

Pollution indices as useful tools for comprehensive evaluation of the soil contamination degree in the vicinity of mining and metallurgical complexes

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Abstract

Soil pollution levels in the copper mining and metallurgical area were evaluated at 14 sampling sites in the City of Bor and its surroundings in regard to Al, As, Cu, Fe, Pb and Zn contents, as well as single and integrated pollution indices. The significance of single pollution indices provides information about pollution by a specific element, while integrated pollution indices offer an insight into cumulative pollution by the examined elements. The mean soil concentrations of As, Cu, Pb and Zn were several times higher than the world average values. The exceedances of soil remediation values were most pronounced for As (at seven sites) and Cu (at eleven sites), more than 3 and 13 times, respectively. According to the geoaccumulation index, the enrichment factor and the contamination factor, the highest soil contamination was with As and Cu, especially at the urban-industrial site. Pollution load index, Nemerow pollution index and the improved Nemerow index confirmed that the most contaminated soils were from the sites in the vicinity of the metallurgical complex and flotation tailing ponds, as well as from the sites in the prevailing wind directions compared to the less polluted soils affected by the ore mining processes. The areas affected by the serious cumulative contamination from the pyrometallurgical copper production need continuous pollution prevention, monitoring and remediation measures.

Keywords: Environmental pollution; soil quality; single and integrated soil pollution indices.

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1. INTRODUCTION

Emissions of hazardous substances into the atmosphere from the mining and smelting activities, mostly contain acid gases such as SO₂ and particulate matter (PM) with different trace elements, some of which can potentially be harmful to humans by inhalation or could be bioaccumulated through the food chain [1-3]. These pollutants cause concern for all the receiving matrices (airborne particulates, vegetation, soil, sediments, dust, waters, etc.) [4-6].

Essential elements (such as Cu, Ni, Zn, etc.) are necessary for many metabolic functions in soil and living organisms but could become toxic at concentrations above the allowed values, whereas non-essential elements (such as Pb and Cd) are potentially toxic even at very low concentrations in the soils [7]. Hazardous substances emitted into the atmosphere in the form of particulate matter and gases precipitate on the soil surfaces and vegetation in the form of dry or wet depositions [4,8].

Areas that are located in the direction of the prevailing winds are usually polluted with emissions from non-ferrous mining and smelting operations, as well as with dust and fugitive emissions rich in trace elements (e.g. As, Cu, Pb, etc.) [9,10]. Many studies aimed to assess environmental pollution at different locations, such as specific mining and smelting areas [2-4,9,11-19], urban and traffic areas [6], industrial areas [8,20-23], farmland and cultivated soils [24], national parks [5,25,26], sediments [27], etc.

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Data regarding the air pollution obtained from the measuring stations, usually located in the urban and residential areas, as well as the data obtained by biomonitoring using different plant species and soil, *e.g.* [11,12,28], could be used for estimation of potential influences of industrial, *e.g.* [8,21,29] and traffic, *e.g.* [6] pollution on the environment.

Soil as the most important and conditionally renewable resource in the environment, which acts as the main receiving surface area of the atmospheric deposition, represents one of the main topics in the environmental quality investigations [25,30]. Contaminated soil has a negative effect on biogeochemical cycles, the state of surface and underground waters and could result in leaching of the deposited pollutants in the substrates [4,8]. Phytoremediation represents an environmentally friendly method for recovery and the removal of trace elements from the contaminated soils [8,31,32].

Many studies conducted worldwide showed the use of different indices in the comprehensive evaluation of the soil contamination by anthropogenic activities [33-35], and for soil classification [36-38]. Some investigation benefits of using soil pollution indices are: easy and fast calculation; large international experience; national and international agencies recognising pollution assessment by indices and simple environmental risk assessment. The pollution indices could denote the presence of trace elements both from natural and anthropogenic processes [22,39]. According to literature, the occurrence of false-negative results or lack of comparability studies could be regarded as disadvantages of soil pollution indices [7].

The aim of this study was to assess the soil pollution level in the area of copper mining and smelting. In order to determine the pollution level of soil, concentrations of trace elements were compared to the corresponding limit values (*LV*) and remediation values (*RV*), proposed by the Serbian regulation, as well as by using single and integrated indices of soil pollution, such as: geoaccumulation index (I_{geo}), enrichment factor (*EF*), contamination factor (*CF*), pollution load index (*PLI*), Nemerow pollution index (*NI*), and improved (modified) Nemerow index (*INI*). In this study, the pollution indices were used for determining distribution patterns of the emitted elements and highlighting the polluted sampling sites.

2. MATERIALS AND METHODS

2. 1. Study area description

Mining activities and pyrometallurgical production of copper from sulfide ores in the City of Bor (Eastern Serbia) and its surroundings have been performed for more than 120 years. Huge quantities of acid gases such as SO_2 , particulate matter (PM) and atmospheric depositions containing different elements, some of which can be highly toxic and carcinogenic, are emitted during smelting of sulfide copper concentrates, containing chalcopyrite ($CuFeS_2$), chalcosine (Cu_2S) and coveline (CuS) [28,40]. Long-term pollution of the environment resulting from the emissions of toxic elements, especially with As, characterised the Bor area as one of the most polluted in Europe, *e.g.* [13,15]. For that reason, the City of Bor and its surroundings were chosen as the study area. The climate in this area is moderately continental. The prevailing winds are in the W and WNW directions, followed by the NW, E, and ENE winds, while the least frequent winds are in the ESE, S and SSW directions [3].

The sampling sites (Fig. 1) were chosen considering the pollution sources such as: the copper smelter (denoted as the primary pollution source), and open pits, ore waste heaps, and flotation tailing ponds (secondary pollution sources). Also, the prevailing wind directions were taken into account, due to the significant influence on the dispersion and transport of the polluting substances.

Description of location of 14 sampling sites, their distance from the primary pollution source, as well as the dominant winds affecting the pollution level at the sampling sites, is presented in Table 1.

