

Spatial and vertical distribution of ^{137}Cs in soils in the erosive area of southeastern Serbia (Pčinja and South Morava River Basins)

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Received: 28 November 2014 / Accepted: 1 July 2015 / Published online: 15 July 2015
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Abstract

Purpose The area of southeastern Serbia, the Pčinja and South Morava River Basins, is under the influence of very strong erosion, and the aim of this study was to investigate the vertical and spatial distribution of the ^{137}Cs in the eroded soils of this area.

Materials and methods Vertical soil profiles were collected with 5-cm increments from the uppermost layer down to 20, 25, 30, 40, and 50 cm of depth, depending on the thickness of the soil layers, i.e., down to the underlying parent rocks. Measurements of ^{137}Cs activity concentration were performed by using the HPGe gamma-ray spectrometer ORTEC-AMETEK (34 % relative efficiency and high resolution 1.65 keV at 1.33 MeV for ^{60}Co), from its gamma-ray line at 661.2 keV.

Results and discussion The mean ^{137}Cs activity concentration across all 18 soil profiles (for all soil layers) was found to be 20 Bq kg⁻¹. In the greatest number of soil profiles, the ^{137}Cs activity concentration was generally highest in the first soil layer (0–5 cm) and decreased with soil depth, while in a few soil profiles, the peak of either the ^{137}Cs activity concentration occurred in the second soil layer (5–10 cm) or the ^{137}Cs

activity concentration was almost equal throughout the entire soil profile. The mean ^{137}Cs activity concentration in the first soil layer (0–5 cm) was found to be 61 Bq kg⁻¹, and the high coefficient of variation of 92 % pointed out high spatial variability and large range of the ^{137}Cs activity concentrations in the study area.

Conclusions The obtained results indicate that in the greatest number of soil profiles, ^{137}Cs is present in the upper layers, with concentration decreasing with depth, as is typical in uncultivated soil. Its spatial distribution was very uneven among the surface soil layers of the investigated sites. One of the main reasons for such pattern of ^{137}Cs in the study area may be soil erosion. Additional investigations which would support this hypothesis are required.

Keywords ^{137}Cs · Spatial distribution · Vertical distribution · Southeastern Serbia · Pčinja and South Morava River Basins

1 Introduction

Radioactive fallout has been globally monitored since the early 1950s. Radionuclides with low mobility remain in the uppermost layers of the soil for a long time and may significantly contribute to higher doses in humans and biota. On the other side, if their migration is fast, they may reach the groundwater pathways and cause contamination. The migration of radionuclides in the environment consists of a horizontal and a vertical component, although on flat surfaces, the vertical component is dominant (Bossey and Kirchner 2004). The vertical migration of radionuclides in soil is a complex process and depends on a number of factors such as the characteristics of the radionuclide, the soil type and its chemical and physical characteristics, and the land use (Ivanov et al. 1997; Al-Masri 2006; Petrović et al. 2013). In order to investigate the vertical

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distribution of caesium in soil, many models have been developed over the years such as exponential (Isaksson and Erlandsson 1995), compartmental (Kirchner 1998), and convection–diffusion (Schuller et al. 1997; Smith and Elder 1999; Likar et al. 2001; Bossew and Kirchner 2004; Krstic et al. 2004). Two basic processes control the migration of the fallout in the undisturbed soil profile. Firstly, the particles may percolate downwards with rain water; secondly, the solvable phase is subjected to convection and diffusion with the soil solution and sorption to the soil matter described by the convection–diffusion model equation (CDE) (Bossew et al. 2004). It is generally accepted that the migration of ^{137}Cs in soil may be modeled by the Fokker–Planck equation governing convection activities and diffusion (Krstic et al. 2004).

