COMPARISON OF BROWN RENDZINA AND COLLUVIAL SINKHOLE SOIL IN THE AREA OF THE NATIONAL PARK "UNA" WITH SPECIAL EMPHASIS ON THE DISTRIBUTION OF CADMIUM, NICKEL AND ARSENIC

ORIGINAL SCIENTIFIC PAPER

Fatima Muhamedagić[1]✉, Ekrem Pehlić[2]

DOI: 10.5281/zenodo.6919834

RECEIVED 2022-02-02 **ACCEPTED** 2022-03-04

[1] University of Bihac, Biotehnical faculty, Luke Marjanovica bb, 77000 Bihac, Bosnia and Herzegovina
[2] University of Bihac, Faculty of Health Studies, Nositelja hrvatskog trolista 4, 77000 Bihac, Bosnia and Herzegovina
✉ fatima.muhamedagic@gmail.com

ABSTRACT:
The paper presents the results of a comparison of brown rendzina and colluvial sinkhole at approximately the same altitudes and the same land use. The sites were located in the Kalati area within the National Park „Una". More precisely, the places referred to the top of the sinkhole (brown rendzina - top) and the bottom of the sinkhole (colluvial soil - bottom). The main objective of the research, in addition to the physical and chemical parameters of soil quality, was to determine the distribution of the total content of cadmium (Cd), nickel (Ni) and arsenic (As). The total content of these elements werw observed in composite samples at two depths of 0 – 10 and 0 – 20 cm and horizons profile. Their total content was measured by atomic adsorption spectrometry – AAS. The results were statistically analyzed using Kruskal - Walisovog test at the significance level of $p \leq 0.05$ using correlation coefficient χ^2. The results showed a single legality of the distribution of Cd, Ni and As in samples of soil profile, while the average soil samples showed unique legality of the distribution of observed elements.

KEYWORDS: brown renzina, colluvial soil, distribution, cadmium, nickel, arsenic

INTRODUCTION

The entire area of the National Park is located in the Federation of BiH within the Una-Sana Canton and belongs to the municipality of Bihac.

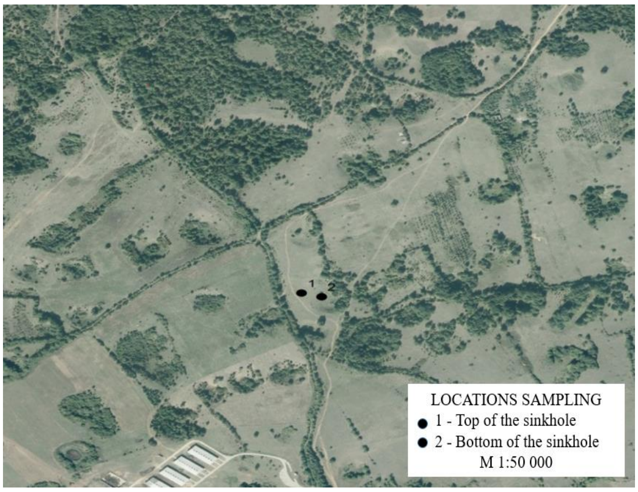

Figure 1. Locality Kalati in the National Park

National Park Una includes the area of the canyon part of the upper course of the river Una upstream from Lohovo, the area of the canyon part of the lower course of the river Unac to its confluence with the river Una, and the interspace between Una and Unac on a total area of 19,800 ha [1]. National Park Una continues to arouse the interest of many scientists and researchers of various profiles [2]-[4]. There are researches especially from the aspect of distribution of toxic elements under the influence of wind, different way of land use and others [5]-[7]. However, the transfer factors of toxic elements can also be colluvial processes in sinkholes. The colluvial process is a geomorphological process that occurs under the direct action of gravity [8]. Sinkhole is the most diagnostic feature of karst terrains [9]. There are several types of sinkholes [10]. This paper presents the results of a comparison of different soil type variants, ie. the association of brown rendzina (top of the sinkhole – TOP) – the first variant and the colluvial soil of the sinkhole (bottom of the sinkhole – BOTTOM) – the second variant. These variants are on the same site are the same land use and with the same sampling depths of average samples. The study has placed a special emphasis on the distribution of the total content of cadmium (Cd), nickel (Ni) and arsenic (As) in the average samples (0 – 10 and 0 – 20 cm) and by depth of the soil profile. Both variants of the locality were from the association of the soil of the brown rendzina with the colluvial soil of sinkholes, of different depths, approximately the same altitudes (501 – 509 m), the same pressure: 812,73 hPa, the same way of use – neglected natural pastures and diferent wind speed (TOP: 12 m/s, BOTTOM: 5 m/s). The appearance of the place is shown in Figures 1.