All the sampling sites were under the influence of particular pollution sources, except the sampling site B, which represented the background site. This sampling site is surrounded by mountain massifs and protected from the air pollution.

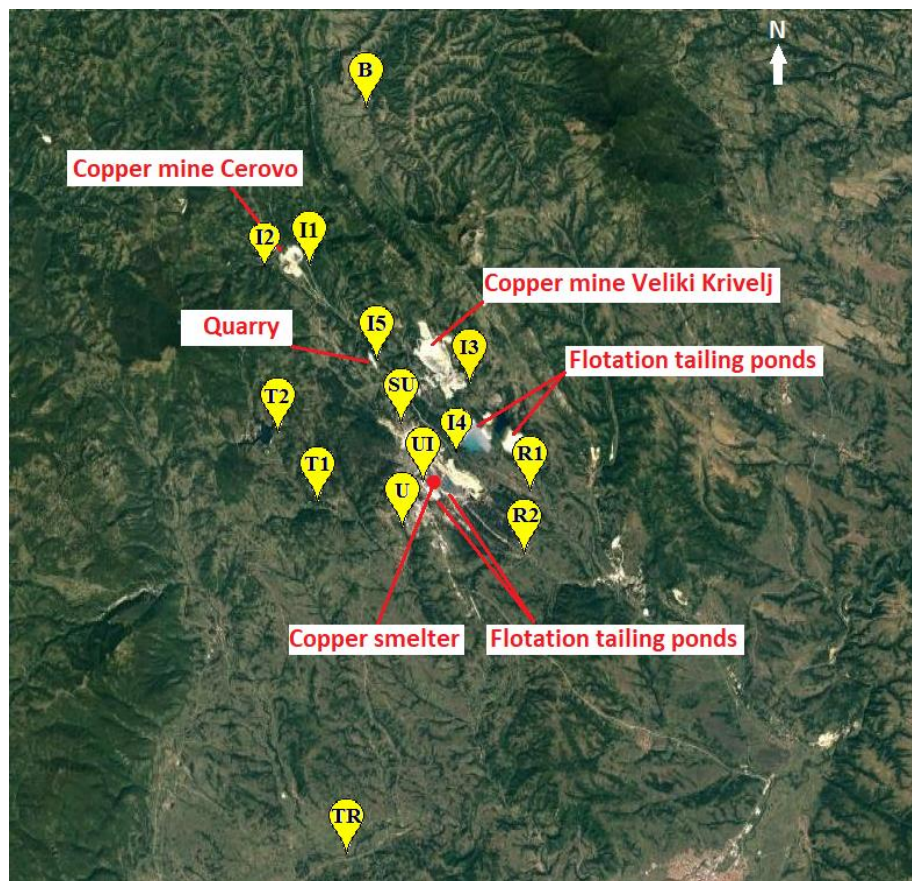


Figure 1. Map of the study area

Table 1. Description of the sampling sites

Sampling site	Location	Pollution sources	Wind direction
UI	Urban-industrial site, 0.5-2.5 km SW from the copper smelter, located in the city center	Copper smelter, flotation tailing ponds, city heating plant, traffic	ENE, NE
U	Urban site, >2.5 km SW from the copper smelter, residential area of the city	Copper smelter, traffic	ENE, NE
SU	Suburban site Brezonik, 2.5 km NW from the copper smelter	Copper smelter, flotation tailing ponds	SE, ESE
I1, I2	Industrial sites in the surroundings of the copper mine Cerovo, 11 km NW from the copper smelter	Open pit, ore waste hips	SE
I3	Industrial site in the surroundings of the copper mine Veliki Krivelj, 5.5 km N from the copper smelter	Ore waste hips, flotation tailing ponds, copper smelter	S
I4	Industrial site in the surroundings of the flotation tailing ponds of the copper mine Veliki Krivelj, 4.5 km NNE from the copper smelter	Flotation tailing ponds, ore waste hips, copper smelter	SW
I5	Industrial site in the surroundings of the quarry, 6 km NNW from the copper smelter	Dust emissions from exploiting and crushing of limestone	SSE, SE
R1	Rural settlement Oštrelj, 4.5 km ESE from the copper smelter	Flotation tailing ponds, copper smelter	WNW, WN
R2	Rural settlement Slatina, 6.5 km SE from the copper smelter	Copper smelter	WNW, WN
T1	Tourist area of the local Brestovac spa, 4.5 km WSW from the copper smelter	Periodical emissions from the copper smelter	ENE, E
T2	Tourist area of the local Bor lake, 7 km WNW from the copper smelter	Periodical emissions from the copper smelter	ESE, E
TR	The local traffic road, 20 km SSW from the copper smelter	Traffic pollution	N, NNE
B	Rural settlement Gornjane, 17 km N from the copper smelter	No air pollution from the copper smelter	S

2. 2. Sampling pattern and soil analysis

Collection of the soil samples was carried out at 14 sampling sites (Fig. 1). The soil was sampled at a depth of 10-20 cm. Soil preparation included air-drying of samples at room temperature and grinding to a fine powder [3]. Prior to chemical analyses, the soil samples were digested, following the U.S. EPA method 3050B [41], in a microwave oven (Ethos E, Milestone), with “aqua regia”, *i.e.* a mixture of HNO₃ and HCl (volume ratio 1:3) (65 % HNO₃, J.T. Baker; 36.5-38 % HCl, J.T. Baker) [3]. Chemical analyses of the soil samples were performed in the accredited chemical laboratory, at the Mining and Metallurgy Institute Bor (Bor, Serbia). The concentrations of the analysed elements were determined by the simultaneous dual view inductively coupled plasma atomic emission spectrometer (ICP-AES, Spectromodel Blue). The quality of the obtained analytical data was verified by the blanks and three replicates of the same sample of soil. The concentrations of all the analysed elements are given as mg kg⁻¹ dry mass.