Due to its long half-life, ^{137}Cs has been widely used for tracing the movement of soil and sediment particles within the landscape (Ritchie and McHenry 1990). The soil types differ in content of minerals and organic matter at one hand and water and air proportions at the other. It is known that the transport rate is greater in soils with coarser texture and porosity (Barišić et al. 1999). Positive ions such as Cs^+ are adsorbed on the surface of negatively charged particles of soil, such as clay minerals. The number of available sites for adsorption is a measure of the sorption capacity and is often described by the cation exchange capacity. It is known that organic matter has high cation exchange capacity, but Cs^+ ion is weakly adsorbed on it, because ions with high-charge density are favored (Dumat et al. 1997). The climate

properties, e.g., rainfall, snowfall, and temperature, affect the downward migration of ^{137}Cs (Smith and Elder 1999).

In Serbia, ^{137}Cs mainly originated from the Chernobyl accident while its much smaller portion derives from atmospheric nuclear weapons tests (Petrović et al. 2013; Janković-Mandić et al. 2014). After the Chernobyl accident, the spatial and vertical migration of the ^{137}Cs in soil has become the main goal of a number of studies in Serbia (Bikit et al. 2005; Dugalic et al. 2010; Grdović et al. 2010; Dragović et al. 2012a; Petrović et al. 2013). The aim of this study was to investigate vertical and spatial distribution of ^{137}Cs in soils in the southeastern Serbia, particularly the Pčinja and South Morava River Basins which is known as an area with very strong soil erosion.

2 Study area

The study area (Fig. 1) spreads from the Krševica River Basin in the west (South Morava River Basin) to the western slopes of Dukat Mountain in the east (Pčinja River Basin). In the north, the border of the area coincides with the mountain watershed of Zladovačka Mountain and Patarica, and in the south, the physical and geographic boundaries are presented by the watershed of Široka Mountain (along the border with the Former Yugoslav Republic of Macedonia—FYR Macedonia). The upper part of the Pčinja River Basin belongs to Serbia with an area of 1,247 km², and the lower one to the FYR Macedonia, with an area of 1,893 km².

