays, can be a huge source of Cd, Pb, Cu, Co and Zn [25, 34]. Furthermore, high levels of Pb at those sites are probably due to vehicular related emissions from dense traffic network of local roads, as a result of the long use of petrol with lead additives in Serbia in the past.

#### 3.4. Health risk analysis

The magnitudes of chronic non-carcinogenic health issues are summarized in Table 4. As it can be seen, the risk from the ingestion of soil particles by children and adults comprises almost whole cumulative HI. Dermal risk exists only for the exposition to Cd in soil, and it is negligible in both categories ( $4 \times 10^{-9}$  to 0.003). The risk arising from inhalation exposure is not respective because calculated HI is so benevolent with a maximal value of  $1 \times 10^{-8}$ . This is consistent with the conclusion obtained by Tang et al. in the study of children's exposure to heavy metals in soils around the coal-fired TEPP in China (Huainan City) [8].

Although none of *HQ* for any single heavy metal was above the reference value of one, aggregate *HI* for children fell in the range from 1.040 to 2.604 (mean 1.793). Cobalt, Fe, and Mn were identified as contaminants of most concern with *HI* values ranged between 0.110 and 0.441 for Mn, 0.419 and 0.936 for Fe, and 0.465 and 1.000 for Co. All of them are considered essential nutrients, but their gastrointestinal intake in higher concentration may cause adverse health effects. In already mentioned study of Tang et al. the total *HI* for children due to As, Cd, Cr, Cu, Mn, Ni, Pb, V, and Zn in soil was 1.502, and excluding *HQ* for As and V it was 0.545 which is lower than *HI* obtained in this research, but without contribution from Co and Fe [8].

It is known that oral Co exposure in humans can induce respiratory, cardiovascular, gastrointestinal, hematological, hepatic, renal, endocrine, dermal, ocular, hypothermic, and body weight effects. Available data have not clearly defined whether a risk for children is greater than for adults from exposure to Co [35]. The different studies conducted in the world have shown that average Co intake varied among countries. It is estimated to be 0.12 mg day<sup>-1</sup> in the United Kingdom, 5–40 μg day<sup>-1</sup> in the US, 11 μg day<sup>-1</sup> in Canada and 29 μg day<sup>-1</sup> in France [36]. Calculated *CDI*<sub>ing</sub> for the highest Co concentration in this research was 0.3 μg kg day<sup>-1</sup>, and the ingestion of total Co content by 15 kg child would result in a daily intake of 4.5 μg day<sup>-1</sup>. This is still less than reported daily intakes for mentioned countries and allows additional intake through the food. All things considered, although the maximal value of *HI* for Co in soil was 1, likely, Co would not provoke adverse effects in children.

**Table 4** The potential non-carcinogenic risk for resident population due to minimal, mean and maximal heavy metal contents in soil