2. 3. Data analyses

Descriptive analysis of the examined element concentrations in the soil was conducted using the SPSS version 17.0. Based on the obtained concentrations and defined limit and remediation values set by the Serbian Regulation [42], classification of the soil from each sampling site was carried out. According to this Regulation, concentration of the specific element in soil above the limit value indicates that functional properties of the soil and sustainable soil quality are endangered. The remediation value represents the concentration, which indicates that the fundamental functions of the soil are seriously disturbed, and the soil requires re mediation, recovery and other measures.

Two types of the commonly used pollution indices, for assessing the environmental implications of the element contents (mg kg⁻¹) in the soil, are single (I_{geo} , EF , CF) and integrated (PLI , NI , INI). The single indices provide information about the pollution degree by the specific element, whereas the integrated indices provide information about pollution by more than one element, giving a comprehensive evaluation of soil contamination [26].

Geo-accumulation index (I_{geo}) is widely used for assessment of soil pollution by the specific element compared with its respective geochemical background concentration. This index was calculated by using the following equation [33,43,44]:

$$I_{geo} = \log_2 \frac{C_n}{1.5B_n} \quad (1)$$

where: C_n is the concentration of the studied element in the soil; B_n is the geochemical background concentration of the corresponding element; 1.5 is the factor used for reduction of possible variations in the background values caused by the natural pedogenic processes. In this study, the concentration of the element in soil, which is not affected by anthropogenic activities was used as the local geochemical background concentration (B_n) [33,44].

The values of I_{geo} were categorised into seven soil contamination classes [22,24,33]: $I_{geo} < 0$, unpolluted; $0 < I_{geo} \leq 1$, unpolluted to moderately polluted; $1 < I_{geo} \leq 2$, moderately polluted; $2 < I_{geo} \leq 3$, moderately to heavily polluted; $3 < I_{geo} \leq 4$, heavily polluted; $4 < I_{geo} \leq 5$, heavily to extremely polluted; $I_{geo} > 5$, extremely polluted.

Enrichment factor (EF) is used for assessment of the possible anthropogenic influence on the element concentration in the soil. EF was calculated by using the equation [10,24,36]:

$$EF = \frac{\left(\frac{C_n}{C_{ref}} \right)_{sample}}{\left(\frac{C_n}{C_{ref}} \right)_{background}} \quad (2)$$

where: C_{ref} is the concentration of the selected reference element. In this study, Al was selected as the reference element for the geochemical normalisation purposes, mostly due to its stability in soil and the lack of contamination with this element in the study area. It is considered that the enrichment of the soil originating from anthropogenic sources is present when $EF > 2$ [36]. However, soil enrichment has five categories [10,24,26,36]: $EF < 2$, deficiency to minimal enrichment; $2 \leq EF < 5$, moderate enrichment; $5 \leq EF < 20$, significant enrichment; $20 \leq EF \leq 40$, very high enrichment; $EF > 40$, extremely high enrichment.

Contamination factor (CF) represents the ratio between the concentration of the specific element in the soil and the corresponding background soil concentration [19,23,35]:

$$CF = \frac{C_n}{C_b} \quad (3)$$

where: C_b represents the geochemical background concentration of the corresponding element.

The CF value indicates the anthropogenic influence on soil contamination with the specific element [23], classified as [19,35,45,46]: $CF < 1$, low contamination; $1 \leq CF < 3$, moderate contamination; $3 \leq CF < 6$, considerable contamination; $CF \geq 6$, very high contamination. Higher CF values indicate lower retention times of the element in the soil and a higher risk to the environment [47]. The CF values are also used for calculation of the Nemerow pollution index (NI) [22]. The CF values usually indicate higher degrees of soil contamination with elements in comparison to the I_{geo} values [35].

Calculated values of the Pollution load index (PLI) represent the overall toxicity status of the soil [23]. The PLI value integrates all the determined elements for each sampling site, according to the equation [19,23,37]:

$$PLI = (CF_1 CF_2 CF_3 \dots CF_n)^{1/n} \quad (4)$$

where: CF represents the value of the contamination factor for each element; n represents the number of the determined elements.

According to the values of PLI , the assessed pollution of soil with trace elements is classified as follows: $PLI < 0.7$, unpolluted; $0.7 \leq PLI < 1$, slightly polluted; $1 \leq PLI < 2$, moderately polluted; $2 \leq PLI < 3$, severely polluted; $PLI \geq 3$, heavily polluted soil [19].

Nemerow pollution index (NI) is used for comprehensive evaluation of the soil quality, highlighting the influence of the element with the highest contamination factor (CF_{max}) [37,38] and it is calculated by the following equation [26]:

$$NI = \sqrt{\frac{CF_{mean}^2 + CF_{max}^2}{2}} \quad (5)$$

where: CF_{mean} is the mean value of contamination factors calculated for all the investigated elements; CF_{max} is the maximum value of the contamination factors calculated for all the investigated elements.

According to the values of NI , the quality of the soil is classified as: $NI < 0.7$, safe; $0.7 < NI \leq 1.0$, precaution; $1.0 < NI \leq 2.0$, slightly polluted; $2.0 < NI \leq 3.0$, moderately polluted; $NI > 3.0$ seriously polluted [26,38].

Improved (modified) Nemerow index (INI) more accurately reflects the state of the soil by substituting other similar pollution indices, such as CF with the I_{geo} . The following equation was used for INI calculations [27,31,39]:

$$INI = \sqrt{\frac{I_{geo-max}^2 + I_{geo-ave}^2}{2}} \quad (6)$$

where: $I_{geo-max}$ is the maximum value of all the I_{geo} values and $I_{geo-ave}$ is the average value of the I_{geo} values calculated for all the investigated elements.

The values of INI are used for soil classification into following classes: $INI < 0.5$, uncontaminated; $0.5 \leq INI < 1$, uncontaminated to moderately contaminated; $1 \leq INI < 2$, moderately contaminated; $2 \leq INI < 3$, moderately to heavily contaminated; $3 \leq INI < 4$, heavily contaminated; $4 \leq INI < 5$, heavily to extremely contaminated; $INI \geq 5$, extremely contaminated soil [31,48]. It should be noted that INI is considered as considerably more accurate than the NI value for the assessment of environmental contamination risks [31].

