DEPTH DISTRIBUTION OF ^{137}CS IN SOILS FROM SPECIAL NATURE RESERVE BANAT SANDS, SERBIA AND ASSESSMENT OF DOSES TO NON-HUMAN BIOTA

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Abstract: A study was carried out to estimates the profile distribution of artificial radionuclide ^{137}Cs in soils from Special Nature Reserve Banat Sands, Serbia, the largest European sand area, and the dose rates to terrestrial biota resulting from exposure to the Chernobyl-derived radionuclide. The relatively low level of the ^{137}Cs was detected in soils from the study area. Calculated dose rates were below the screening level of 10 μGy h^{-1}, indicating no significant risks to the terrestrial biota. Findings presented in this study could serve as a baseline investigation for a subsequent exposure of plants and animals in their natural habitats due to the increasing level of the ^{137}Cs into the environment as a consequence of possible new release.

Key words: ^{137}Cs · Soil · Depth distribution · Non-human biota · ERICA tool

1. Introduction

Artificial, i.e., man-made radionuclide ^{137}Cs ($t_{1/2}$ = 30.17 years) is one of the essential radionuclides released into the environment as a result of atmospheric nuclear weapons tests and accidents at Chernobyl (1986) and Fukushima Daiichi (2011) nuclear power plants. According to the available data, before the 1986 Chernobyl accident ^{137}Cs activity concentrations in the Serbian soils were below 5 Bq kg^{-1} (Popović and Spasić-Jokić, 2006).

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Following the Chernobyl accident, soils on the territory of Serbia became contaminated with ^{137}Cs, and its spatial distribution was reported to be influenced by local meteorological conditions and site-specific variables (Dragović et al., 2012b). Results of extensive study show that ^{137}Cs activity concentrations in surface soils (0-2 cm) collected during 2001 from Vojvodina, northern province of Serbia, varied from 1.1 Bq kg^{-1} for Horgoš to 55 Bq kg^{-1} for Bavanište (Bikit et al., 2005). The ^{137}Cs from the Fukushima nuclear accident was not detected in Vojvodina (Bikit et al., 2012). The assessment of radiation doses to human and non-human biota based on soil ^{137}Cs activities is very important due to long half-life of this radionuclide. Numerous models/approaches have been developed to assess the radiation risk to non-human biota (Beresford et al., 2008; Stark et al., 2015; Vives i Batlle et al., 2007). ERICA (Environmental Risk from Ionising Contaminants: Assessment and Management) Tool software system was applied in a number of studies worldwide to calculate dose rates to non-human biota based on activity concentrations of natural and artificial radionuclides in soils (Černe et al., 2012; Ćujić and Dragović, 2018; Oughton et al., 2013; Sotiropoulou et al., 2016; Wood et al., 2008). The assessment of radiation doses to terrestrial non-human biota is of particular importance in Banat Sands (also there is few synonyms currently used in literature: Deliblato Sands or Banat Sandstone), for several reasons: it is the largest continental sand area in Europe; it has been under the protection as a nature reserve since 1977; it is very important natural habitat for numerous species of wild animals, birds (in 1989 was declared an Important Bird Area - IBA (Simić and Puzović, 2008) and plants (in 2005 was declared an Important Plant Area - IPA (Radford and Odé, 2009); and the dune grasslands of this area are used for livestock grazing.

2. Materials and methods

Study area

Banat Sands is located in the southeastern sector of the Pannonian Plain in the southern part of Serbian geographic region Banat (Fig. 1). It extends the length of about 35 km and a maximum width of about 15 km. It covers an area about 300 km^2, with elliptical shape stretching in southeast-northwest direction. The landscape of Banat Sands is unique among other parts of the Pannonian Plain because altitudes gradually increase from southeast to northwest but also from southwest to northeast. The dune-deflation relief dominates, as the result of intensive eolian and accumulation processes (Dragović, 2001).

