

Research Article

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The application of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ methods in soil erosion research of Titel loess plateau, Vojvodina, Northern Serbia

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Abstract: Soil erosion is one of the largest global problems of environmental protection and sustainable development, causing serious land degradation and environmental deterioration. The need for fast and accurate soil rate assessment of erosion and deposition favors the application of alternative methods based on the radionuclide measurement technique contrary to long-term conventional methods. In this paper, we used gamma spectrometry measurements of ^{137}Cs and unsupported $^{210}\text{Pb}_{\text{ex}}$ in order to quantify the erosion on the Titel Loess Plateau near the Tisa (Tisza) River in the Vojvodina province of Serbia. Along the slope of the study area and in the immediate vicinity eight representative soil depth profiles were taken and the radioactivity content in 1 cm thick soil layers was analyzed. Soil erosion rates were estimated according to the profile distribution model and the diffusion and migration model for undisturbed soil. The net soil erosion rates, estimated by ^{137}Cs method range from $-2.3 \text{ t ha}^{-1} \text{ yr}^{-1}$ to $-2.7 \text{ t ha}^{-1} \text{ yr}^{-1}$, related to the used conversion model which is comparable to published results of similar studies of soil erosion in the region. Vertical distribution of natural radionuclides in soil profiles was also discussed and compared with the profile distribution of unsupported $^{210}\text{Pb}_{\text{ex}}$ measurements. The use of diffusion and migration model to convert the results of $^{210}\text{Pb}_{\text{ex}}$ activities to soil redistribution rates indicates a slightly higher net erosion of $-3.7 \text{ t ha}^{-1} \text{ yr}^{-1}$ with 98% of the sediment delivery ratio.

Keywords: radiocaesium, unsupported lead, gamma-spectrometry, soil erosion, gully erosion, Titel loess plateau

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1 Introduction

Land degradation is a broad term that relates essentially to the gradual loss of soil functions by the interaction of natural and anthropogenic factors [1]. Its consequences are reflected in the loss of fertility, desertification, vulnerability to floods, vulnerability to climate change and accelerated human activities. The complexity of the soil degradation phenomenon could be explained by categorizing the term in four basic groups: erosion (soil deposition), depletion (taking of fertile material), accretion (chemical changes in the form of accumulation of heavy metals) and compaction (physical disturbance) [1]. Every year about 10,000,000 ha of arable land on the global level is lost only by erosion, with several tens of times faster loss compared to the time it takes to renew [2]. The consequences of soil erosion are reflected in reduced agricultural productivity (on-site) and increased flooding of lake-river-sea systems due to intense sedimentation [3]. Furthermore, cur-

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rent land loss rates are unsustainable and can grow significantly in recent times due to significant changes in soil coverage and rainfall intensity as a matter of human impact and climate change. Therefore, the need to monitor the extent of land loss is highly justified. Conventional monitoring techniques have numerous limitations in terms of representativeness of data, temporal and spatial resolution, costs, long-term measurements with high uncertainties [4]. During the last fifty years, empirical and semi-empirical models of various complexities (e.g. USLE, EPIC, WEPP, RUSLE and EUROSEM) were designed to take into account the spatial and temporal pattern of variables, such as watershed topography, soil properties, land use and management, climate, as well as the relationship between them [5]. The resolution and reliability of input variables directly affect the precision of the applied model [6, 7]. Therefore, the utilization of fallout radionuclide tracers in soil, mostly bound to clay minerals and organic matter, such as the fission product ^{137}Cs and the naturally occurring excess lead $^{210}\text{Pb}_{\text{ex}}$ are being increasingly used for soil erosion and sediment assessment [7]. These radionuclides have been spread in the atmosphere and adsorbed on the surface layer of soil, and subsequent redistribution of them indicates soil erosion and provides the possibility for precise determination of annual soil erosion/deposition.

Due to the long half-life of 30.17 yr and also to fast and strong sorption of this radionuclide onto soil particles, the ^{137}Cs is an optimal erosion tracer and also its measurement in environmental samples using gamma-spectroscopy (an intensive gamma line at 661.7 keV) is relatively easy and accurate without the need of special chemical separation. Radiocaesium ^{137}Cs , like a fission product, has become a part of global atmospheric deposition since the nuclear weapon tests, with the maximum caesium deposition in 1963 [3, 8]. During these tests, ^{137}Cs has reached the stratosphere and the subsequent fallout affected the whole Northern Hemisphere and part of the Southern one [9]. Unlike nuclear tests, caesium released by nuclear accidents has a local character and the deposition is highly conditioned by meteorological conditions [10]. The most notable was the Chernobyl accident [11]. The contamination related to the accident was spatially inhomogeneous and depended on the trajectories of main radioactivity clouds, and on the precipitation in the area of interest at that time [12]. According to the atlas of the spatial distribution of residual levels of caesium deposition from the atmospheric testing of nuclear weapons in 1986 just prior to the accident, the average values were between 2.5 kBq m^{-2} and 3.0 kBq m^{-2} for study area [13]. There were three phases of Chernobyl radioactive cloud transport across Europe [14]. Radionuclides released

in the atmosphere between May 1st and 5th formed the third radioactive cloud, which moved south of Chernobyl, crossing former Yugoslavia, Greece, and western Turkey, and then be turned by the wind to the north towards Scandinavia. Despite the fact that majority of studies worldwide emphasize nuclear testing as the most important source of Cs in the environment, the obtained average value of surface contamination by ^{137}Cs in 1986 for territory of former Yugoslavia of 14.0 kBq m^{-2} (from 0.8 kBq m^{-2} in the zone of minimal contamination to 83 kBq m^{-2} in the zone of maximal contamination) shows that the Chernobyl fallout had relatively large contribution to total ^{137}Cs deposition in our region [15].

In alluvial environments, the penetration of radiocaesium in the vertical profile is more profound, mostly related to high water content, loose soil and the distribution of soil granulation [16]. In attempting to address these limitations of the ^{137}Cs method, the using of unsupported lead $^{210}\text{Pb}_{\text{ex}}$, as an alternative and complementary tracer in soil erosion investigations, was introduced [7, 8]. ^{210}Pb is a natural product of the ^{238}U decay series derived from the decay of gaseous radon ^{222}Rn , the daughter of ^{226}Ra . Diffusion of the ^{222}Rn from the soil introduces ^{210}Pb into the atmosphere, and its subsequent fallout provides a constant input of this radionuclide to surface soils and sediments which is not in equilibrium with its non-gaseous parent ^{226}Ra . This is the reason why the fallout component is termed as "unsupported" or "excess". The ^{210}Pb half-life of 22.26 years is also similar to that of ^{137}Cs . However, because ^{210}Pb was not present in the Chernobyl fallout, it doesn't face the problems associated with interpreting ^{137}Cs measurements in those areas that received inputs of both nuclear weapon tests and Chernobyl-derived radiocaesium.