3. RESULTS AND DISCUSSION

3. 1. Descriptive statistics for concentrations of elements in soil

Descriptive statistics of the obtained concentrations of trace elements in the soil samples are summarised in Table 2.

The mean concentrations of As, Cu, Pb and Zn in the soil of the study area were several times higher than the world average values, while the content of Al and Fe were within these values or a little higher [49]. Classification of the soil from the study area depending on the LVs and RVs [42], is presented in Table 3 and in Supplementary material (Fig. S1).

Table 2. Results of descriptive statistical analyses of Al, As, Cu, Fe, Pb and Zn concentrations in soils sampled at the 14 sites compared to the corresponding world average values

Element	Concentration, mg kg ⁻¹				Standard deviation	World average soil concentrations, mg kg ⁻¹ [49]
	Range	Min	Max	Mean		
Al*	2.85	3.15	5.99	4.75	0.92	1.00-5.00
As	155.30	9.67	164.98	70.29	54.56	6.83
Cu	2543.11	28.41	2571.52	708.14	651.52	38.9
Fe*	4.11	2.57	6.68	3.97	1.11	3.50
Pb	100.99	18.59	119.58	60.00	28.30	27.00
Zn	238.90	65.35	304.25	149.89	70.04	70.00

*concentration in %

The concentrations of As and Cu in soil samples exceeded the corresponding LVs at three and two sampling sites, respectively, while the RVs were exceeded at seven and eleven sampling sites, respectively. According to the exceedances of the RVs for As (up to 3 times) and Cu (up to 13 times), fundamental functions of the soil at most sampling sites were seriously disturbed and require remediation and other recovery measures. The exceedances of the LVs for Pb and Zn were recorded at three and six sampling sites, respectively, while exceedances of the RVs were not found for these metals. The limit and remediation values for Al and Fe were not defined by the Serbian Regulation. According to the LVs and RVs, the soil from the B site was classified as unpolluted in regard to all the studied elements. Soils from the sampling sites UI, U and SU, which were closest to the copper smelter and under the influence of dust from the flotation tailing ponds, were classified as polluted. The obtained concentration ranges of the investigated elements in the sampled soil (Table 2), as well as the exceedances of the RVs and LVs, indicated that the hazardous substances released from the copper smelter and dispersed from the flotation tailing ponds had the highest influence on the soil pollution in the study area.

Table 3. Classification of soil from the study area compared to the limit and remediation values

Element	LV / mg kg ⁻¹ [42]	RV / mg kg ⁻¹ [42]	Unpolluted soil	Polluted soil	
				C > LV	C > RV
As	29	55	I1, I3, TR, B	I2, I4, I5	UI, U, SU, R1, R2, T1, T2
Cu	36	190	B	I1, I2	UI, U, SU, I3, I4, I5, R1, R2, T1, T2, TR
Pb	85	530	I1, I2, I3, I4, I5, R1, R2, T1, T2, TR, B	UI, U, SU	/
Zn	140	720	I1, I2, I4, I5, R1, R2, T2, B	UI, U, SU, I3, T1, TR	/

/ - no exceedances of the RVs

3. 2. Assessment of the soil quality using single and integrated pollution indices

The influence of pollution from anthropogenic sources on the soil contamination around the mining and metallurgical area was evaluated by using the selected single and integrated indices of contamination, as stated in Section 2 (Materials and methods).

3. 2. 1. Geo-accumulation index (I_{geo})

The I_{geo} values for Al, As, Cu, Fe, Pb, and Zn in the soil at 13 sampling sites are shown in Figure 2. Based on the calculated I_{geo} values, soil in the study area was the most polluted with Cu, reaching the categories of heavily to extremely polluted soil at UI site, while extremely polluted soil was noted at five sites. The highest I_{geo} values for As were in the category of heavily polluted at UI, R2, T1 and T2 sites and moderately to heavily polluted soil at three sites. Moderate to heavy pollution of soil with Pb was noted at UI site. Regarding I_{geo} values, the most endangered sites with Cu, As and Pb were in immediate proximity to the copper smelter (*i.e.* UI, U, SU) and in the direction of the prevailing winds (*i.e.* R1, R2, T1, T2), indicating a strong influence of the industrial pollution. Also, it is important to emphasise that ore mining processes have less influence on the environmental pollution compared to the smelting of copper concentrates. According to I_{geo} values, soil from the study area was unpolluted to moderately polluted with Zn,

unpolluted and unpolluted to moderately polluted with Fe, while soil pollution with Al was not found. A lower category of the soil pollution with Fe and Al could indicate different origin of these elements compared to Cu, As and Pb.

In the study by Okonkwo *at al.* [19], the I_{geo} values for soils sampled around pegmatite mining sites showed that soil was unpolluted to heavily polluted by Zn, unpolluted to moderately polluted with Pb, while the I_{geo} values for Cu indicated that soils were in the category of unpolluted soil or moderately to heavily polluted soil, which was the consequence of the application of sewage sludge and copper containing fungicides. In another study [43], the I_{geo} values, calculated by using the median values of the examined elements in soils sampled in the surroundings of a tailing pond, indicated minimal soil pollution with As, Pb and Zn, whereas the sampled soil was at least moderately polluted with Cu. The highest values of I_{geo} suggested high pollution with Cu [43]. Contrary to that, based on the I_{geo} classification, the studied soils from an industrial area were not contaminated with Cu, Pb, Zn and Ni [33]. According to the I_{geo} values in an urban-rural area, low to moderate contamination with Cu, Pb and Zn was found [50]. Compared to these studies, the mean I_{geo} values for fertile agricultural soils were negative, indicating that the soils in this area were unpolluted with Al, As, Fe, Cu, Pb and Zn [24].