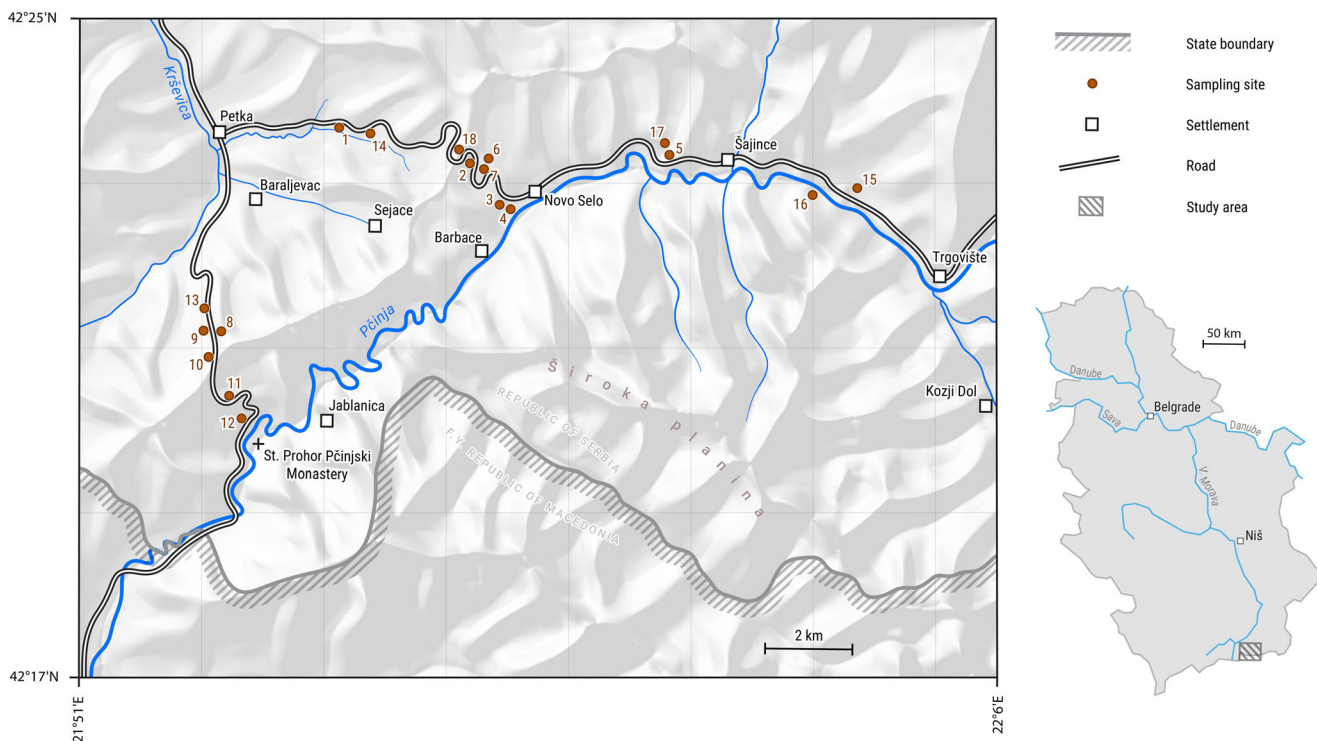


Fig. 1 The simplified map showing the location of the study area within Serbia and the locations of sampling sites

The sites where the soil samples were taken mostly belong to the Serbian–Macedonian Mass, while others are located in the Vardar tectonic unit. Fluvial sediments formed by exogenous agents in the recent phase have been mosaically arranged in the area between the towns Trgovište and Barbace. Oxidized sandstones, fanglomerate, and non-stratified clay–sandy material, as well as clay, marl, and breccia, have greater ranges, indicating a short and intense sedimentation (Babović and Cvetković 1977).

The temperate-continental climate of the Pčinja River Basin has been modified under the influence of the direction of the valley, the distance from the Aegean Sea, altitude, configuration, coverage, and type of vegetation. According to the criterion of continentality and aridity, the basin itself has the characteristics of sub-Mediterranean and modified Aegean climate, but the mountain heights above the altitude of 1,300 m are exposed to the influence of air masses coming from the south and west quadrants. Western winds usually bring precipitation in early winter and late spring, but the air mass coming from the Aegean Sea occurs as a modifier from continental to warm valley climate.

In the upper part of the Pčinja River Basin, soil is exposed to erosion processes, especially to pluvial erosion and denudation. The precipitation is an important factor in soil erosion. The precipitation rate in the depression (lowland of the Basin) ranges from 614 mm in Trgovište to 667 mm in Bujanovac (Republic Hydrometeorological Service of Serbia 2011). In the mountains that frame the basin, the precipitation rate is about 1,000 mm. The intensive soil erosion and reduction of the soil profile occur as a result of the terrain slope, soil erodibility, intensity of rainfall, exposure to frontal rainfall series, and coverage of vegetation of deep and branched roots.

The geological composition of the terrain and pedogenetic processes influenced the distribution of certain types, subtypes, and forms of soil. The acidic and brown podzolic soils are of dominant distribution, i.e., Haplic Cambisols and Luvic Planosols. From Šajince to Trgovište, young undeveloped soils are represented by Lithic Leptosols and Nudilithic Leptosols on gneiss. Young, shallow, sandy, and sandy loam soils are widespread on steep slopes affected by intense pluvial erosion and denudation. Haplic Cambisols and Haplic Regosols are present along the valleys of the upper Pčinja River Basin, especially in the central and northwestern parts of the area (Zlatković 2011). At the contact of the mountain slopes and the Pčinja River Valley and on the river terraces, the soils of two types, Colluvic Regosols and Leptic Regosols, are accumulated. Permeable and shallow crumbly humus-silicate soils (Humic Leptosols) were recorded throughout the area (Dugalić and Gajić 2012; FAO 2006). Another representative group of soils is Cambisols. In areas inclined at an angle of about 45° with intense pedogenetic processes, the dominant soil type is Humic Leptosols. Haplic Cambisols were registered at higher altitudes, especially on Široka