According to pedological maps of BiH [11] and pedological interpreters (Bihać 3), soils in selected localities alternate by different types (variants) and depths. Lighter mechanical composition (TOP). In the surface layers, the mechanical composition is lighter, and at the bottom it is heavier (BOTTOM). The soils are weakly humus, poor in phosphorus (P_2O_5) and calcium carbonate ($CaCO_3$), potassium is medium (K_2O). Water capacity (K_w) is medium for air (K_a) is good. Their color is usually brown and yellow-brown.

MATERIALS AND METHODS

This paper presents results of the field research, sampling and laboratory testing. All analyzes of soil samples were determined according to standardized methods: Mechanical composition (Modified method B pipettes, ISO 11277); Structure (Method by Sekera); The rights specific gravity, Porosity (Conventional method, ISO 11508); The volume spacific gravity (Gravimetric method by Kopecky, ISO 11272); Actual humidity, Capacity of soil for water and Air capacity (Gravimetric method, ISO 11465); Humus (Spectrophotometric method, ISO 10694); $CaCO_3$ (Volumetric method ISO 10693); pH (Electrometric method, ISO 10390); El.conductivity (Electrometric method, ISO 11265); P_2O_5 and K_2O (Al method, ISO 19730) and The total content of metals and metalloids in the soil (Extraction HCl/HNO_3 = 3:1, ISO 11466). A total of two pedological profiles were opened at the sites of research and average samples were taken from two soil depths: 0 – 10 cm and 0 – 20 cm.

Preparation of soil samples was carried out at the Biotechnical Faculty of the University of Bihac. All analyses of soil samples were carried out in the laboratory of the Institute of Soil Science, Agrochemistry and Reclamation of the Faculty of Agricultural and Food Sciences, University of Sarajevo, except for the content of arsenic that was carried out at the Faculty of Agriculture, University of Zagreb, and the Institute for Medical Research and Occupational Health in Zagreb.

Statistical analysis was performed by the Kruskal - Wallis test at the level of significance of $p \leq 0.05$. To determine the relation and distribution of Cd, Ni and As in the soil we used the correlation coefficient (χ^2) [12]. Data were statistically analyzed using SPSS 17. The maximum allowable amounts of total contents were determined in accordance with the current regulations [13] in relation to the soil texture (MRL) and the use of soil in organic farming (MRL-OF). Used are orthophoto imagery in the scale of 1:5000 and 1:75000, topographic maps and GIS.

RESULTS AND DISCUSSION

Results of the study on the comparison of different types (variants) of soil provide:

Description of the soil profile of the brown rendzina at the top of the sinkhole (TOP) (Figure 2)
Site: Kalati – brown rendzina (top of the sinkhole – TOP) (N 44° 34.066´, E 16° 03.434´)
Vegetation: natural pasture
Use: pasture
Parent substrate: dolomite
Ah 0-40 cm: surface layer medium overgrown with grass roots, dark brown in color, coarsely crumbly structure and according to the texture – clay loam (sand = 28,7 %, powder = 40,5 %, clay = 30,8 %). Skeleton present.
C > od 40 cm: coarse dolomite

Description of the colluvial soil profile of the sinkhole (BOTTOM) (Figure 3)
Site: Kalati – colluvial soil (bottom of the sinkhole – BOTTOM) (N 44° 34.063´, E 16° 03.448´)
Vegetation: natural pasture
Use: pasture
Parent substrate: dolomite

I 0-19 cm: layer overgrown with grass roots and weeds, dark brown in color, finely crumbly structure. Texture – powdery clay loam (sand = 16.6 %, powder = 43.5 %, clay = 39.9 %)

II 19-42 cm: in the layer still evident roots of weed plants, dark yellow color, layer moist, crumbly structure and texture – powdery clay (sand = 15.3 %, powder = 41.5 %, clay = 43.2 %).