Based on the presented studies and results obtained here for I_{geo} , it could be observed that the soil from our study area was more polluted, especially with Cu.

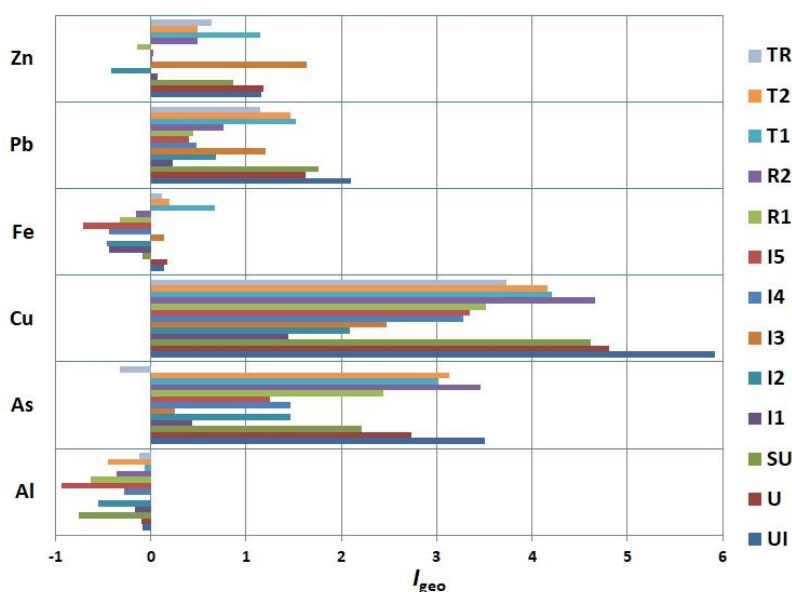


Figure 2. Geo-accumulation index (I_{geo}) values for Al, As, Cu, Fe, Pb and Zn in soils at 13 sampling sites in the study area

3. 2. 2. Enrichment factor (EF)

Calculated EF values for As, Cu, Fe, Pb, and Zn in the soil are presented in Figure 3. The most pronounced enrichment of the soil was noted for Cu. The soil at the sites UI and SU were extremely highly enriched, very highly enriched at the three sites (U, R2 and T2), while significantly enriched soil with Cu was noted at almost all the other sites. Significant enrichment was the highest category of soil enrichment with As and it was noted at the sites UI, U, SU, R1, R2, T1 and T2, as well as for Pb at the SU site. Enrichment of soil with Zn was minimal to moderate. The EF values for Fe in the soil at all the sampling sites were < 2 , indicating that there was no enrichment with Fe and the origin of this element could be predominantly natural in the study area. EF soil index also revealed that enrichment of the examined soil was the highest with As, Cu and Pb, similar to the observations obtained with I_{geo} index.

Soil contamination in the vicinity of a former Zn-Pb ore treatment plant was estimated in literature [36]. The mean EF values for Cu and Fe in the topsoil were in the moderate contamination category, soil enrichment with Zn was high, while soil was in the extremely high category of contamination with Pb. In another study [43], based on the values of EF for Zn, As, and Pb in the soil from the surroundings of the tailing ponds, the soil was in the category of deficiency to minimally enriched, while there was moderate enrichment of soil with Cu. Very high enrichment with As ($EF = 29.5$), significant

enrichment with Cu ($EF = 6.1$), and deficiency to minimal enrichment with Zn ($EF = 1.4$) and Pb ($EF = 1.1$) were noted around Khatoon Abad Cu smelter (SE Iran) [10]. Such high values of EF for As and Cu confirmed the negative influence of the anthropogenic emissions of the Cu-smelter, which was in accordance with the results obtained for the soil sampled in our study. Varol [24] reported that the mean value of EF for As indicated moderate enrichment of the soil, while minimal soil enrichments were noted for Cu, Fe, Pb and Zn, in the Harran Plain located in Şanlıurfa province in Turkey.

Considering the data reported in the literature, it can be observed that the enrichment of soil with Cu was higher in our research, while the EF value for As was higher in the study of Forghani *et al.* [10]. The values of EF for Pb, Zn and Fe were higher in our study compared to the literature data.

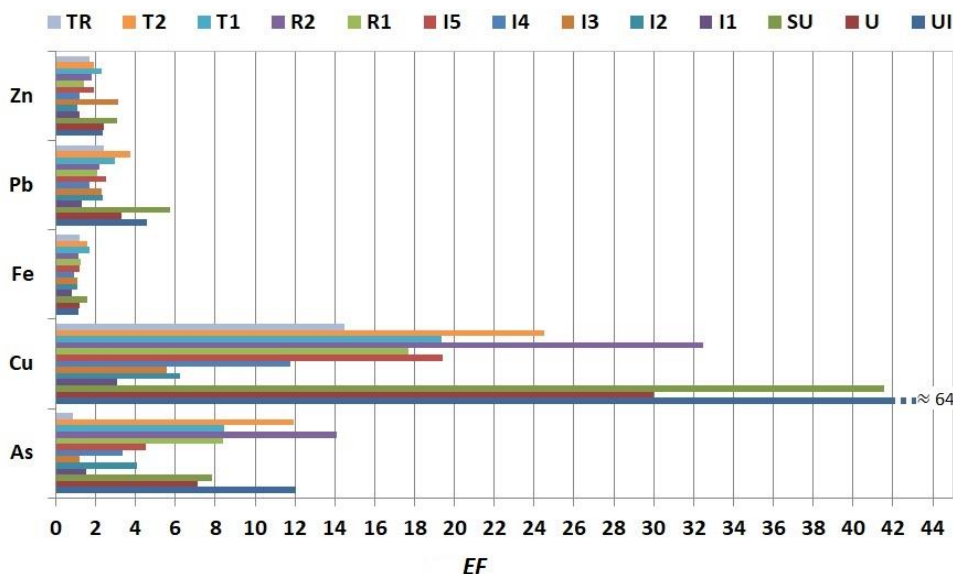


Figure 3. Enrichment factor (EF) values for As, Cu, Fe, Pb and Zn in the soils at 13 sampling sites in the study area