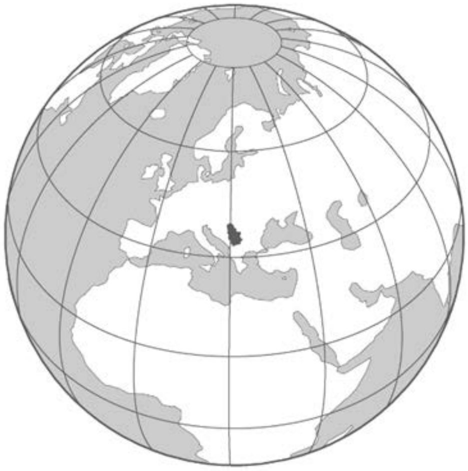

Fig. 1. Banat Sands region within Serbia

The relief of Banat Sands is created by phase action of dominant north-east and south-east winds, by deflation and sand sedimentation (Ivanović, 1975). The major part of the sand-loess complex of Banat Sands is composed of eolian sand sediments, initially formed as alluvial sands that were then blown up by the southeastern wind and accumulated as aero-sediments at the places of the lower wind intensity. The pure eolian sands are of coarse-grained and pulverulent texture, composed mainly from quartz, feldspar, mica, granat, epidote, and iron oxides (Ivanović, 1975). According to Cholnoky (1910) and Milojević (1949), the eolian sands are of Pliocene age. Sandy-loess accumulations spread over the northwestern part of Banat Sands are presented by fine-grained sands, which are accumulated either over eolian ones or lake sands. In this area, there are many loess sinkholes, shallow, oval, and oblong in shape, formed during the processes of dissolution of carbonates from loess and their sedimentation as concretions at the sinkhole bottom. The sandy loess is characterized by the combined intergrain and capillary porosity (Ivanović, 1975). The belts of loess accumulation spread over the western part of Banat

Sands are characterized by intergrain and capillary porosity in their surface horizons (Ivanović, 1975).

The climate of the investigated area is semi-arid continental with elements of steppe climate and belongs to b type according to Köppen climate classification (Köppen, 1936). The winters are cold with low snow precipitation, summers are warm and dry, and autumns are warmer than springs. The mean annual temperature is 10.4 °C, with minimum values by the end of January and the beginning of February and maximum values by the end of July and the beginning of August. The mean annual precipitation is 660 mm, with two maxima, in June and November. According to the values of the Lang rain coefficient (calculated as a ratio between mean annual precipitation and mean annual temperature) of about 60 (with decreasing values towards peripheral areas up to 40), the area belongs to the climate of low forests (Lang, 1920). The southeastern Košava wind significantly influences the climate of the area, blowing from the Carpathian Mountains in the southeast. Soils are represented by a number of varieties of basic soil types. Among undeveloped soils the most abundant is *Albic Arenosol*. Above them the fragments of *Cambisol* and *Eutric Cambisol* occur. They are characterized by low water-holding capacity. For almost two centuries of afforestation, the quicksand is stabilized, and processes of establishment of forest and meadow soils began. Flora of Banat Sands is represented by over 900 species, with some rarities and relicts. Vegetation cover was formed in several phases. Steppe grass and shrub xerophile formations were established as pioneer vegetation, which stabilized the sand and enabled the occurrence of the first stands of dendroflora. The most abundant representatives are acacia and Scotch and black pine. About 97% of forest stands of Banat Sands are formed by afforestation. Fauna of Banat Sands is represented by typical steppe-forest species of insects and wild game.

Soil sampling and preparation

A total of fifteen soil profiles were collected during the period of 2012/2013 from five different locations from the edge of the Banat Sands area (Fig. 1), where dunes with steppe-grassland plains were found. At each location, soil profiles were collected from the dune top, leeward slope, and inter-dune depression (hollow). Soil samples were collected at 5 cm intervals to a depth of 40 cm, obtaining a total of 120 interval samples. All collected soil samples were dried and homogenized mechanically.