Mountain, Patarica, Zladovačka Mountain, and western slopes of Dukat Mountain. Undeveloped alluvial soils (Fluvisols) are present on the sedimentary layers of the Pčinja River and along its tributaries: Kočurica and Kozjedolska River (Zlatković 2011). The soils of south-east Serbia are formed from geological substrate composed mainly of crystalline schists and Neogene lake sediments. Different soil types with their varieties and forms, formed during further pedological processes, have a significant role in understanding soil erodibility. The dominant soil types in the investigated area, Leptosols, Cambisols, and Fluvisols, are generally susceptible to erosion (FAO 2006).

Previous investigation in the study area has shown variability of ^{137}Cs activity concentrations in soil ranging from 4.5 to 84 Bq kg⁻¹ with the mean of 29 Bq kg⁻¹ (Dragović et al. 2012a, b).

3 Materials and methods

3.1 Soil sampling

Samples of 18 vertical soil profiles were collected in 2013 (Fig. 1). The vertical soil profiles were collected with 5-cm increments from the uppermost layer down to 20 cm (soil profiles I, XIV, and XVI), 25 cm (soil profiles VIII, XIII, XV, and XVII), 30 cm (soil profile XI), 40 cm (soil profiles X and XII), or 50 cm (soil profiles II–VII, IX, and XVIII) of depth, depending on the thickness of the soil layers, i.e., down to the underlying parent rock. All the soil profiles were collected from the area where strong soil erosion was noted in the past (bare rocks, low soil profile). At the time of sampling, the erosive processes seemed to be significantly slowed down due to the expansion of primary vegetation as a consequence of lower intensity of cattle grazing. Basic descriptive statistics of particle size distribution (sand (2.0–0.05 mm), silt (0.05–0.002 mm), and clay (<0.002 mm)) in the analyzed soil profiles are presented in Table 1. According to USDA classification (USDA 1999), the analyzed soil samples belong to the following textural classes: sandy loam (47.0 %), loam (32.1 %), loamy sand (8.2 %), silt loam (7.5 %), lay loam (3.7 %), and silty clay loam (1.5 %). The soils in the study area are slightly acidic with very low organic matter content. The soil profiles were collected from the slopes with an angle of 10 to 63° (Table 2). Agricultural activities or other anthropogenic disturbances were not observed in the study area. The dominant vegetation in the study area is low-grown grassland.

The vertical soil profiles were sampled from the small area with similar amount of precipitation along two transects, i.e., from 6 sampling sites (8, 9, 10, 11, 12, and 13) along a 3-km long north–south-oriented transect and from 12 sampling sites (1, 2, 3, 4, 5, 6, 7, 14, 15, 16, 17, and 18) along an east–west-oriented 12.5-km-long transect (Fig. 1). According to the

Table 1 Basic descriptive statistics of particle size distribution in the analyzed soil profiles

	Sand (2.0–0.05 mm) (%)	Silt (0.05–0.002 mm) (%)	Clay (<0.002 mm) (%)
Mean	51	38	11
Median	52	39	6
Mode	28 ^a	29 ^a	3
Std. deviation	17	11	8
Variance	306	111	71
Range	68	48	32
Minimum	19	9	0.5
Maximum	86	57	33

^a Multiple modes exist. The smallest value is shown

erosion map of the Republic of Serbia (Lazarević 1983), this area is under the influence of strong water erosion. The coordinates of sampling sites are given in Table 2.

From each location, two to three subsamples of soil were collected, using the template method (IAEA 2004). For each soil sample collected, an area of 1 m × 1 m was marked and cleaned from decayed organic matter, plant roots, stones, and other debris. All samples were taken with a stainless steel spade, packed in plastic bags, and

transported to the laboratory. In the laboratory, all soil samples were further cleared from the residual foreign material (the rocks and plant roots), dried at room temperature to constant weight, mechanically ground (using mortar and pestle), and then sifted through a sieve (2-mm-mesh size) (IAEA 1989). The soil samples were then packed into 0.5-L Marinelli beakers, and the total weight of each sample was recorded. The weights of dried samples ranged from 432 to 845 g.