3. 2. 3. Contamination factor (CF)

A comparative presentation of CF values for Al, As, Cu, Fe, Pb, and Zn in the soil, depending on the sampling site is shown in Figure 4. The calculated CF s for Cu suggested very high contamination at almost all the sites and considerable contamination at site I1. The noted CF values for As, similarly as for Cu, were in the highest category of contamination, *i.e.* very high contamination at the sites UI, U, SU, R1, R2, T1 and T2, and considerable contamination at the sites I2, I4 and I5, while moderate contamination was obtained for three sites. Also, CF values for Pb indicated very high contamination at the site UI and considerable contamination at the sites U, SU, I3, T1, T2 and TR, and moderate contamination level at the remaining sites. Considerable soil contamination with Zn was noted at the sites UI, U, I3 and T1, while in other places soil contamination was moderate. The highest values of CF highlighted the strong negative influence of industrial activities, as well as the influence of prevailing wind on pollution dispersion, which was in accordance with the obtained values for I_{geo} and EF . The CF values for Al and Fe were predominantly in the category of moderate and low contamination, similarly to the data observed for I_{geo} and EF .

In a study on a mining area, the CF values indicated that soil contamination was low to considerable for Zn (0.54 to 4.00), and low to very high for Fe (1.00 to 11.34), Pb (0.70 to 10.00), and Cu (0.66 to 6.46) [19], which were similar to the results obtained in the present study. In another study [23], the CF values indicated that the soil contamination varied from low to considerable level, in a suburban zone around brick kilns (Jhenaidah District, south-western Bangladesh). However, the level of soil contamination with As was low, while the level of soil contamination with Al, Fe, Cu, Pb and Zn was low to moderate. In yet another research [26] the CF values obtained for Cu and Pb showed low to considerable contamination of soil inside the Vesuvius National Park, suggesting the influence of vehicular exhaust emissions in the tourist zone. Varol *et al.* [24] reported moderate contamination of soil with As, while they observed low contamination with Cu, Pb and Zn in agricultural area in Şanlıurfa province in Turkey. Low soil contamination with



Cu and Zn, and moderate contamination with Pb was noted at Sargodha, one of the most productive agricultural areas of Pakistan, with a fast-growing industrial setup [47].

Generally, in our study, the calculated values of *CF* for As, Cu, Pb and Zn were higher compared to the corresponding values of *CF* reported in the literature regarding different examined areas.

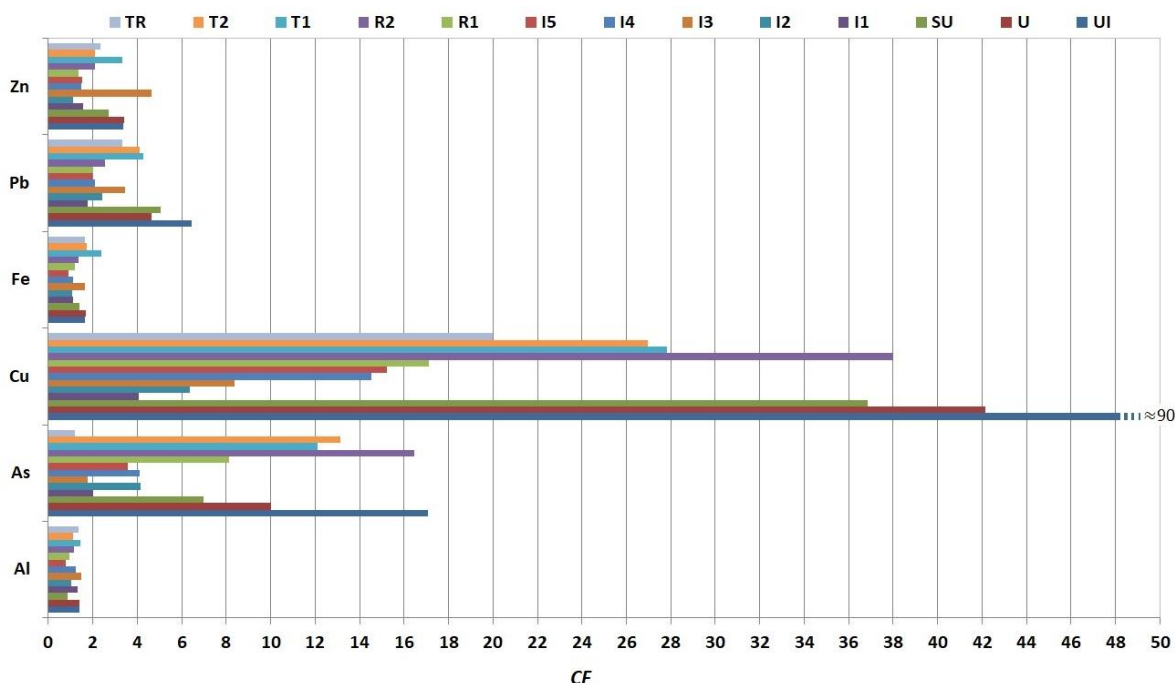


Figure 4. Contamination factor (*CF*) values for Al, As, Cu, Fe, Pb and Zn in the soils at 13 sampling sites in the study area

3. 2. 4. Pollution load index (PLI)

The overall soil contamination with the studied elements was estimated using *PLI* values shown in Figure 5. The obtained values of *PLI* ranged from 1.8 at the site I1 to 6.5 at the site UI. According to the *PLI* classification, the site I1 belongs to the moderately polluted, the sites I2, I3, I4 I5, R1 and TR are severely polluted, while the sites UI, U, SU, R2, T1 and T2 are in the category of heavily polluted soil. Within the elements used for the calculation of *PLI* values, As and Cu are the main pollution contributors.