Heavy metal			Child	ren			Adults			
		$HQ_{\rm ing}$	$HQ_{\mathrm{inh}}$	$HQ_{ m derm}$	HI	$HQ_{\rm ing}$	$HQ_{\mathrm{inh}}$	$HQ_{ m derm}$	HI	
Cd	min	0.005	$2 \times 10^{-11}$	$5 \times 10^{-4}$	0.006	$5 \times 10^{-4}$	$2 \times 10^{-11}$	$8 \times 10^{-5}$	$6 \times 10^{-4}$	
	mean	0.011	$5 \times 10^{-11}$	0.001	0.012	0.001	$5 \times 10^{-11}$	$2 \times 10^{-4}$	0.001	
	max	0.031	$1 \times 10^{-10}$	0.003	0.034	0.003	$1 \times 10^{-10}$	$5 \times 10^{-4}$	0.003	
Co	min	0.465	$1 \times 10^{-9}$	-	0.465	0.044	$1 \times 10^{-9}$	-	0.044	
	mean	0.729	$2 \times 10^{-9}$	_	0.729	0.068	$2 \times 10^{-9}$	_	0.068	
	max	1.000	$2 \times 10^{-9}$	_	1.000	0.094	$2 \times 10^{-9}$	_	0.094	
Cr(VI)	min	0.013	$2 \times 10^{-11}$	_	0.013	0.001	$2 \times 10^{-11}$	_	0.001	
	mean	0.041	$5 \times 10^{-11}$	-	0.041	0.004	$5 \times 10^{-11}$	-	0.004	
	max	0.067	$9 \times 10^{-11}$	_	0.067	0.006	$9 \times 10^{-11}$	-	0.006	
Cu	min	0.004	_	_	0.004	$4 \times 10^{-4}$	_	-	$4 \times 10^{-4}$	
	mean	0.012	_	-	0.012	0.001	-	_	0.001	
	max	0.021	_	_	0.021	0.002	_	-	0.002	
Fe	min	0.419	_	_	0.419	0.039	_	-	0.039	
	mean	0.695	_	-	0.695	0.065	-	-	0.065	
	max	0.936	_	_	0.936	0.088	_	-	0.088	
Mn	min	0.110	$2 \times 10^{-9}$	-	0.110	0.010	$2 \times 10^{-9}$	-	0.010	
	mean	0.242	$5 \times 10^{-9}$	_	0.242	0.023	$5 \times 10^{-9}$	_	0.023	
	max	0.441	$9 \times 10^{-9}$	_	0.441	0.041	$9 \times 10^{-9}$	-	0.041	
Ni	min	0.020	$2 \times 10^{-10}$	_	0.020	0.002	$2 \times 10^{-10}$	_	0.002	
	mean	0.058	$6 \times 10^{-10}$	_	0.058	0.005	$6 \times 10^{-10}$	_	0.005	
	max	0.094	$9 \times 10^{-10}$	_	0.094	0.009	$9 \times 10^{-10}$	-	0.009	
Zn	min	0.003	_	_	0.003	$2 \times 10^{-4}$	_	-	$2 \times 10^{-4}$	
	mean	0.005	_	-	0.005	$4 \times 10^{-4}$	-	-	$4 \times 10^{-4}$	
	max	0.011	_	-	0.011	0.001	-	_	0.001	
HI	min	1.039	$4 \times 10^{-9}$	$5 \times 10^{-4}$	1.040	0.097	$4 \times 10^{-9}$	$8 \times 10^{-5}$	0.097	
	mean	1.792	$7 \times 10^{-9}$	0.001	1.793	0.168	$7 \times 10^{-9}$	$2 \times 10^{-4}$	0.168	
	max	2.601	$1 \times 10^{-8}$	0.003	2.604	0.244	$1 \times 10^{-8}$	$5 \times 10^{-4}$	0.244	

The US EPA derived *RfD* of 0.14 mg kg<sup>-1</sup> day<sup>-1</sup> for the total oral intake of Mn [37]. By strictly following the value of Mn in the soil of the study area there would not be any significant risk, but instead of this value *RfD* of 0.024 mg kg<sup>-1</sup> day<sup>-1</sup> was used for the nonfood items. This is also proposed by US EPA due to numerous uncertainties regarding the risk associated with Mn, and likely due to the strong homeostatic control of Mn absorption [22]. Studies conducted on children have indicated that extremely high doses may induce deleterious effects on the central nervous system. The tolerable upper intake level (UL) for Mn, for children, varies from 2 to 9 mg day<sup>-1</sup> depending on the age and body weight [38]. Obtained *CDI*<sub>ing</sub> for children in this work for the highest Mn level in soil was 0.0106 mg kg day<sup>-1</sup>, which would result in a case of 15 kg child with a daily intake of 0.159 mg Mn. Such intake is well below the lowest UL, so the risk of harmful effects due to excess Mn intake appears to be very low.

Iron is a component of many proteins, including enzymes and hemoglobin, but it also can be damaging when it accumulates in the body. The Fe content of the body is highly conserved because the body normally absorbs less Fe if its stores are full. For this reason it is important to perform a risk assessment of such an element. Vomiting and diarrhea

characterize the initial stages of intoxication, and later toxicity is expressed in the cardiovascular, central nervous system, kidney, liver, and hematologic organ systems. First symptoms appear with doses between 20 and 60 mg kg<sup>-1</sup> [38]. Lethal doses of Fe are in the range of 200–300 mg kg<sup>-1</sup> [39]. The value of UL for infants and children up to the age of 13 is 40 mg day<sup>-1</sup>, and for adolescents (age of 14–18) is 45 mg day<sup>-1</sup> [38]. Comparing to those values, calculated maximal  $CDI_{ing}$  for children for Fe in the soil of the study area (0.486 mg kg<sup>-1</sup> day<sup>-1</sup>) will yield a daily Fe intake of 7.29 mg day<sup>-1</sup>, which is approximately 56% of the lowest UL recommended.

The values for HQ for adults of all heavy metals were below the threshold value of one. The HI was ranged from 0.097 to 0.244 which indicates negligible risk level.