The main aim of this study was to apply ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ nuclear methods to quantify erosion and deposition rates for the soil in a specific part of the complex gully system in the area of Loess Pyramid (LP) [19] on the southeastern part of Titel Loess Plateau (TLP). This loess plateau is a palaeoclimatic and paleoecological witness of the last 5 glacial-interglacial cycles occurred during the last 650,000 yr [17]. As a matter of this and other natural and social characteristics, TLP was proclaimed as a Special Nature Reserve. The main aim of this study is to explore soil rate erosion at the uncultivated rim of the TLP as a unique geomorphologic unit to control and manage the soil loss by pluvial erosion. In addition, the plateau is under intensive agricultural activity and there are no studies on the erosion and land degradation of the Plateau at all, especially for gully erosion.

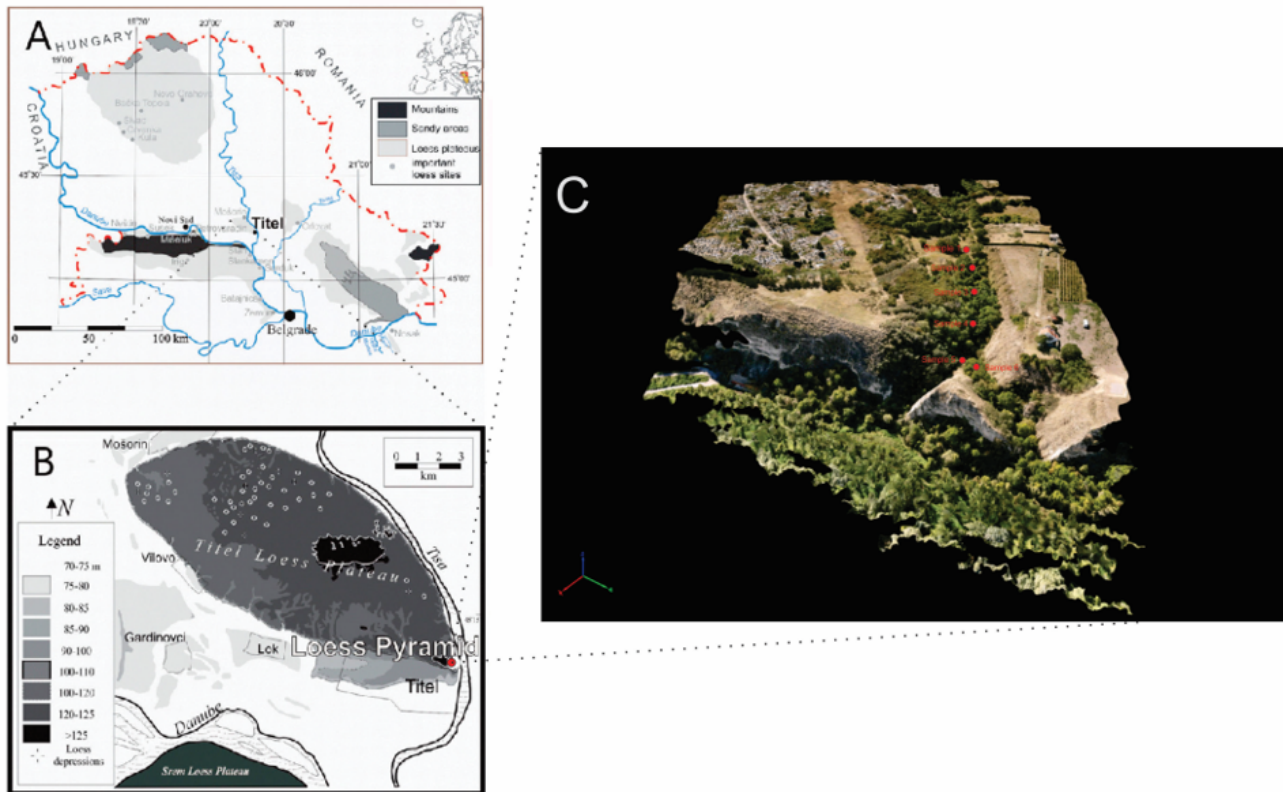


Figure 1: The geographical position of the TLP (B) on the geomorphological map of Vojvodina Province (A) [19] with assigned sampling points of study area on in-situ drone image (C).

2 Study area

The former valleys, and today the gullies, are the dominant geomorphologic forms on the edge of the TLP, which is located at the mouth of the Tisa (Tisza) River into the Danube near the settlement Titel in the Vojvodina Province of Serbia (Figure 1).

The valleys are formed on loess and intersect clay horizons, suggesting post-loess genesis and evolution. The majority of gullies on the northeastern, eastern and south-eastern edge of TLP are uncultivated and overgrown with trees, shrubs, and grass. These are generally short gullies characterized by steeper longitudinal profiles, especially in the lower part, with a continuous decline towards the Tisa River. In the cross-section they have V-shape. The side of the gullies is stretched in the upper part and narrowed in the lower part. Such as two almost overgrown gullies, 233 m north of the settlement of Titel, near the geo-locality LP in the gully system were selected for this research (Figure 2). They are divided by a watershed of an average width of 2 m. In older literature, they have named the North gully and the South gully. Since the South gully does not cut vertically, terminologically it can be characterized by a fossil

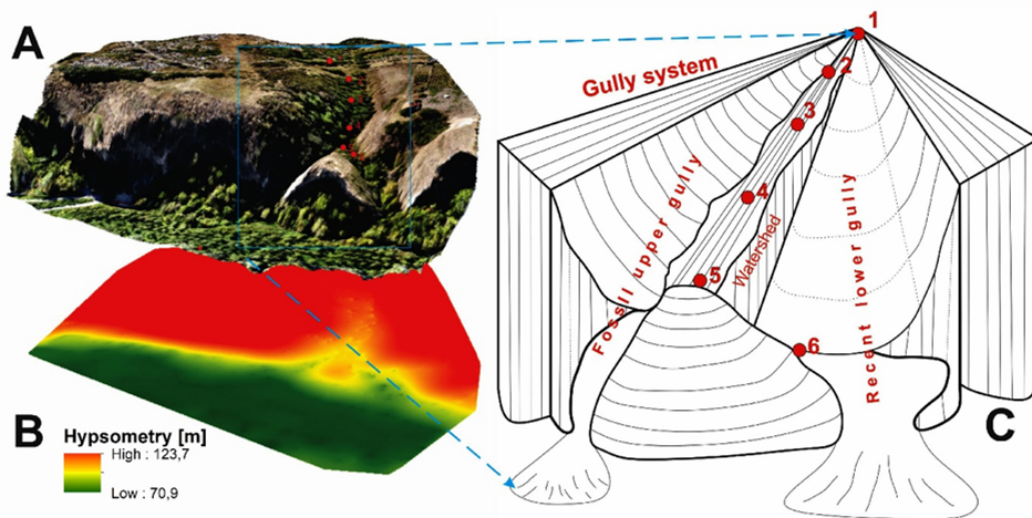
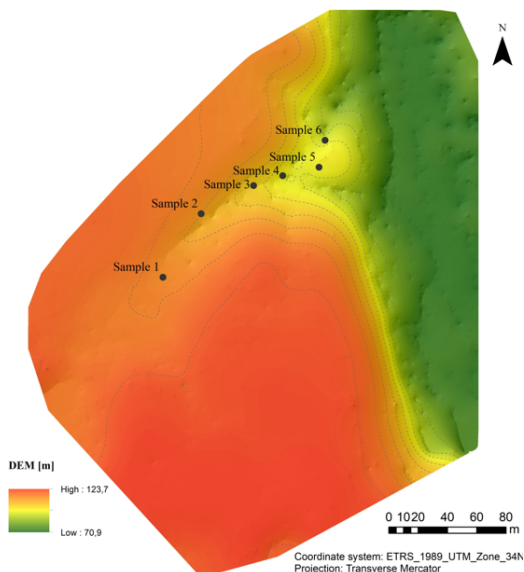
upper gully (valley). On the other hand, the North gully is vertically cut and because of this feature is characterized as a recent lower gully (valley). Due to the stronger lowering of the watershed in the future, the phenomena piteraria will occur, and depending on the configuration of the terrain, one of the aforementioned gullies (valleys) will take over the part of the whole basin of the other [18, 19].