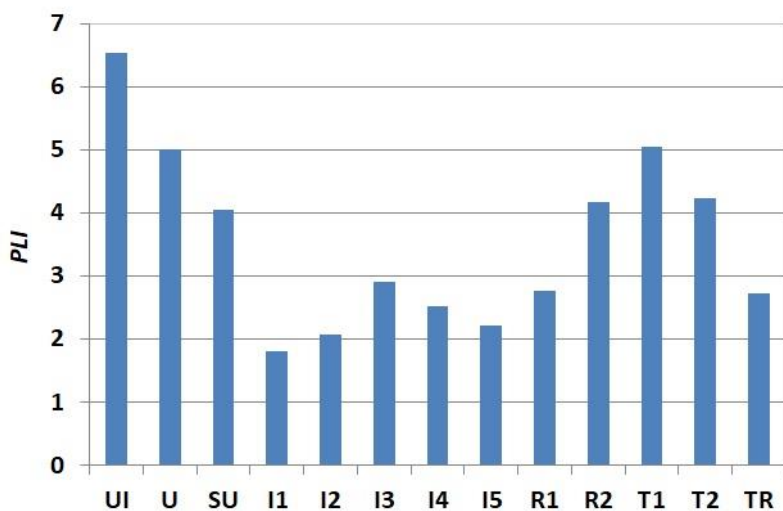


Figure 5. Pollution load index (*PLI*) values for the soils at 13 sampling sites in the study area



Based on the results obtained in the study by Yang *et al.* [38], the values of *PLI* for the urban soil ranged from 0.46 to 2.78, with a mean value of 0.96. The *PLI* values for soil samples from different sampling sites were reported to range from 0.8 in a nature reserve park to 2.3 in a lead/zinc mining region [37]. Low pollution levels were noted in the background area, while in the agricultural, industry, salt-lake and urban area, pollution was mainly moderate. Pollution in the mining area was the most serious [37]. The investigation of Okonkwo *et al.* [19] showed that *PLI* for soil at all the locations in the pegmatite mining area were in the range from 2 to 14, suggesting severe to heavy pollution of the soil, as in our study, but their calculated values were several times higher.

3. 2. 5. Nemerow index (NI)

The *NI* values of the examined elements in the soil samples were very high, ranging between 3.2 at the site I1 and 65.6 at the site UI (Fig. 6), indicating serious pollution at the sampling sites affected by pollution in the study area.

A study carried out in Peru investigated the area polluted by former mining activities. The *NI* values of trace elements in the soils from the leach pad area were very high, ranging between ~31 and 345, and in the soil from the mine dump area, they ranged between 28 and 211, indicating serious contamination at both sites [31], similarly as in our study, but with several times higher *NI* values. Contrary to that, in the study performed by Cui *et al.*, [20] at the old industrial area, the *NI* values ranged from 0.23 to 0.91 and from 0.17 to 0.62, indicating that soils from all the sampling sites were safe. In a study conducted in the Vesuvius National Park, the *NI* values ranged from ~1 and ~4, indicating slight to serious pollution of the soil [26]. The highest values of *NI* were noted at the end of the intense touristic activity (from April to October) that occurred in the park. In the soil from the urban Zhejiang Province located in south-eastern China, the *NI* values from all the evaluated sites were reported to range from 0.79 to 8.92, with a mean value of 1.74. Most of the values were in the category of precaution (15 %) and slightly polluted (64 %), while approximately 8 % of the sites had the $NI > 3$, which indicated serious pollution of the soil [38]. *NI* values of elements in soil sampled from different zones in the northeastern Qinghai-Tibet Plateau, were unexpectedly high, ranging from 1 to 378 [37]. At 58 sampling sites, soil pollution was high, at 21 sites, the samples showed moderate *NI* values, and at 48 sites, the samples showed low pollution level, while pollution of soil from the background area was low to moderate [37].

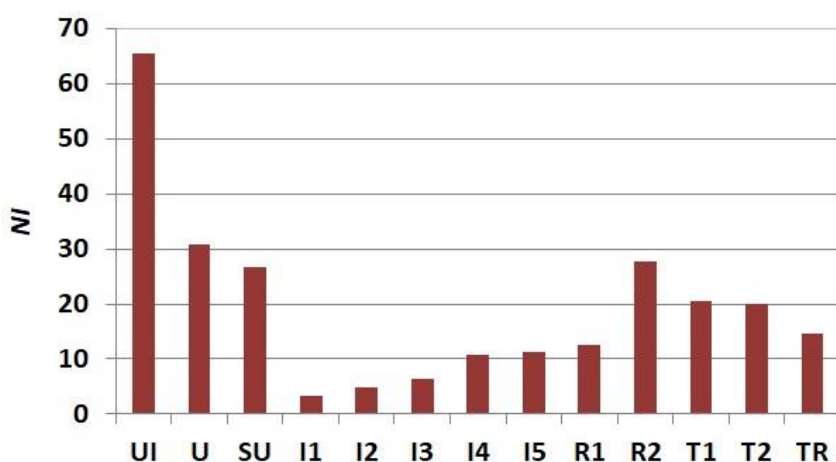


Figure 6. Nemerow index (*NI*) values for the soils at 13 sampling sites in the study area

PLI and *NI* showed the same trends with the highest values in the soil at the UI and the lowest at the site I1, indicating that the sites which were in the vicinity of the copper smelter and flotation tailing ponds were more polluted with the investigated elements. The sampling site I1 was located on the edge of the open pit, indicating that ore mining processes had a lower influence on the element concentration in the soil, compared to the smelting of copper concentrate and ore processing.

3. 2. 6. Improved Nemerow index (INI)

According to the calculated *INI* values (Fig. 7), the level of soil contamination was relatively high, ranging from moderately contaminated at the sites I1, I2 and I3, moderately to heavily contaminated at the sites I4, I5, R1 and TR, heavily contaminated at the sites U, SU, R2, T1 and T2, and heavily to extremely contaminated at the site UI. These data showed that soils from the most sampling sites in the study area were heavily contaminated.