Table 2 Coordinates and slope of the sampling sites and ¹³⁷Cs activity concentration in surface soil layers

Sampling site	Coordinates		Slope (%)	¹³⁷ Cs activity concentration in surface soil layers (0–5 cm)
	Latitude	Longitude		
1	42.395	21.921	28	10
2	42.388	21.958	25	25
3	42.379	21.966	63	56
4	42.378	21.967	22	32
5	42.386	22.010	38	32
6	42.386	21.961	24	2
7	42.386	21.961	16	14
8	42.353	21.887	12	115
9	42.353	21.887	10	7
10	42.348	21.886	15	53
11	42.340	21.891	23	86
12	42.336	21.897	18	184
13	42.358	21.885	54	97
14	42.394	21.929	24	29
15	42.382	22.061	48	61
16	42.382	22.050	63	84
17	42.387	22.010	24	184
18	42.390	21.954	25	21
Standard deviation (Bq kg ⁻¹)				56
Mean (Bq kg ⁻¹)				61
Coefficient of variation (%)				92
Minimum (Bq kg ⁻¹)				2
Maximum (Bq kg ⁻¹)				184

3.2 Analytical methods

Measurements of the ^{137}Cs activity concentration were performed using the HPGe gamma-ray spectrometer ORTEC-AMETEK (34 % relative efficiency and high resolution 1.65 keV at 1.33 MeV for ^{60}Co), from its gamma-ray line at 661.2 keV. The activity concentration of ^{137}Cs was calculated based on the equation:

$$A_{E_i} = \frac{N_{E_i}}{\varepsilon_E \times t \times \gamma_d \times m} \quad (1)$$

where A_{E_i} —the activity concentration of nuclide i based on energy E (Bq kg^{-1}); N_{E_i} —the net peak area for peak at energy E ; ε_E —the detector efficiency at energy E ; t —the live time; γ_d —the gammas/disintegration for energy E of this nuclide; and m —the sample mass (kg).

For the analyses of the gamma-ray spectra, the Gamma Vision 32 MCA emulation software was used (ORTEC 2001). The standard reference material MBSS type, provided by the Czech Metrological Institute, Prague, with the total activity of 38 kBq on 16 August 2010 (^{241}Am , ^{109}Cd , ^{139}Ce , ^{57}Co , ^{60}Co , ^{137}Cs , ^{113}Sn , ^{85}Sr , ^{88}Y , ^{203}Hg) was used for calibration of the detector. The total measurement errors of the activity concentrations were up to 10 %. The values of the ^{137}Cs activity concentration presented in this study refer to date of sampling.

For particle size analysis (sand (2.0–0.05 mm), silt (0.05–0.002 mm), and clay (<0.002 mm)), the pipette method was used (Rowell 1997).

For descriptive statistic of the results, the software package SPSS 16.0 (Statistical Package for the Social Sciences) for Windows was used (SPSS 2007).

4 Results and discussion

The ^{137}Cs activity concentration obtained for the top layer (0–5 cm) of each soil profile is presented in Table 2. The ^{137}Cs activity concentration ranged from 2 to 184 Bq kg^{-1} with the average of 61 Bq kg^{-1} and standard deviation of 56 Bq kg^{-1} . The high coefficient of variation (ratio of the standard deviation to the mean values) of 92 % indicates high spatial variability and large range of the ^{137}Cs activity concentrations in the study area. The spatial variation in the ^{137}Cs activity concentration in the study area may be due to differences in the quantity of ^{137}Cs activity concentration fallen on a surface of study area after the Chernobyl accident (differences in deposition) or due to concentration factors (factors which determine an uneven repartition of radioactivity in soils). In the case of primary direct deposition, the most important factors affecting the creation of horizontal radioactivity patterns are the amount of precipitation and type of rainfall (Nimis 1996).