The watershed with a length of 180.0 m and the lower part of the recent lower gully with a length of 74.4 m were chosen as the study area. The total length of the slope through sampling points is 254.4 m. Five soil profiles were sampled at the watershed and one soil profile at the bottom of the recent lower gully. The GPS coordinates of the measuring sites were determined using Garmin Oregon 450 instrument. The digital elevation model of the study area obtained by ArcMap 10.2.1 software is plotted in Figure 3.

It appears that the sampling point 1 is outside of the gully system and it has been selected as a potential reference site in an early phase of the investigation, but it turned out that it showed erosion because of the slightly inclined slope of the terrain. Therefore, to select a new reference point, two profiles were sampled on undisturbed

Table 1: Description of sampling locations with vegetation covering.

Sampling site	Latitude [°N]	Longitude [°E]	Altitude [m]	Slope [°]	Vegetation
1	45.21011	20.30824	118	7.20	grass
2	45.21050	20.30857	116	5.82	grass
3	45.21068	20.30902	112	19.75	grass
4	45.21074	20.30927	107	20.10	grass
5	45.21079	20.30959	102	13.11	poor grass
6	45.21096	20.30964	101	32.97	bare soil
7	45.25142	20.15698	97	0.47	grass
8	45.25157	20.15680	97	1.26	grass

**Figure 2:** The orthophoto model (A) with Digital Elevation Model (DEM) of the study area (B); and the sketch of the study area (C).**Figure 3:** Digital elevation model of the study area.

and flat terrain at the southwest part of the TLP, near the village of Vilovo (Figure 1). In the geomorphological sense, the selected site is a miniature loess terrace covered with grass.

Description of sampling sites (longitude, latitude, altitude and slope), average soil densities for each depth profile and vegetation that was covering soil are given in Table 1. In each layer of sampled soil profiles, the magnetic susceptibility and density of the soil were precisely determined, because the procedure for the magnetic susceptibility determination considers homogeneously packing of soil samples in plastic containers of constant volume 6.4 cm^3 and mass measurement of such prepared samples.

3 Methods

The sampling of soil in the gully was undertaken in spring 2017 after heavy precipitation in the investigated area. The

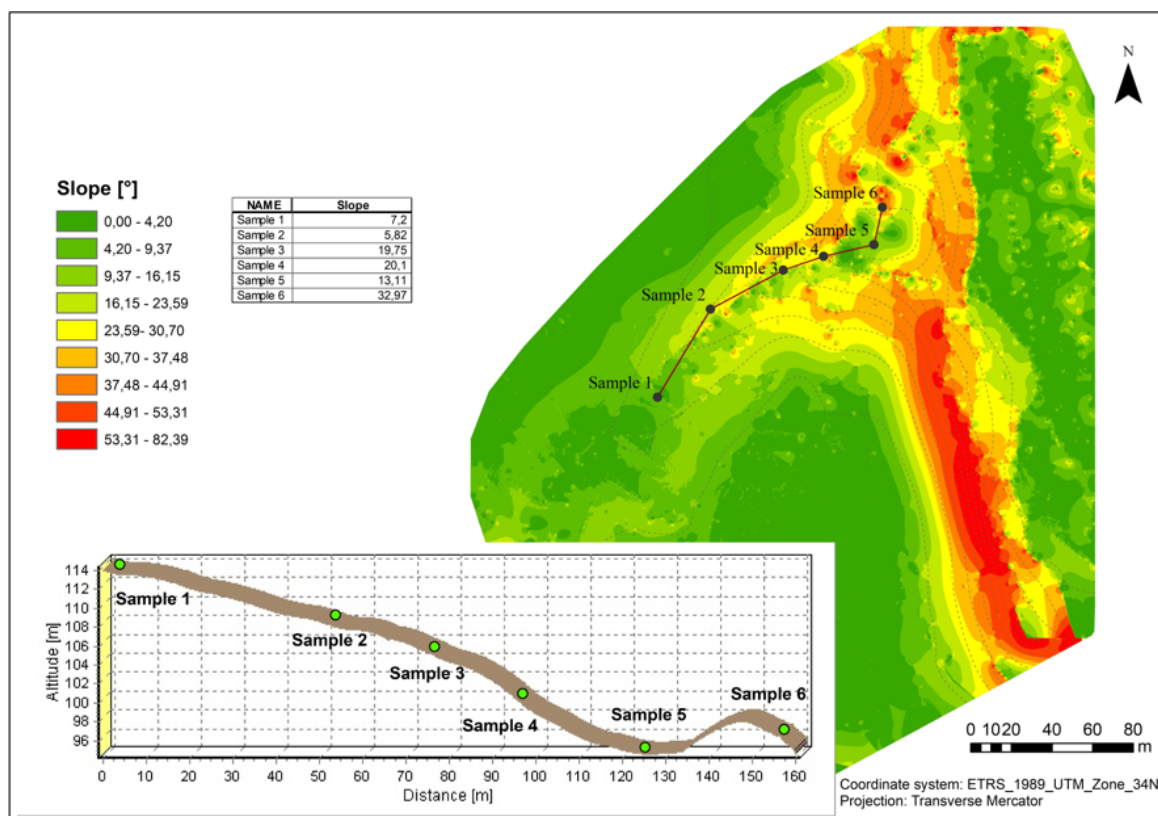


Figure 4: Slope map of the study area with a topographic profile through sampling points.

sampling points were selected on the basis of altitude and features of the surrounding terrain that favor erosion or soil accumulation at the site and are approximately equally distributed along the slope, at an adequate distance to reveal differences in ^{137}Cs activity concentration in soil (Figure 4). DEM of the study area was obtained by the Pix4D program and Figures 1, 2, 3 and 4 were generated in ArcMap. The dominant soil type is Calcerous Chernozem on TLP according to the soil map of Vojvodina Province [20]. Terrestrial gamma dose measurements were made using the Radiation Alert Inspector, GM counter with a range of the ambient dose equivalent rates from $0.01 \mu\text{Sv h}^{-1}$ to 1 mSv h^{-1} and combined measurement uncertainty of $\pm 20\%$. Only maximal values measured at contact to the soil for a period of 5 minutes were reported. Display of the instrument updates every 3 seconds, showing the average for the past 30-second time period at normal levels.