Pb toxicity has been known ever since this element is omitted from the non-cancerogenic risk assessment. Lead is a systemic poison with multisystem toxicity that affects the gastrointestinal tract, kidneys, hematopoietic, cardiovascular, nervous, immune, and reproductive system. Studies of Pb toxicity have not established a 'safe' exposure limit for Pb, therefore, the US EPA has not established an RfD for lead [40]. Young children are most vulnerable to Pb toxic effects due to their rapid growth rate, development of central nervous systems and substantially higher ability to absorb Pb than adults. In general, approximately 10-15% of Pb ingested by adult humans is absorbed, whereas in children this value may be as high as 50%. More than 90% of inhaled Pb particles deposited in the lower respiratory organs enter the bloodstream. Inorganic Pb is not efficiently dermally absorbed and this route does not contribute markedly to the total body Pb burden [22]. Peripheral nerve dysfunction occurs at blood Pb levels of 30-50 µg dm<sup>-3</sup>, and peripheral nerve damage at 400–600 μg dm<sup>-3</sup>. The primary target for Pb is the central nervous system. Irreversible brain damage has been observed at blood Pb levels exceeding 1000 µg dm<sup>-3</sup> in adults and at 800– 1000 μg dm<sup>-3</sup> in children [22]. The calculated  $CDI_{ing}$  for maximal Pb concentration in the soil for children and adults in this work was  $1.2 \times 10^3$  mg kg day<sup>-1</sup>, and  $1.1 \times 10^4$  mg kg day<sup>-1</sup>. In the worst-case scenario, assuming that total volume of blood in 15 kg child is 1.125 dm<sup>-3</sup>, and 100% Pb bioavailability, and if all Pb from soil would enter the bloodstream, the resulting blood level would be 15.6 µg dm<sup>-3</sup> which would have not been enough to cause any severe health effect in children.

The cancer risks are quantified in Table 5. Among heavy metals measured, only Cd, Co, Cr(VI), Ni and Pb are classified as human carcinogens [22]. The total cancer risk in the present study ranged from  $1.04 \times 10^{-5}$  to  $5.24 \times 10^{-5}$ . The US EPA considers cancer risk below  $10^{-6}$  to be negligibly small, and risk of  $10^{-4}$  to be sufficiently large that remediation is desirable. Cancer risk between  $10^{-6}$  and  $10^{-4}$  is generally considered acceptable. The exposure route that has the highest contribution to the  $ELCR_{total}$  appears to be the ingestion of soil, followed by an insignificant contribution from inhalation. There is no risk induced by dermal exposure.

**Table 5** Carcinogenic lifetime risk for the resident population based on minimal, mean and maximal heavy metal concentration in soil

Heavy meta	al	$ELCR_{ing}$	$ELCR_{inh}$	$ELCR_{derm}$	$ELCR_{total}$
Cd	min	-	$1.55 \times 10^{-16}$	_	$1.55 \times 10^{-16}$
	mean	-	$3.36 \times 10^{-16}$	_	$3.36 \times 10^{-16}$
	max	-	$9.24 \times 10^{-16}$	_	$9.24 \times 10^{-16}$
Co	min	-	$2.06 \times 10^{-14}$	_	$2.06 \times 10^{-14}$
	mean	_	$3.22 \times 10^{-14}$	_	$3.22 \times 10^{-14}$
	max	_	$4.43 \times 10^{-14}$	_	$4.43 \times 10^{-14}$
Cr(VI)	min	$1.02 \times 10^{-5}$	$1.52 \times 10^{-13}$	_	$1.02 \times 10^{-5}$
	mean	$3.10 \times 10^{-5}$	$4.63 \times 10^{-13}$	_	$3.10 \times 10^{-5}$
	max	$5.13 \times 10^{-5}$	$7.66 \times 10^{-13}$	_	$5.13 \times 10^{-5}$
Ni	min	_	$1.73 \times 10^{-15}$	_	$1.73 \times 10^{-15}$
	mean	-	$4.92 \times 10^{-15}$	_	$4.92 \times 10^{-15}$
	max	_	$8.01 \times 10^{-15}$	_	$8.01 \times 10^{-15}$
Pb	min	$2.07 \times 10^{-7}$	$4.25 \times 10^{-17}$	_	$2.07 \times 10^{-7}$
	mean	$7.10 \times 10^{-7}$	$1.46 \times 10^{-16}$	_	$7.10 \times 10^{-7}$
	max	$1.12 \times 10^{-6}$	$2.30 \times 10^{-16}$	_	$1.12 \times 10^{-6}$
ELCR <sub>total</sub>	min	$1.04 \times 10^{-5}$	$1.75 \times 10^{-13}$	_	$1.04 \times 10^{-5}$
Total.	mean	$3.17 \times 10^{-5}$	$5.01 \times 10^{-13}$	_	$3.18 \times 10^{-5}$
	max	$5.24 \times 10^{-5}$	$8.19 \times 10^{-13}$	_	$5.24 \times 10^{-5}$

According to data obtained from the Institute of Public Health of Serbia, the real standardized cancer incidence for 2013 in Kolubara County (where study area belongs) was  $3.24 \times 10^{-3}$  for males and  $2.40 \times 10^{-3}$  for females [41]. These values are very high in comparison to the risk assessed in this study. Therefore, the risk provoked by exposure to heavy metals in the soil makes a portion of the real cancer risk that is completely insignificant.