Although the study area is very small, the influence of the amount of precipitation and type of rainfall on the spatial variability of the freshly deposited ^{137}Cs fallout over the study area cannot be completely excluded. The differences in ^{137}Cs activity concentration in surface soil layer (0–5 cm) among sampling locations within transects (Table 2) were also very pronounced, although the locations are very close to each other. When ^{137}Cs are deposited from the atmosphere, one part of the ^{137}Cs reaches the ground while another part is being intercepted by vegetation. The intercepted ^{137}Cs may be absorbed by vegetation and released to soils when vegetation dies and decays (depending on vegetation type and climate conditions) or adsorbed and then washed off by precipitation and moved to the soil (Ritchie and McHenry 1990). The area over which samples were collected is a relatively simple system, i.e., grassland, where interception and retention of the freshly deposited ^{137}Cs fallout by vegetation may have just slight influence on the observed spatial variation in the ^{137}Cs activity concentration in the area. According to Nimis (1996), the horizontal patterning of radioactivity is less pronounced in simpler systems (grasslands) in comparison with structurally more complex systems (multi-layered forests). As the soil profiles were sampled from the slope, it is possible that very small part of fresh ^{137}Cs fallout was moved from the landscape during heavy rainfall events before being adsorbed by soil (Ritchie and McHenry 1990). This might be one of the reasons for the observed spatial variation in the ^{137}Cs activity concentration in the study area. After reaching the soil surface, ^{137}Cs was rapidly and strongly adsorbed by organic colloids and clay particles (Nimis 1996), after which it was essentially non-exchangeable. Major causes of redistribution of the ^{137}Cs in soils and its movement from soils to water are physical processes associated with erosion, transport, and deposition of sediment particles, while mobility by biological and chemical processes are limited (Ritchie and McHenry 1990; Nimis 1996). As ^{137}Cs is rapidly and strongly fixed after reaching the soil surface as fallout, and any subsequent redistribution of ^{137}Cs within the landscape will occur only in association with the erosion, transport, and deposition of soil and sediment particles, ^{137}Cs may be used as an effective sediment tracer and provide a basis for estimating erosion and deposition rates (Walling and He 1999; Porto et al. 2001, 2003; Hacıyakupoglu et al. 2005; Walling et al. 2006).

The erosion may be the main reason for the observed spatial variation in ^{137}Cs activity concentration in the study area, due to fact that erosion in the study area is an extremely pronounced long-term process (Lazarević 1983). Rogowski and Tamura (1970) found that movement of ^{137}Cs by runoff and erosion may be significant. Bonnett (1990) observed the redistribution of ^{137}Cs and ^{134}Cs by erosive processes over an area of a drainage basin. Slope gradient, length, and shape also affect runoff generation and soil erosion (Şensoy and Kara 2014). As the soil profiles in this study were collected from

Table 3 Basic descriptive statistics of ¹³⁷Cs activity concentration in the analyzed soil profiles

Parameter	¹³⁷ Cs (Bq kg ⁻¹)
Mean	20
Standard error of mean	3
Median	7
Mode	0.8
Standard deviation	36
Skewness	3
Standard error of skewness	0.2
Kurtosis	11
Standard error of kurtosis	0.4
Range	184
Minimum	0.3
Maximum	184

slopes at randomly selected locations (Table 2), it is not possible to directly correlate the ¹³⁷Cs activity concentration with slopes.

The basic descriptive statistics of the ¹³⁷Cs activity concentration across all 18 soil profiles (for all soil layers) collected from the study area are presented in Table 3. The mean value of ¹³⁷Cs was found to be 20 Bq kg⁻¹, ranging from 0.3 to

184 Bq kg⁻¹. The reported value is in accordance with the previous study conducted on the territory of Serbia (Dragović et al. 2012a).