Undisturbed soil cores were collected at seven sites, using an Eijkelkamp split tube sampler of a 53 mm diameter inserted to the depth of 40 cm. After freezing, soil profiles were unpacked and described in detail. Then the cores were carefully cut into layers of 1 cm, dried on 105°C to constant mass and after grinding were packed in containers of cylindrical geometry 30 mm 67 mm. However,

the height of the samples within a container was different depending on the mass of the samples. The radionuclides content of soil samples was determined by low-level gamma spectrometry in the Laboratory for Radioactivity and Dose Measurements, Faculty of Sciences at the University of Novi Sad. In most cases, the first 1 cm of the profiles was the layer of grass and roots and was not considered in the calculations. In order to achieve a lower MDA (minimum detectable activity) time of measurement was about 80 ks and all samples from the same profile were measured on the same detector. In total 256 samples were prepared and measured in cylindrical geometry. The samples were sealed for 30 days in order to establish a secular equilibrium in the series of uranium. Two HPGe detectors with high spectral resolution were used. The first one was an ORTEC GMX type detector with a relative efficiency of 32.4% with extended energy range from 10 keV to 3 MeV inside a 12 cm thick lead shield with 3 mm Cu lining inside. The second HPGe detector was a 100% relative efficiency detector (extended range also 6 keV – 3 MeV), manufactured by Canberra USA, model GX10021 in an original lead shield of 15 cm thick. Efficiency calibration was carried out with certified reference material - a mixture of radionuclide gamma emitters in the matrix of resin of cylindrical

geometry Cert.No:1035-SE-40001-17. Different detection efficiency curves corresponding to various sample heights were obtained by the semi-empirical approach using the ANGLE v.3.0 software [21]. Gamma spectra of soil samples were acquired and analyzed using the GENIE 2000 Spectroscopy System Software program. Activity concentrations of ^{238}U were determined by a low energy 63 keV gamma line of its first progeny ^{234}Th . Having in mind the interference of 185.7 keV line (^{235}U) and 186.1 keV line from ^{226}Ra decay, it is difficult to measure ^{226}Ra based on 186.1 keV line. Thus the ^{226}Ra activity was determined from post radon gamma lines, i.e. by considering the photopeaks of ^{214}Pb (295.2 keV and 352.0 keV) and ^{214}Bi (609.3 keV). Similarly, the ^{232}Th activity was obtained from the mean activity of the photopeaks of its daughters: ^{212}Pb at 238.6 keV, ^{208}Tl at 583.1 keV and ^{228}Ac at 911.1 keV. The total ^{210}Pb activity was measured at a 46.5 keV line. Since that ^{210}Pb line (46.5 keV) represents a low energy gamma line with a pronounced absorption within the sample matrix, this absorption was taken into account by ANGLE v.3.0 software where density and elemental content of the sample was incorporated. Excess $^{210}\text{Pb}_{\text{ex}}$ concentrations were calculated by subtracting the ^{226}Ra concentrations (supported ^{210}Pb) from the total ^{210}Pb concentrations. After gamma-spectrometry measurements, soil samples were carefully compressed with a non-magnetic pistil and packed in non-magnetic plastic boxes (vol. 6.4 cm^3), enabling the precise determination of the density of the layers by measuring the mass of such prepared samples. Magnetic susceptibility was measured in low-frequency χ_{lf} ($10^{-8}\text{ m}^3\text{kg}^{-1}$) and in high-frequency magnetic field χ_{hf} ($10^{-8}\text{ m}^3\text{kg}^{-1}$) using a dual-frequency Bartington MS2/MS2B sensor at two frequencies. The relative frequency-dependent susceptibility in% expressed as

$$\Delta\chi = (\chi_{\text{lf}} - \chi_{\text{hf}})/\chi_{\text{lf}}, \quad (1)$$

which characterized magnetic properties of soil was calculated for each layer of 1 cm thickness. According to obtained results, 35 targeted layers of soils were selected for grain size measurements related to extreme values of magnetic susceptibility. The soil grain sizes distribution was determined by laser diffraction dry method with particle size analyzer Mastersizer 2000, Malvern Instruments at the Faculty of Technical Sciences, Novi Sad.

Having in mind the variety of procedures and models, used for estimation of erosion rate or deposition, based on measured ^{137}Cs activity concentrations, a distinct difference between agricultural soil and non-agricultural soil (meadow, pasture) should be emphasized. In the case of agricultural soil, due to tillage, the mixing of surface and deep soil layers occurs which eliminates the difference

in ^{137}Cs activity concentration among soil layers. On the other hand, ^{137}Cs is concentrated in the surface layer of non-agricultural soil, within 10 cm depth, and decreases exponentially with depth (migrated no deeper than 10-20 cm), which reflects the atmospheric artificial origin of this radionuclide [22, 23]. The migration and distribution of radiocaesium in the soil depth profile vary depending on soil properties such as soil granulation, NH_4 concentration, and organic matter content, soil clay content, exchangeable K status and pH, as well as on climatic conditions, land use and management practices [24].

The traditional approach to using the ^{137}Cs method is based on the comparison between the inventory (total radionuclide activity per unit area) at a given sampling site and values measured at a reference site located in a flat and undisturbed stable area [8, 9]. This proportional model indicates erosion at sites with a lower ^{137}Cs inventory compared to the reference site and sediment deposition processes at sites with a greater ^{137}Cs inventory.

Total ^{137}Cs activity per square meter for sampling points was calculated as following [8]:

$$^{137}\text{Cs}_{\text{inventory}} = \sum_{i=1}^n C_i B D_i D_i, \quad (2)$$

where i is a number of the particular layer, n is the maximum number of layers with registered ^{137}Cs concentrations, C_i is ^{137}Cs activity concentration (Bq kg^{-1}) for a particular layer, $B D_i$ – density of dried soil (kg m^{-3}) for particular layer and D_i is layer thickness (m).

The reduction of total ^{137}Cs activity per square meter relative to the reference value is derived from [8], as

$$X = \frac{A_{\text{ref}} - A}{A_{\text{ref}}} \times 100\% \quad (3)$$

where A_{ref} is a total activity of ^{137}Cs per square meter for a reference point (Bq m^{-2}), A is a total activity of ^{137}Cs per square meter for sampling points (Bq m^{-2}).

The profile shape model is a convenient way of assessing erosion for uncultivated soil due to the exponential decrease of ^{137}Cs mass concentration with depth down an undisturbed soil profile that may be described by the following function [8], as

$$A'(x) = A_{\text{ref}} \left(1 - e^{-\frac{x}{h_0}}\right), \quad (4)$$

where $A'(x)$ is the amount of ^{137}Cs above the depth x (Bq m^{-2}), x is the mass depth from the soil surface (kg m^{-2}) and h_0 is coefficient describing profile shape (kg m^{-2}) referred to the penetration of ^{137}Cs into the soil.

Guided by the assumption that total ^{137}Cs fallout occurred in 1986 and that the depth distribution of caesium

Table 2: Mean cumulative particle size distribution D50 with ambient dose equivalent rates $H^*(10)$ ($\mu\text{Sv h}^{-1}$) and results of ^{137}Cs inventories and $^{210}\text{Pb}_{\text{ex}}$ inventories for each sampled profile.