The total cancer risk from the ingestion of Cr(VI) was between  $1.02 \times 10^{-5}$  and  $5.13 \times 10^{-5}$ , which means that almost all  $ELCR_{total}$  consists of risk coming from oral intake of this element. These findings should be taken with caution. Namely, Cr(VI) is known to be carcinogenic in humans by the inhalation route associated with increased lung cancer risk among workers in certain industries. Nevertheless, Cr(VI) by ingestion is also likely to be carcinogenic. Accordingly, a more health-protective approach for cancer risk assessment due to Cr(VI) exposure via ingestion is being applied. In that respect, SF of  $0.5 \text{ mg}^{-1}$  kg day was used for the calculation of oral cancer risk [42]. This figure is quite low and primarily was developed for drinking water. Under those circumstances, it is likely that oral Cr(VI) cancer risk is overestimated.

The second-largest contributor to the overall cancer risk is Pb. The most common cancers provoked by Pb exposure are lung and brain cancer [22]. Although the risk caused by this element is two orders of magnitude lower than in the case of Cr, it should not be ignored since the calculated lifetime risk for maximal Pb content is  $1.12 \times 10^{-6}$ , and exceeded general guidance risk value of  $10^{-6}$ .

In the end, it is important to realize that the US EPA model used for risk assessment assumes an absorption efficiency of 100% for all heavy metal concentrations used. Since the concentrations determined in this work were total concentrations, the final results have to be considered as very conservative because no total concentrations are bioavailable to humans.

#### 6. CONCLUSION

This study evaluated the potential ecological and human health risk due to heavy metals in soils in the proximity of the largest Serbian TEPP and provides baseline data regarding the distribution of heavy metals in soil. The analysis of the potential ecological risk showed that soil is considered a relatively low to moderate ecological risk. Contamination was observed in the studied soil samples, most of all with Cd, following with Ni, Cu, and Pb. Analysis of the heavy metals spatial distribution revealed that soil in proximity to the TEPP and the downwind area had a higher level of analyzed heavy metals, while soil away from the TEPP was under greater influence from natural processes and other anthropogenic impacts (agriculture and traffic). The estimated carcinogenic risk in this research is within the acceptable range. Figures for non-carcinogenic risk suggest that adults are not endangered due to heavy metals in soil, while the children population is under elevated risk from deleterious health effects. Ingestion of soil was identified as a primary pathway of heavy metal sources which harm human health. This study may be valuable data source for long-term monitoring of soil quality and may provide a basis for the development of further activities related to the prevention of excessive exposure of humans, especially children, to heavy metals present in the soil. A future study applying a sequential extraction procedure is recommended to determine the proportion of the total metal pool that might be bioavailable.

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# PROCENA EKOLOŠKOG I ZDRAVSTVENOG RIZIKA OD TEŠKIH METALA U ZEMLJIŠTU U OKOLINI TERMOELEKTRANE "NIKOLA TESLA A"

Ekološki i zdravstveni rizik su procenjeni u odnosu na koncentracije odabranih teških metala (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb i Zn) u trideset površinskih uzoraka zemljišta iz okoline najveće termoelektrane u Srbiji. Ekološki rizik je procenjen putem različitih pokazatelja zagađenja i njihove vrednosti pokazuju niski do srednji ekološki rizik. Rizik po zdravlje usled izlaganja stanovništva teškim metalima u zemljištu je izračunat primenom modela Agencije za zaštitu životne sredine Sjedinjenih Američkih Država. Ingestija zemljišta je identifikovana kao najvažniji put izlaganja. Procena rizika je pokazala da ekspozicija Co, Fe i Mn rezultuje neprihvatljivim nekancerogenim rizikom za decu. Cr i Pb su bili jedini elementi koji su doprinosili u većem obimu kancerogenom riziku, ali je ukupan kancerogeni rizik bio u granicama tolerantnog.

Ključne reči: zagađenje, zemljište, teški metal, pokazatelji zagađenja, Informacioni sistem za procenu rizika, Nikola Tesla A