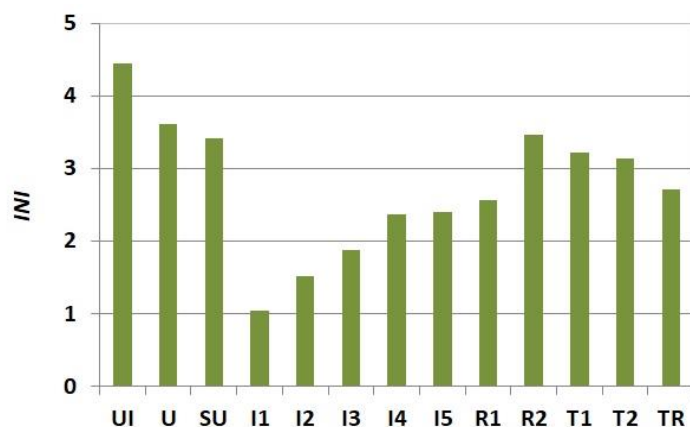


Figure 7. Improved Nemerow index (*INI*) values for the soils at 13 sampling sites in the study area

In a study in literature [31], the average *INI* values indicated extreme contamination (>5.0) and heavy to extreme contamination (4.67) of the soils from the mining area. Such high contamination indices highlighted the importance of effective contamination control and remediation. In the study by Santos-Francés *et al.* [48], taking the *INI* values into account, the soil was moderately to heavily contaminated, in the mining areas of La Zanja and Colquirrumi (Department of Cajamarca) and Julcani (Department of Huancavelica).

The calculated values of *INI* in the literature were similar as in our study, emphasising the effect of the industrial processes on the environmental contamination.

4. CONCLUSIONS

The purpose of this study was to provide information about the level of soil contamination with Al, As, Cu, Fe, Pb and Zn in the area endangered by pollution from the processes of copper mining and smelting. The mean concentrations of As, Cu, Pb and Zn in the soil of the study area were several times higher than the world average values. Concentrations of As and Cu in the soil exceeded the corresponding *RVs* at almost all the sampling sites, even up to 3 and 13 times, respectively. The exceedances of *LVs* for Pb and Zn were noted only at the most polluted sites. According to the single and integrated pollution indices, the quality of the soil was determined. Calculated values of single pollution indices provided insight into the degree of pollution by a specific element, while integrated pollution indices indicated cumulative pollution by the elements of interest. Based on the values of I_{geo} , *EF* and *CF* (single pollution indices) it was concluded that the highest pollution was with Cu and As, in the soils sampled in the close vicinity of the mining-metallurgical complex and flotation tailing ponds (UI, SU, U sites), as well as at the sites in the prevailing wind directions (R1, R2, T1, T2). The sampling sites significantly enriched with As actually indicated that the polluting substances containing As had the ability of dispersion over a large area due to their nature and properties, as well as that flotation tailings also were the source of particulate matters rich in As content. Mainly moderate contamination of the soil was observed in the case of Pb and Zn. The soil in the study area was not polluted with Al and Fe, suggesting their predominantly natural occurrence in the soil. The values of the *PLI*, *NI* and *INI* (integrated pollution indices) also showed serious cumulative contamination of the soil with the analysed elements from the sites closest to the copper smelter and in the prevailing wind directions, suggesting the influence of the anthropogenic activities in the study area. The main contributors of the soil pollution were Cu and As, which was expected due to the smelting of sulphide copper ore

with a high quantity of these elements. The results of this investigation indicate that the soil in the vicinity of the metallurgical complex, as well as from the sampling sites at which the prevailing winds bring the pollution from the copper smelter and flotation tailing ponds show the most serious pollution. These areas need effective environmental pollution control and prevention, as well as soil remediation particularly for highly endangered sites.

SUPPLEMENTARY MATERIAL

Additional data are available electronically at <https://www.ache-pub.org.rs/index.php/HemInd/article/view/1133>, or from the corresponding author on request.

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Indeksi zagađenja kao koristan alat za sveobuhvatnu procenu stepena kontaminacije zemljišta u blizini rudarskih i metalurških kompleksa

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(Naučni rad)

Izvod

U ovom radu procenjen je nivo zagađenja zemljišta elementima Al, As, Cu, Fe, Pb i Zn na 14 mesta u gradu Boru i okolini u oblasti rudarstva i metalurgije bakra, kao i procena nivoa zagađenja pojedinačnim i integrisanim indeksima zagađenja. Značaj pojedinačnih indeksa zagađenja pruža informaciju o zagađenju određenim elementom, dok integrisani indeksi zagađenja pružaju uvid u kumulativno zagađenje ispitivanim elementima. Prosečne koncentracije As, Cu, Pb i Zn u zemljištu bile su nekoliko puta više od svetskih prosečnih vrednosti. Prekoračenja remedijacionih vrednosti bila su najizraženija za As (na sedam mesta) i Cu (na jedanaest mesta), više od 3 i 13 puta, redom. Prema geoakumulacionom indeksu, faktoru obogaćenja i faktoru kontaminacije, najveća kontaminacija zemljišta je bila sa As i Cu, naročito na urbano-industrijskom mestu. Indeksom opterećenja zagađenjem, Nemerovim indeksom zagađenja i poboljšanim (modifikovanim) Nemerovim indeksom potvrđeno je da su najviše kontaminirana zemljišta bila na mestima u blizini metalurškog kompleksa, flotacijskih jalovišta, kao i sa mesta koja se nalaze na pravcima najučestalijih vetrova u odnosu na manje zagađena zemljišta koja su bila pod uticajem procesa iskopavanja rude. Područjima pogođenim ozbiljnom kumulativnom kontaminacijom poreklom iz pirometalurške proizvodnje bakra potrebna je kontinualna prevencija zagađenja, monitoring i remedijacione mere.

Ključne reči: Zagađenje životne sredine; kvalitet zemljišta; pojedinačni i integrisani indeksi zagađenja zemljišta