Sampled profile	D50 _{mean} [μm]	$H^*(10)$ [$\mu\text{Sv h}^{-1}$]	^{137}Cs inventory A [Bq m^{-2}]	$^{210}\text{Pb}_{\text{ex}}$ inventory A [Bq m^{-2}]
1	33.46	0.220	3568	19209
2	37.52	0.200	5090	50484
3	36.34	0.150	5154	20068
4	38.01	0.173	5154	55960
5	47.51	0.137	4025	16520
6	45.24	0.167	3061	45948
7	39.22	0.260	6174	23475
8	47.12	0.150	2128	53234

is independent of time, the annual soil erosion rate Y ($\text{t ha}^{-1} \text{yr}^{-1}$) for an eroding point (with total ^{137}Cs inventory less than local reference inventory A_{ref}) can be estimated from the following equation:

$$Y = \frac{10}{t - 1986} \ln \left(1 - \frac{X}{100} \right) h_0 \quad (5)$$

where t is a year of sampling and h_0 is a factor that describes the shape of profile (kg m^{-2}).

For a depositional site, the deposition rate R' can be estimated from the excess caesium inventory $A_{\text{ex}}(t)$ (Bq m^{-2}) (defined as $A - A_{\text{ref}}$) and the ^{137}Cs concentration of deposited sediment C_d [8] in the following form, as

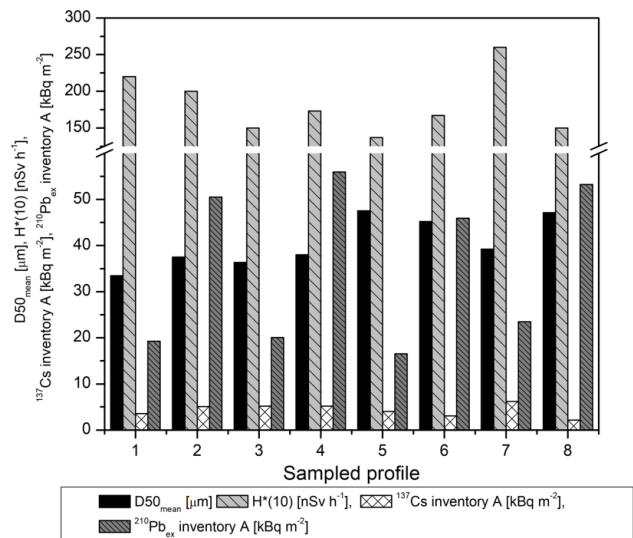
$$R' = \frac{A_{\text{ex}}(t)}{\int_{t_0}^t C_d(t') e^{-\lambda(t-t')} dt'} \quad (6)$$

where λ is decay constant for ^{137}Cs (yr^{-1}).

Although the profile shape model is a simple way to convert ^{137}Cs measurements to quantitative estimates of erosion and deposition rates, it does not take account of time-dependent ^{137}Cs fallout input and the progressive redistribution of this radionuclide in the soil profile after deposition from the atmosphere. The redistribution of caesium in uncultivated soils can be described using a one-dimensional diffusion and migration model characterized by an effective diffusion coefficient D ($\text{kg}^2 \text{m}^{-4} \text{yr}^{-1}$) and downward migration rate V ($\text{kg m}^{-2} \text{yr}^{-1}$) of ^{137}Cs in the soil profile [25]:

$$C_u(t) \approx \frac{I(t)}{H} + \int_0^{t-1} \frac{I(t') e^{-\frac{R}{H}}}{\sqrt{D\pi(t-t')}} e^{-\frac{V^2(t-t')}{(4D)-\lambda(t-t')}} dt' \quad (7)$$

where $C_u(t)$ (Bq kg^{-1}) is the variation of the ^{137}Cs concentration in surface soil with time t (yr), $I(t)$ is the annual ^{137}Cs deposition flux ($\text{Bq m}^{-2} \text{yr}^{-1}$); H (kg m^{-2}) is the relaxation mass depth of the initial distribution of fresh fallout

**Figure 5:** The mean grain size D50mean (μm), the ambient dose equivalent rates $H^*(10)$ (nSv h^{-1}) and results of ^{137}Cs inventories and $^{210}\text{Pb}_{\text{ex}}$ inventories (kBq m^{-2}) along with the soil profiles.

^{137}Cs at the surface of the soil profile and R is the erosion rate ($\text{kg m}^{-2} \text{yr}^{-1}$).

For a depositional site, the deposition rate R' can be derived from the ^{137}Cs concentration of deposited sediment $C_d(t')$ and the excess ^{137}Cs inventory $A_{\text{ex}}(t)$ (defined as the total measured ^{137}Cs inventory A_u less the local reference inventory A_{ref}) using the following relationship:

$$R' = \frac{A_{\text{ex}}}{\int_{t_0}^t C_d(t') e^{-\lambda(t-t')} dt'} = \frac{A_u - A_{\text{ref}}}{\int_{t_0}^t C_d(t') e^{-\lambda(t-t')} dt'} \quad (8)$$

$^{210}\text{Pb}_{\text{ex}}$ has a comparable half-life (22.3 years) to ^{137}Cs , but its natural origin and continuous fallout provide the potential for the assessment of longer-term erosion rates. The diffusion and migration model for ^{137}Cs could also be modified for use with $^{210}\text{Pb}_{\text{ex}}$ measurements obtained from uncultivated soils. It is assumed that $^{210}\text{Pb}_{\text{ex}}$ fallout input

Table 3: The results of particle size analysis of soil layer samples from different depths with the weight percentages of five defined fractions.

Profile	Depth [cm]	Clay [%]	Silt [%]	Fine sand [%]	Medium sand [%]	Coarse sand [%]	D50 [μm]
1	7	9.48	49.72	16.86	6.35	17.57	48.74
	18	16.92	63.13	15.83	4.03	0.14	26.54
	28	16.16	60.49	14.14	7.43	1.77	28.59
	35	14.77	61.18	16.58	6.29	1.22	29.97
2	4	6.38	59.07	19.93	8.51	6.16	41.78
	13	10.97	57.00	16.08	6.46	9.47	36.78
	18	12.58	57.73	17.75	8.14	3.78	34.81
	29	9.98	58.43	19.46	8.28	3.55	36.71
3	6	9.41	60.00	17.21	6.36	7.03	37.95
	15	9.62	61.34	14.84	7.28	6.96	35.13
	25	11.82	59.94	15.31	5.04	7.88	34.50
	34	10.31	56.81	15.41	5.47	10.55	37.79
4	6	9.87	61.34	18.63	7.80	2.67	36.78
	12	8.87	56.05	17.91	7.45	9.74	42.35
	23	12.30	59.68	14.79	4.23	9.02	34.51
	29	11.29	56.37	14.89	5.29	12.15	38.39
5	5	4.18	59.18	33.61	2.98	0.00	50.37
	14	5.86	57.12	33.36	6.65	0.00	50.25
	21	6.29	57.99	32.64	3.10	0.00	49.11
	32	8.89	60.02	25.03	2.78	0.30	40.29
6	3	5.14	58.03	24.64	7.24	4.92	47.64
	7	6.16	63.01	24.60	5.70	0.51	42.40
	13	7.40	58.52	22.12	8.26	5.17	44.22
	17	6.87	56.49	22.37	6.95	7.28	47.18
7	22	7.66	58.63	22.77	6.29	4.62	44.44
	24	8.72	55.38	21.62	7.10	7.13	45.54
	6	9.98	68.61	16.98	1.27	3.19	29.53
	15	8.23	50.65	13.40	16.46	10.18	46.70
8	21	7.13	57.79	41.16	8.30	13.18	43.09
	27	7.24	62.18	9.06	7.24	14.27	36.83
	31	7.29	57.01	10.96	10.42	14.32	39.97
	4	5.70	52.53	17.20	9.60	14.97	52.35
8	9	5.81	50.68	16.24	15.17	12.09	49.66
	16	7.94	61.96	14.69	10.47	4.95	50.72
	30	6.38	50.59	13.68	17.52	11.85	35.77