Vertical distribution of the ¹³⁷Cs activity concentration in the soil profiles is shown in Fig. 2. In the soil profiles I, XIV, and XVI, sampled to the depth of 20 cm, the ¹³⁷Cs activity concentration decreased with the soil depth, but in the soil profiles XIV and XVI, the ¹³⁷Cs activity concentration decreased more rapidly in comparison with the soil profile I. In the soil profiles VIII, XIII, and XV, sampled to the depth of 25 cm, the ¹³⁷Cs activity concentration decreased rapidly, with a maximum in the first layer (0–5 cm). In the soil profile XVII, sampled to the depth of 25 cm, the ¹³⁷Cs activity concentration decreased slowly from the first (0–5 cm) to the third (10–15 cm) soil layer and below that it rapidly decreased. In the soil profile XI, sampled to the depth of 30 cm, the ¹³⁷Cs activity concentration decreased rapidly, with a maximum in the first layer (0–5 cm). In the soil profiles X and XII, sampled to the depth of 40 cm, the ¹³⁷Cs activity concentration goes down very quickly from the first (0–5 cm) to the second (5–10 cm) soil layer. The measured ¹³⁷Cs activity concentrations in the soil profiles II–VII, IX, and XVIII, sampled to the depth of 50 cm, were significantly lower in comparison to the previous soil profiles, while the decrease of the ¹³⁷Cs activity concentration with the soil depth was much less pronounced. In the soil profiles II, IV, V, and XVIII, the maximum of the