III 42-64 cm: yellow-brown color, pea-shaped structure, the layer dries a little in relation to the upper one, evident smaller parts of the skeleton with a diameter of about 2 cm, texture – powdery clay to clay (sand = 13.3 %, powder = 39.3 %, clay = 47.4 %).

IV 64-120 cm: yellow-brown color, layer moist and sticky, visible larger parts of the skeleton 5 - 8 cm, texture – clay (sand = 11.7 %, powder = 34.7 %, clay = 53.6 %).

Figure 2 – 3. Profiles of soil at the sites Kalati TOP (left) and BOTTOM (right)

Physical properties of soil at the sites Kalati of TOP and BOTTOM

The stability of the structural aggregates in the depth of the profile was very good to good. The mechanical composition in the surface layer was clay loam (TOP) and powdery clay loam (BOTTOM), in deeper layer – clay (BOTTOM). The soils were porous, with medium soil water and moderate soil air capacities.

Mean values of real and bulk specific density also indicated that the soils were porous. Overview of the physical properties of the soil is provided in Table 1.

Table 1. The physical properties of the soil profile ($\Sigma n_{TOP\text{-}BOTTOM}$ = 18)

Parameters		$X_{mv} \pm X_{se}$	min	max	V	σ
S	TOP	1.50 ± 0.22	1.00	2.00	0.30	0.54
	BOTTOM	2.33 ± 0.31	1.00	4.00	1.07	1.07
P_{sg} (g/cm^3)	TOP	2.65 ± 0.00	2.65	2.66	0.00	0.01
	BOTTOM	2.65 ± 0.01	2.62	2.68	0.00	0.02
V_{sg} (g/cm^3)	TOP	1.26 ± 0.02	1.22	1.30	0.00	0.03
	BOTTOM	1.17 ± 0.31	1.01	1.28	1.09	1.09
P (%)	TOP	50.13 ± 0.59	49.00	51.00	1.05	1.02
	BOTTOM	54.95 ± 0.86	50.0	59.90	8.26	2.87
K_w (%)	TOP	41.73 ± 0.29	41.30	42.30	0.26	0.51
	BOTTOM	43.66 ± 0.80	40.60	47.70	7.13	2.67
K_a (%)	TOP	9.40 ± 0.11	9.20	9.60	0.04	0.20
	BOTTOM	11.29 ± 0.76	8.00	16.40	6.49	2.54

TOP – brown rendzina; BOTTOM – colluvial soil; X_{mv} – mean value, X_{se} – statistical error, min – minimum value, max – maximum value, V – variance, σ – deviation, n –numer of samples, S – soil structure, P_{sg} – the righs specific gravity, V_{sg} – the volume spacific gravity, P – porosity, K_w – capacity of soil for water, K_a – air capacity

Chemical properties of the soil profile and average samples 0 – 20 cm at the sites Kalati of TOP and BOTTOM

In open profiles the reactions of pH values for the TOP and BOTTOM of the sinkhole ranged from neutral to moderately alkaline. In the surface layer, the contents of K_20 ranged from good to poor supply (TOP) and from medium to poor (BOTTOM). The supply of P_2O_5 was generally weak (TOP and BOTTOM). The humus content was moderate to weak (3.9 to 1.9%). Total content of $CaCO_3$ was low with a tendency to decrease with profile depth. Mean values of measured electro-conductivity of the soil were not indicative of soil salinity at the profile.

Results of the observed chemical parameters in samples (0 – 20 cm) had similar characteristics as the parameters of the soil profile of the surface layer (Table 2 – 3).