and the loss by decay represent a steady-state and that the reference inventory will, therefore, remain the same through time. The deposition flux $I(t)$ may be calculated from the local reference inventory A_{ref} , using the equation:

$$I(t) = \frac{A_{ref} \ln 2}{22.3} \quad (9)$$

4 Results

Obtained results of the median diameter of the soil grain size distribution at 50% in the cumulative distribution D50 (μm) with ambient dose equivalent rates $H^*(10)$ ($\mu\text{Sv h}^{-1}$) and results of ^{137}Cs inventories and $^{210}\text{Pb}_{ex}$ inventories for each sampled profile are shown in Table 2 and Figure 5. Soil granulation was classified according to Wentworth scale [26] in five components: clay ($< 3.9 \mu\text{m}$), silt ($3.9\text{--}62.5$)

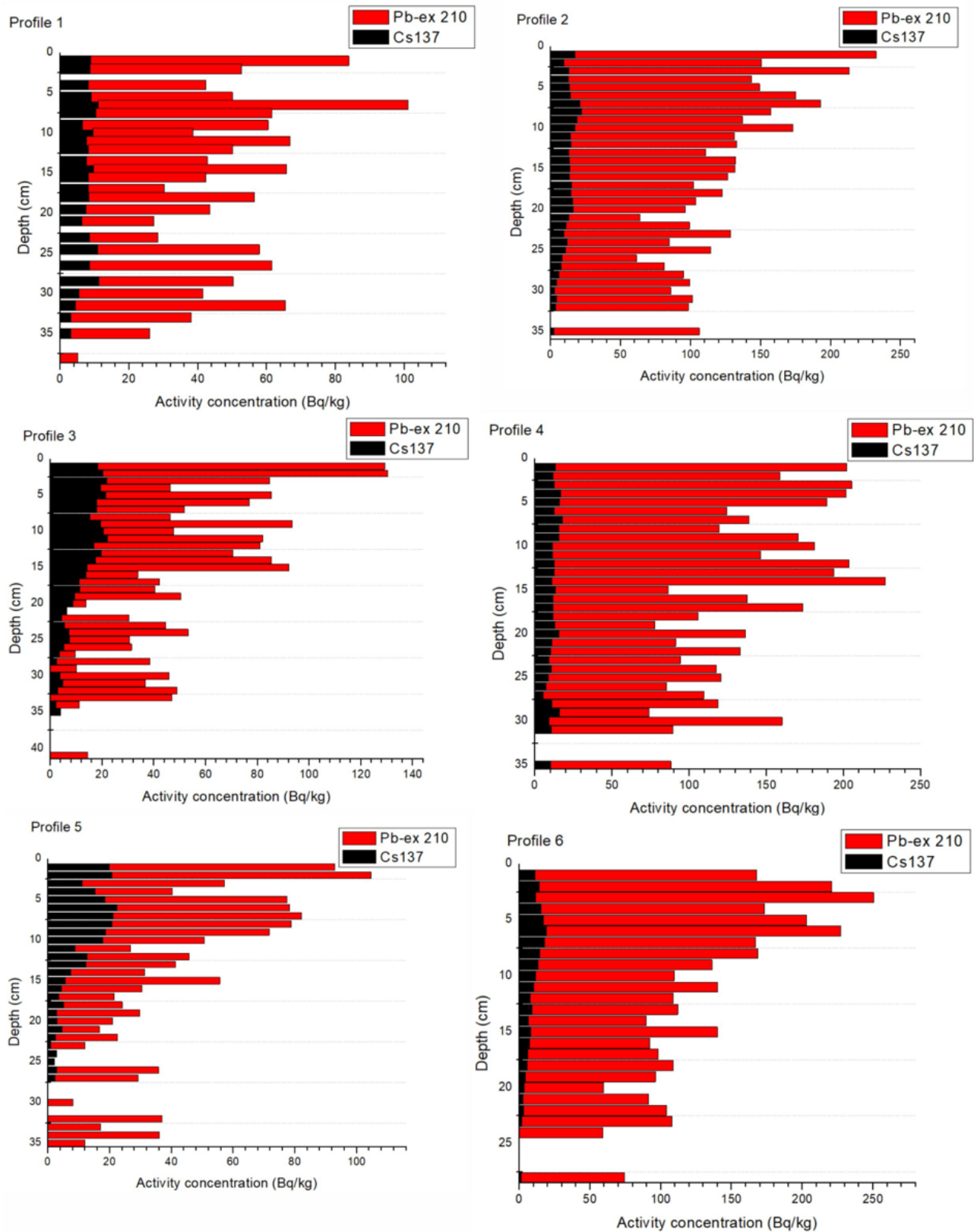


Figure 6: The depth distribution of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activity concentrations for six sampled profiles.

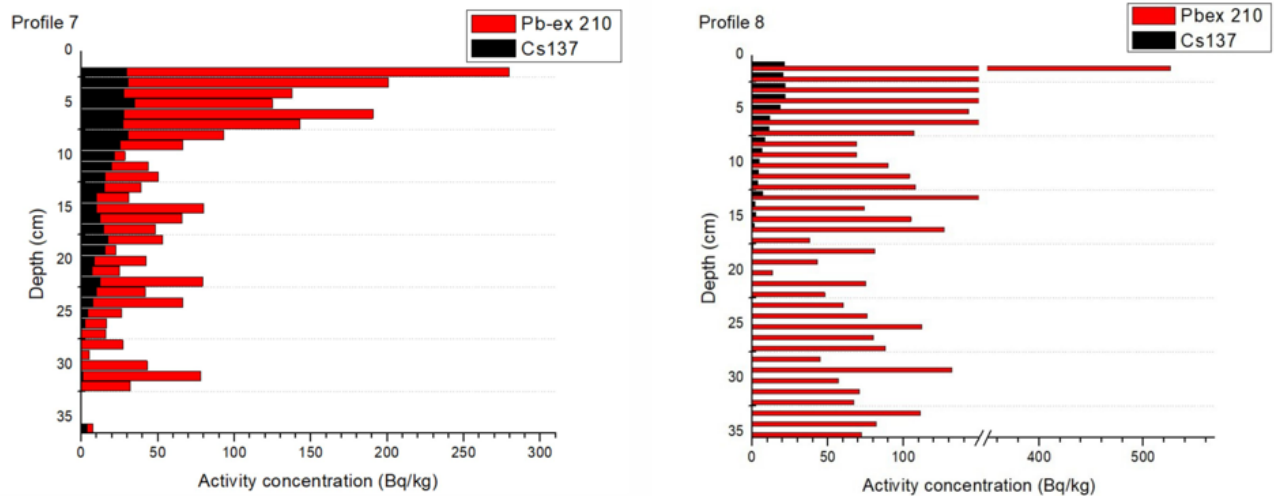


Figure 7: The depth distribution of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activity concentrations for the reference site profiles.