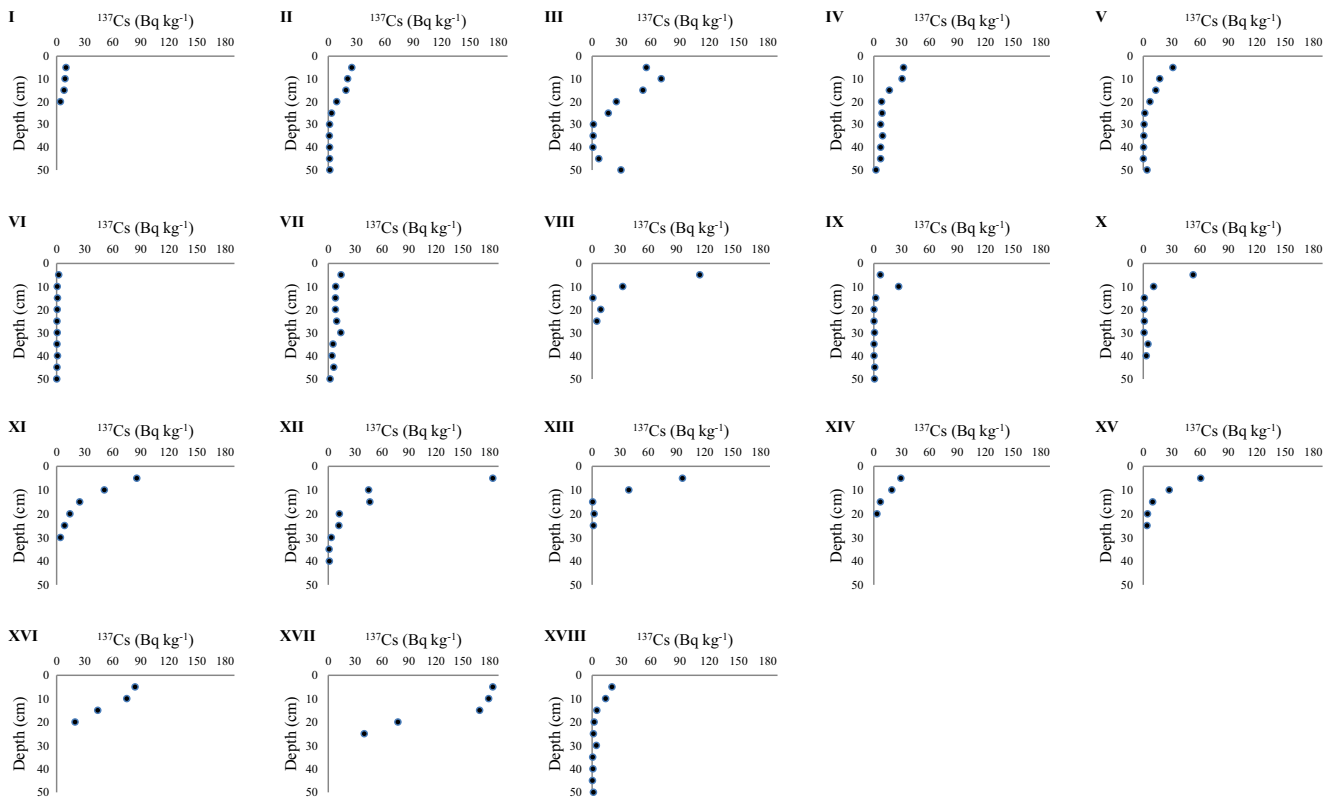


Fig. 2 Vertical distribution of ¹³⁷Cs activity concentration in the soil profiles (I–XVIII) from the erosive area of southeastern Serbia (Pčinja and South Morava River Basins)

^{137}Cs activity concentration occurred in the first soil layer (0–5 cm) and decreased with the soil depth, but the reduction of the activity concentration was just weakly pronounced. In the soil profiles III and IX, the maximum of the ^{137}Cs activity concentration occurred in the second soil layer (5–10 cm), and the soil profile III has shown a peak at the depth of 50 cm that may be caused by a variety of factors. In the soil profiles VI and VII, the ^{137}Cs activity concentration was almost uniform along the entire soil profile.

In the greatest number of the soil profiles, the ^{137}Cs activity concentration was highest in the first soil layer (0–5 cm) and decreased with the soil depth, confirming that these soils have not been cultivated. However, in the soil profiles III and IX, the peak of the ^{137}Cs activity concentration occurred in the second soil layer (5–10 cm), while in the soil profiles VI and VII, the ^{137}Cs activity concentration was almost equal throughout the soil profile. The migration and retention of the ^{137}Cs through the soil profile is affected by small differences in texture across different soil layers. In sandy soils, migration of the ^{137}Cs is faster, due to numerous pores through which water can drain freely. On the other hand, high clay content also reduces the movement of water through the soil and results in a slower migration of ^{137}Cs ; slowed migration can also be a consequence of their higher exchange capacity. Dragović et al. (2012a) found a positive correlation between the clay content and the ^{137}Cs activity concentration, while Petrović et al. (2013) found no correlation between sand or clay content and the ^{137}Cs activity concentration.

Biological, chemical, and physical factors influence migration and retention of ^{137}Cs through the soil profile, causing differences in retention and migration of ^{137}Cs among the soil layers. Further investigations of the soil erosion rate at the sampling location as well as the physicochemical characteristics of the analyzed soil profiles are necessary to explain migration and retention of ^{137}Cs at these locations in more detail.

5 Conclusions

The obtained results indicate that ^{137}Cs is present in the upper layers of the greatest number of soil profiles and that its concentration decreases with depth, as typical for uncultivated land. The ^{137}Cs activity concentration in the soil profiles (for all soil layers across 18 sampling sites) ranged from 0.3 to 184 Bq kg⁻¹, with a mean value of 20 Bq kg⁻¹. In the surface soil layers (0–5 cm), the ^{137}Cs activity concentration has shown a wide range of values from 2 to 184 Bq kg⁻¹ with an average of 61 Bq kg⁻¹, indicating a very uneven spatial distribution of ^{137}Cs activity concentration in the study area. The area is under the influence of strong erosion, which may be one of the main reasons for such behavior of ^{137}Cs in the study area. Additional investigations to verify this hypothesis are required. As distribution of ^{137}Cs is influenced by a

number of physical, chemical, and biological properties of the soil, further investigations should include the analyses of both, the pedogenic factors (pH, specific electrical conductivity, total exchange capacity, organic matter content, carbonates content, metals content, and soil particle size distribution) as well as the examination of bioturbation processes, i.e., activities of biological organisms in the soil.

Acknowledgments This work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia (Project no. III43009).

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