Table 2. The chemical properties of the soil profile ($\Sigma n_{TOP\text{-}BOTTOM}$ = 18)

Parameters		$X_{mv} \pm X_{se}$	min	max	V	σ
pH_{H2O}	TOP	7.61 ± 0.22	6.73	8.12	0.30	0.55
	BOTTOM	7.85 ± 0.09	7.15	8.12	0.10	0.33
pH_{KCl}	TOP	6.39 ± 0.20	5.60	6.90	0.24	0.49
	BOTTOM	6.55 ± 0.07	5.97	6.72	0.06	0.25
EC (μS/cm)	TOP	142.61 ± 5.13	126.6	161.3	158.21	12.57
	BOTTOM	153.88 ± 3.71	131.5	175	165.56	12.86
K_2O (mg/100g soil)	TOP	11.10 ± 2.28	6.50	20.10	31.2	5.58
	BOTTOM	10.50 ± 0.64	7.20	15.50	4.96	2.22
P_2O_5 (mg/100g soil)	TOP	1.38 ± 0.17	0.88	1.85	0.17	0.41
	BOTTOM	1.92 ± 0.20	0.77	3.04	0.5	0.71
Humus (%)	TOP	2.88 ± 0.37	1.89	3.90	0.84	0.92
	BOTTOM	2.60 ± 0.18	1.64	3.50	0.38	0.62
$CaCO_3$ (%)	TOP	0.22 ± 0.04	<0.01	0.23	0.01	0.10
	BOTTOM	0.24 ± 0.13	<0.01	1.47	0.47	0.22

TOP – brown rendzina; BOTTOM – coluvial soil, n – numer of samples, X_{mv} – mean value, X_{se} – statistical error, min – minimal value, max – maximum value, V – variance, σ – deviation

Table 3. The chemical properties of the average samples 0 – 20 cm ($\Sigma n_{TOP\text{-}BOTTOM}$ = 18)

Parameters		$X_{mv} \pm X_{se}$	min	max	V	σ
pH_{H2O}	TOP	7.13 ± 0.12	6.63	7.40	0.09	0.30
	BOTTOM	7.23 ± 0.18	6.76	7.84	0.20	0.45
pH_{KCl}	TOP	6.00 ± 0.14	5.48	6.32	0.13	0,36
	BOTTOM	6.16 ± 0.20	5.57	6.85	0.25	0.5
K_2O (mg/100g soil)	TOP	16.06 ± 2.21	10.50	23.80	29.39	5.42
	BOTTOM	24.25 ± 4.30	14.20	35.80	111.1	10.5
P_2O_5 (mg/100g soil)	TOP	4.10 ± 1.39	1.28	9.54	11.64	3.41
	BOTTOM	2.01 ± 0.33	1.19	3.14	0.69	0.83
Humus (%)	TOP	3.50 ± 0.28	2.84	4.28	0.49	0.69
	BOTTOM	4.00 ± 0.74	0.63	5.70	3.33	1.82
$CaCO_3$ (%)	TOP	0.23 ± 0.27	0.00	0.71	0.14	0.37
	BOTTOM	0.27 ± 0.31	0.00	0.96	0.17	0.41

TOP – brown rendzina; BOTTOM – colluvial soil; n – numer of samples; X_{mv} – mean value; X_{se} – statistical error; min – minimal value; max – maximum value; V – variance; σ – deviation

<u>*Distribution of the total contents of Cd, Ni and As in average samples*</u>

In average samples at the sites of TOP and BOTTOM (0 – 10 and 0 – 20 cm), a significant difference ($p \leq 0.05$) in the total content of Cd, Ni and As was established (Table 4).

In the average samples at the sites TOP and BOTTOM (0 – 10 and 0 – 20 cm), the total mean contens Cd was lower than MRL and MRL-OF (TOP) but higher than MRL and MRL-OF (BOTTOM), the total mean contens As was between (TOP and BOTTOM) while the total mean contens Ni was above the allowable values of MRLand MRL-OF (TOP and BOTTOM). At the sites TOP and BOTTOM in depth 0 – 10 cm (Cd_{TOP} = 0.45 mg/kg; Cd_{BOTTOM} = 2.09 mg/kg; Ni_{TOP} = 78.58 mg/kg; Ni_{BOTTOM} = 83.11 mg/kg; As_{TOP} = 11.49 mg/kg; As_{BOTTOM} = 15.49 mg/kg), and the sites TOP and BOTTOM in depht 0 – 20 cm (Cd_{TOP} = 0.82; Cd_{BOTTOM} = 3.23; Ni_{TOP} = 78.24 mg/kg; Ni_{BOTTOM} = 85.54 mg/kg; As_{TOP} = 12.54 mg/kg; As_{BOTTOM} = 15.13 mg/kg). Generaly, the total content of analyzed elements at the bottom of the sinkhole was higher compared to the soils at the top of the sinkhole, which confirms the fact of possible deposition due to colluvial processes or carrying soil to the bottom of the sinkhole due to gravity. See in the charts 1 – 3.