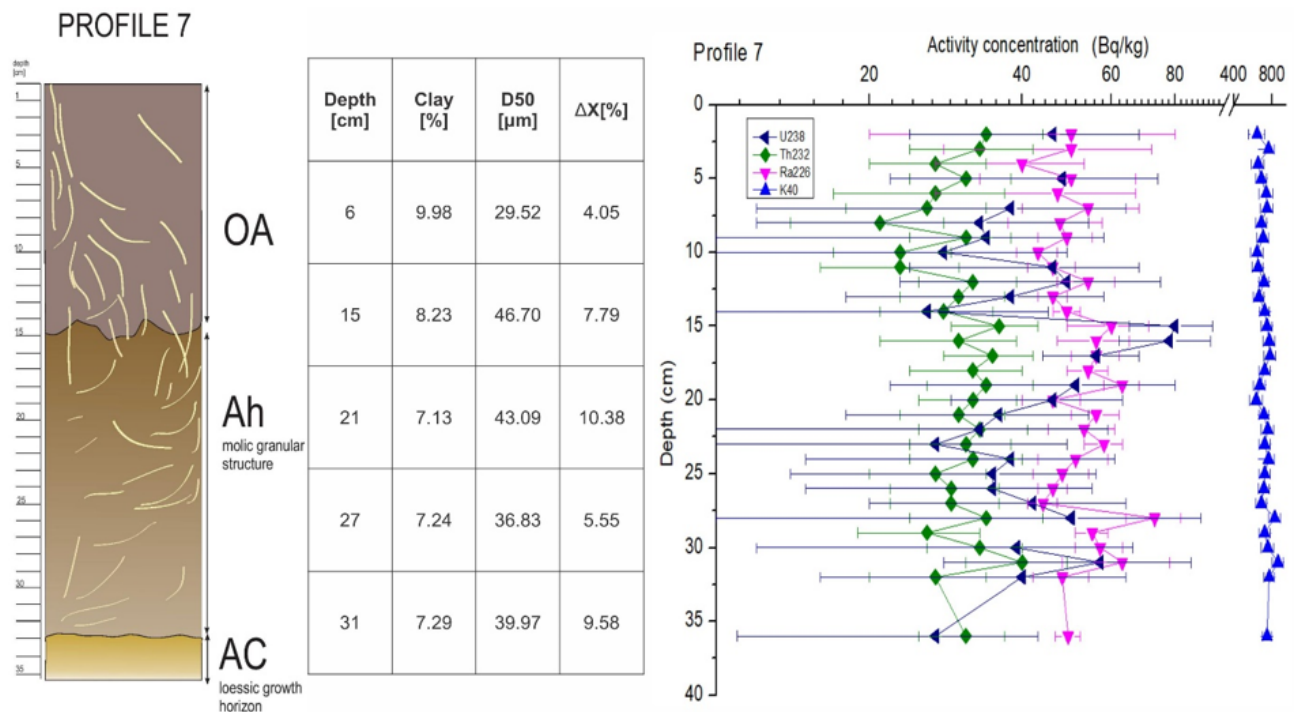


Figure 8: Soil horizons with the vertical distribution of clay content, soil grain size, magnetic data and natural radionuclides for the reference profile 7.

μm , fine sand (62.5-250 μm , medium sand (250-500 μm and coarse sand (500-2000 μm .

Results of particle size analysis carried out on 35 samples from all soil profiles on different depths (Table 3) indicate the dominant presence of silty clay which is soil component with low ability to resist soil erosion.

It is noticed that the percentage of clay content increases with the depth in almost all profiles (except the reference profile 7) which affects the ^{137}Cs vertical distribu-

tion and enables the penetration of caesium ions to deeper layers of soil. The depth distributions of measured ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ activity concentrations for 6 profiles along the study area are presented in Figure 6. Only sampling point 4 shows atypical depth distribution of ^{137}Cs concentrations. The vertical profile distribution of ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ concentrations obtained for the profiles 7 and 8, which are recognized as the reference sites are presented in Figure 7.