Table 4. Distribution of the total contents of Cd, Ni and As in average samples

Observed		$X_{mv} \pm X_{se}$	min	max	V	σ	χ^2	p
Deph 0 – 10 cm (Σn = 9)								
Cd (mg/kg)	TOP	0,45 ± 0,02	0,43	0,49	0,00	0,03	3.97	$p \leq 0.05$
	BOTTOM	2,09 ± 0,00	2,09	2,10	0,00	0,01		
Ni (mg/kg)	TOP	78,58 ± 0,09	78,40	78,68	0,02	0,15	3.97	$p \leq 0.05$
	BOTTOM	83,11 ± 0,02	83,10	83,15	0,00	0,02		
As (mg/kg)	TOP	11,49 ± 0,02	11,45	11,52	0,00	0,03	3.97	$p \leq 0.05$
	BOTTOM	15,49 ± 0,01	15,47	15,50	0,00	0,01		
Deph 0 – 20 cm (Σn = 9)								
Cd (mg/kg)	TOP	0,82 ± 0,16	0,43	1,20	0,16	0,40	8.36	$p \leq 0.05$
	BOTTOM	3,23 ± 0,51	2,09	4,44	1,57	1,25		
Ni (mg/kg)	TOP	78,24 ± 0,15	77,90	78,68	0,14	0,38	8.36	$p \leq 0.05$
	BOTTOM	85,54 ± 1,08	83,10	88,00	7,04	2,65		
As (mg/kg)	TOP	12,54 ± 0,47	11,45	13,60	1,32	1,15	8.36	$p \leq 0.05$
	BOTTOM	15,13 ± 0,16	14,74	15,50	0,15	0,39		

TOP – brown rendzina; BOTTOM – colluvial soil; n – numer of samples; X_{mv} – mean value; X_{se} – statistical error; min – minimal value; max – maximum value; V – variance; σ – deviation; χ^2 – Kruskal – Wallis coefficient; p – level of significance

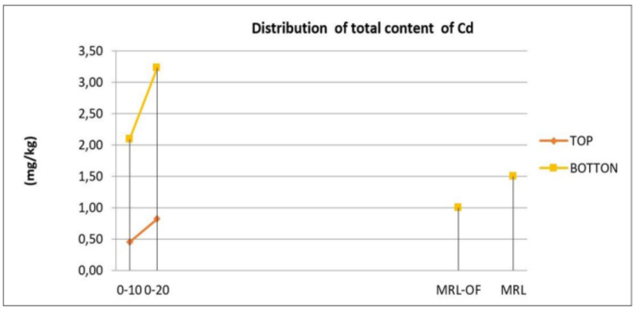

Chart 1. Distribution of the total content of Cd (0 – 10 and 0 – 20 cm)

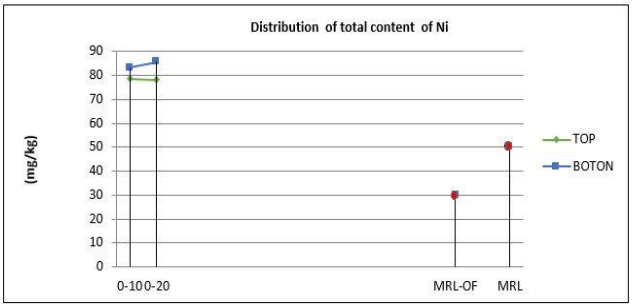

Chart 2. Distribution of the total content of Ni (0 – 10 and 0 – 20 cm)