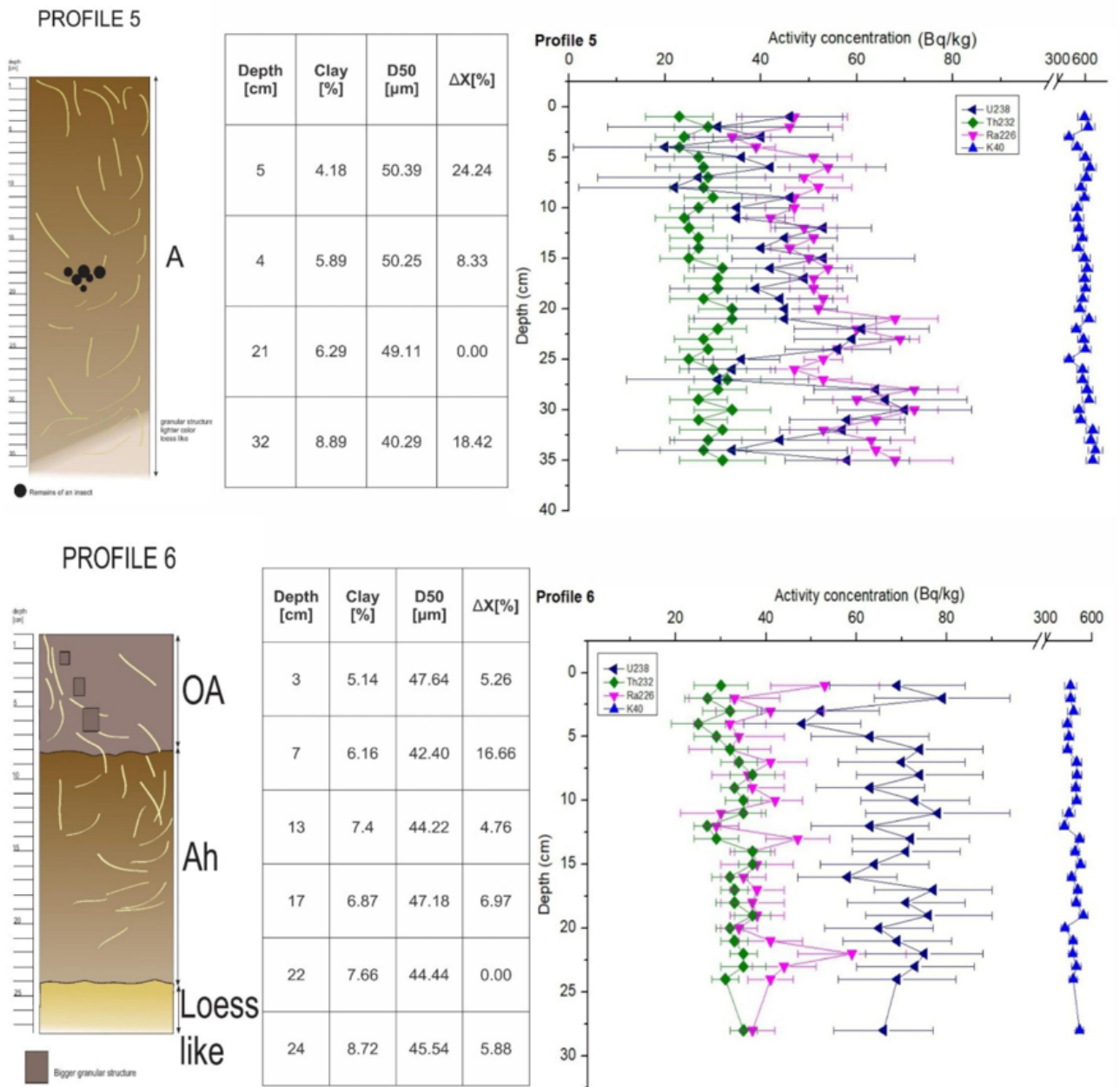


Figure 9: Soil horizons with the vertical distribution of clay content, soil grain size, magnetic data and natural radionuclides for profiles 5 and 6 that reflect erosion.

Soil horizons with the vertical distribution of natural radionuclides (²²⁶Ra, ²³⁸U, ²³²Th and ⁴⁰K) for the reference profile are shown in Figure 8. OA horizon is a transitional horizon at the top of the soil. The middle, strongly developed Ah horizon is rich in humus providing high fertility of chernozems and finally, the AC horizon represents transitional zone between soil and loess. The activity concentrations of the natural radionuclides are detected in all samples at the normal equilibrium environmental levels

and vary in the next ranges: (25 – 85) Bq kg⁻¹ for ²²⁶Ra, (20 – 84) Bq kg⁻¹ for ²³⁸U, (18 – 47) Bq kg⁻¹ for ²³²Th and (372 – 920) Bq kg⁻¹ for ⁴⁰K, respectively. The presence of organic matter in soil affects the content of ²²⁶Ra for which the only variation along the layers is observed that is in good agreement with published results [23]. This claim can't be applied to artificial ¹³⁷Cs and unsupported ²¹⁰Pb_{ex} which exponential decrease with depth from topsoil (Figure 6 and 7) which is typically for fallout radionuclides. Besides the

Table 4: Estimated soil redistribution rates using different conversion models.

Profile	^{137}Cs method			$^{210}\text{Pb}_{\text{ex}}$ method
	PDM (1963)	PDM (1986)	D&M	D&M
1	−4.78	−8.26	−6.9	−7.9
2	−1.68	−2.87	−2.8	−0.6
3	−1.58	−2.68	−2.6	−7.6
4	−1.07	−1.80	−1.8	0.62
5	−3.73	−7.41	−5.6	−8.6
6	−6.12	−10.60	−8.4	−1.5
7	reference inventories			
8				

depth distribution of radionuclides and soil horizon, the variation of soil magnetic data, clay content and grain size D50 with the depth of the soil layer is shown in Figure 8. To prove the claim that radionuclide concentration varies with clay content along the depth similar view of data was presented in Figure 9 for soil profiles 5 and 6 where both ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ method indicate erosion.

The soil erosion rates from measurements of ^{137}Cs inventories were calculated for the investigated sites using two convenient models for uncultivated soils which are explained above. These are the profile distribution model (PDM) and the diffusion and migration (D&M) model. The profile shape coefficient estimated for the profile 7 chosen as the reference site ($h_0=47.1 \text{ kg m}^{-2}$) was needed to assess the soil redistribution rates according to PDM (Table 3). In Table 4 results obtained from the excel add-in program [27] where it is not possible to change the year of total ^{137}Cs input PDM (1963) and from equation (4) with a modified year of total input (1986) are presented and compared. In order to obtain more realistic results D&M model ^{137}Cs method was applied, because this model takes into account post-fallout migration/diffusion of the ^{137}Cs . The contribution of the Chernobyl fallout was assumed to be 80% which is recommended by many studies for our region [28–30]. For the relaxation depth H , diffusion coefficient D and migration rate V , the values reported by Walling and He [27] were adopted. For the $^{210}\text{Pb}_{\text{ex}}$ method, D&M model was only used with profile 8 as a reference site. Estimated results of soil redistribution rates show good agreement with ^{137}Cs method for profiles 1 and 5 indicating a slightly higher net erosion of $-3.7 \text{ t ha}^{-1} \text{ yr}^{-1}$ with 98% of sediment delivery ratio.

5 Discussion and Conclusions

Results of this preliminary soil erosion assessment show that the ^{137}Cs method for the first time in the Vojvodina region gives reliable estimates of the rates of soil loss comparable to results obtained by unsupported $^{210}\text{Pb}_{\text{ex}}$ method for the study area. Differences in vertical distribution shape or in values of their inventories in soil profiles could be explained by the different origin and time consistency of their deposition from the atmosphere.

On most experimental points, erosion is a dominant process, and probably the area of accumulation is in the river Tisza – on river right bank, river's side and riverbed. The values of soil loss estimated from the values of ^{137}Cs inventory range from $-1.58 \text{ t ha}^{-1} \text{ yr}^{-1}$ to $-10.60 \text{ t ha}^{-1} \text{ yr}^{-1}$ related to erosion classified as moderate.

As a rule, gullies do not have organic soil horizons because they have been carried away by the long process of fluvial and pluvial erosion. However, some samples have organic horizons in their vertical profiles (sample 2, sample 6). The existence of organic horizons may indicate that the weakening of the rainfall is the predominant process of washing away the soil from the TLP (sheet erosion). These sediments generally remain within the gully, so they in addition to the transport role-play the accumulation of organic matter. But, during heavy rains, the gully is a source of sediment and transports them to the inundation plane of the Tisza River. This fact may make it difficult to interpret the results obtained by using nuclear methods. Nevertheless, this research confirmed that the ^{137}Cs and $^{210}\text{Pb}_{\text{ex}}$ method is absolutely applicable in the investigation of soil erosion in gullies on the Titel plateau and that the results obtained are consistent with studies dealing with similar issues [29, 30].

The continuation of this research on a larger number of samples and a more detailed selection program of the reference site is necessary. We plan to apply Gavrilovic's method for comparison and validation of results. This method, also known as the Erosion Potential Model EPM, uses the precipitation among another huge number of parameters and that will improve current results.

The usage of more different conversion models, such as profile distribution and diffusion and migration model recommended by many authors for the erosion assessment for undisturbed soils, in combination with particle size analysis gives more accurate estimates.

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