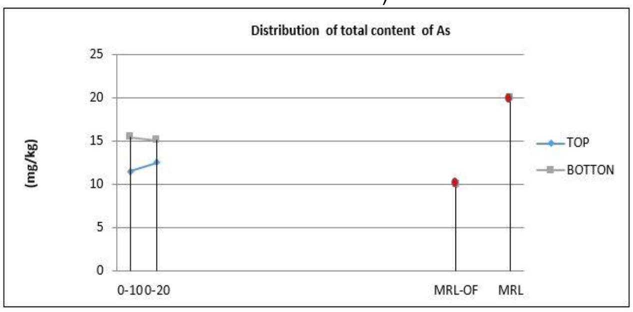

Chart 3. Distribution of the total content of As (0 – 10 and 0 – 20 cm)

<u>Distribution of the total content of Cd, Ni and As in the soil profiles</u>

Results of the analysed profiles TOP and BOTTON sites of the same use (natural pasture) have shown significant differences ($p \leq 0.05$) in concentrations of Cd, Ni and As (Table 5).

Table 5. Distribution of the total content of Cd, Ni and As in the soil profiles
($\Sigma n_{TOP-BOTTON}$ = 18)

Observed		$X_{mv} \pm X_{se}$	min	max	V	σ	χ^2	p
Cd (mg/kg)	TOP	0.99 ± 0.06	0.85	1.15	0.02	0.14	11.41	$p \leq 0.05$
	BOTTON	2.94 ± 0.15	2.35	3.50	0.27	0.52		
Ni (mg/kg)	TOP	72.37 ± 1.99	67.70	76.90	23.76	4.87	11.36	$p \leq 0.05$
	BOTTON	122.35 ±6.20	87.10	138.21	462.13	21.49		
As (mg/kg)	TOP	9.01 ± 0.67	7.45	10.58	2.69	1.64	11.38	$p \leq 0.05$
	BOTTON	16.35 ± 0.38	14.90	18.30	1.79	1.34		

TOP – brown rendzina; BOTTON – colluvial soil; n – numer of samples; X_{mv} – mean value; X_{se} – statistical error; min – minimal value; max – maximum value; V – variance; σ – deviation; χ^2 – Kruskal – Wallis coefficient; p – level of significance

Observed in general in the examined profiles at the TOP and BUTTOM sites, the total mean contents of Cd, Ni and AS in the profile at the bottom of the sinkhole was higher compared to the soil profile at the top of the sinkhole (Chart 4 - 6). In the examined profiles, the total mean Cd content was 66% higher, nickel 40% higher, and arsenic 46% higher at the bottom of the sinkhole (colluvial soil) compared to its top (brown rendzina). The total contents of Cd and Ni tended to increase and exceeded the allowable values (MRL and MRL-OF) while the content of As was between MRL and MRL-OF.

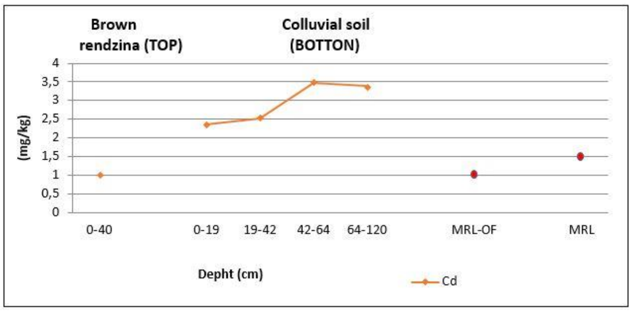

Chart 4. Distribution of the total content of Cd in soil profiles

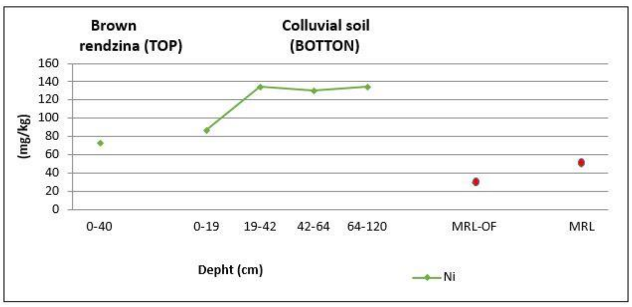

Chart 5. Distribution of the total content of Ni in soil profiles

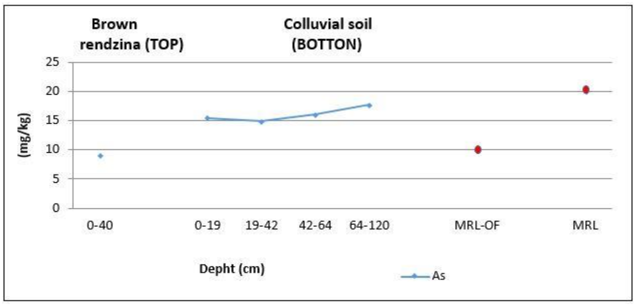

Chart 6. Distribution of the total content of As in soil profiles

CONCLUSIONS

At the sites of TOP and BOTTOM, according to the physical and chemical parameters of soil, stability of structural aggregates declines from good to very good; mechanical composition of the soil in the surface layer – clay loam (TOP) and powdery clay loam (BOTTOM), and in the deeper ones – clay (BOTTOM). The soils were porous, with medium soil water and moderate soil air capacities. In general, the pH values at the sites of TOP and BOTTOM ranged from neutral to moderately alkaline ($pH_{TOP\ in\ H20}$ 6.73 – 8.12; $pH_{TOP\ in\ KCl}$ 5.60 – 6.90; $pH_{BOTTOM\ in\ H20}$ 7.15– 8.12; $pH_{BOTTOM\ in\ KCl}$ 5.97 – 6.72) and average samples ($pH_{TOP\ in\ H20}$ 6.63 – 7.40; $pH_{TOP\ in\ KCl}$ 5.48 – 6.32; $pH_{BOTTOM\ in\ H20}$ 6.76 – 7.84; $pH_{BOTTOM\ in\ KCl}$ 5.57 – 6.85).

In the average samples (0–10 and 0–20 cm) at the sites, total mean content of Cd at the top of the sinkhole was lower than the permitted levels of MRL-OF and MRL, but at the bottom of the sinkhole the Cd content was higher than the permitted levels (Cd_{TOP} = 0.43 – 1.20 mg/kg; Cd_{BOTTOM} = 2.09 – 4.44 mg/kg).

The total mean content of Ni at the top and bottom of the sinkhole was above the permitted levels (Ni_{TOP} = 77.90 – 78.68 mg/kg; Ni_{BOTTOM} = 83.10 – 88.00 mg/kg), while the total mean content As was between the permitted levels (As_{TOP} = 11.45 – 13.60 mg/kg; As_{BOTTOM} = 14.74 – 15.50 mg/kg).

In the surface layer, content of P_2O_5 was low, while the K_2O content ranged from weak to moderate as in humus. Total content of $CaCO_3$ was low with a tendency to decrease with profile depth ($CaCO_{3TOP}$ <0.01 – 0.23 %; $CaCO_{3BOTTOM}$ <0.01 – 1.47 %). The measured levels of soil electrical conductivity were not indicative of the soil salinity (EC_{TOP} 126.6 – 161.3 μS/cm; EC_{BOTTOM} 131.5 – 175.0 μS/cm).

In the distribution of Cd, Ni and As in average samples (0 – 10 and 0 – 20 cm) at the top and bottom of the sinkhole, it was found that there is a significant difference ($p \leq 0.05$) in the total content of Cd, Ni and As.

In general, in the average samples (0 – 10 and 0 – 20 cm) there was a unique tendency to distribute (increase) the total content of Cd, Ni and As at the top and bottom of the sinkhole. This confirms the fact that due to colluvial processes, Cd, Ni and As are deposited at the bottom of the sinkhole. In the profiles at the sites (TOP and BOTTOM), significant differences ($p \leq 0.05$) were determined in the total content of Cd, Ni and As. At the sampling sites at Kalati at the top and bottom of the sinkhole, it was found that the total mean content of Cd, Ni and As at the bottom of the sinkhole was higher compared to the top of the sinkhole, with Cd by 79% with Ni by 9% and As by 9%.

In this research in the area of the National Park „Una“, a possible accumulation of Cd, Ni and AS was determined, which takes place under the influence of colluvial processes. Colluvial processes under the influence of gravity or relief carry the surrounding material from the top of the sinkhole to the bottom of the sinkhole.

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