Analytical methods

The soil samples in the 0.5 L Marinelli beaker were measured using ORTEC-AMETEK HPGe gamma-ray spectrometer. Obtained gamma-ray spectra were analyzed using Gamma Vision 32 MCA emulation software (ORTEC, 2001). The activity of ^{137}Cs in the soil samples was determined using its gamma line at 661.6 keV, and the ^{137}Cs concentration was expressed as activity per unit mass (Bq kg^{-1}). The MBSS2 calibration source in 0.5 L Marinelli beaker (containing a mixture of radionuclides) was used for energy, and efficiency calibration of the HPGe gamma-ray spectrometer and the calibration was checked using IAEA-RGU-1 and IAEA-RGTh-1 reference materials. Soil properties (sand, clay and silt contents, carbonate content, pH(in H_2O), specific electrical conductivity, organic matter content) were determined using standard methods (ISO 10390, 2005; ISO 10693, 1995; ISO 11265, 1994; Rowell, 1997; Simakov, 1957). The ERICA Assessment Tool (version 1.2.1) was applied in order to calculate external, internal and total dose rates for reference organisms of terrestrial ecosystem available in model (ERICA, 2007). Details on ERICA Tool can be found in different studies (Brown et al., 2008, 2016). The input data consisted of maximal measured activity concentration of ^{137}Cs in soils in order to ensure that the maximum possible value of dose rates to non-human biota was below the screening dose rate criterion of 10 μGy h^{-1}. For statistical analysis of data the Statistical Package for Social Science - SPSS 16.0 software package was used (SPSS, 2007).

3. Results and discussion

Distribution of ^{137}Cs in soils and its relationship with soil properties

Results obtained in this study show that ^{137}Cs was detected in collected soil samples (Table 1), and varied between 0.2 and 168 Bq kg^{-1} (mean value: 15 Bq kg^{-1}). Results of extensive study show that ^{137}Cs activity concentrations in surface soils (0-5 cm) collected during the 2003 from 15 locations in Serbia and Montenegro, varied from 5.25 Bq kg^{-1} to 112 Bq kg^{-1} (mean value: 48.3 Bq kg^{-1}) (Dragović and Onija, 2006), which is in accordance with values reported in this study.

Table 1. Basic descriptive statistics of ^{137}Cs activity concentrations (Bq kg^{-1}) in the soil profiles.

Depth (cm)	Mean	Std. Deviation	Minimum	Maximum
0-5	46	47	4.9	168
5-10	29	22	4.1	81
10-15	17	17	2.0	59
15-20	12	15	0.2	47
20-25	4.4	4.5	0.3	17
25-30	3.6	3.8	0.3	13
30-35	3.7	4.4	0.3	14
35-40	2.7	3.2	0.3	12
Total	15	24	0.2	168

Depth distributions of the ^{137}Cs activity concentrations in the soil profiles down to 40 cm are presented in Fig. 2. In soil most of the analyzed soil profiles, the maximum activity concentration was found in the top 0-5 cm layer, and a concentration of ^{137}Cs decreased with soil depth (see Fig. 2), the shape of the profiles conforms to that expected for an uncultivated site. In soil profiles, 2-c, 3-c, and 5-c collected from the inter-dune depression, the highest ^{137}Cs concentration was observed at deeper soil layers (see Fig. 2). In all soil profiles collected from location 4, the highest ^{137}Cs concentration was found in deeper layers (see Fig. 2). Vertical and horizontal migration of ^{137}Cs in soils and resulting profile distributions are site-specific and depend on number of factors, such as type of soil, land use and soil management practices, soil properties (*e.g.* texture, organic matter content (SOM) and pH), climatic conditions (*e.g.* rainfall, temperature, or humidity), bioturbation etc., physical processes of soil erosion and deposition can be involved in the redistribution of ^{137}Cs in soils (Al-Masri, 2006; Begy et al., 2017; Gaspar and Navas 2013; Müller-Lemans and van Dorp, 1996; Owens and Walling, 1996; Ramzaev and Barkovsky, 2018; Iurian et al., 2012). According to Kadović et al. (2016) area of Banat (Deliblato) Sands is highly sensitive to degradation, 43.18% of total area belongs to medium sensitivity class, and 56.26% to high sensitivity class. In the recent study about wind erosion conducted in the same study area, based on ^{137}Cs and $^{210}Pb_{ex}$ measurements, significant erosion rates, especially on the tilled area were found (Krmar et al., 2015). By considering all analyzed soil profiles, most of the ^{137}Cs (87.7%) was found within the first 20 cm of soils, and a significant decrease of ^{137}Cs with depth was observed (Fig. 2, Table 1). Below 20 cm soil depth low levels of ^{137}Cs activity concentrations were found, and ^{137}Cs showed minimal change with depth (Fig. 2, Table 1). Yan and Shi

(2004) reported different ^{137}Cs depth profile in dune land, in shift dunes, ^{137}Cs was distributed throughout the entire sand layer while in fixed dunes ^{137}Cs decreased sharply below the 15 cm depth.

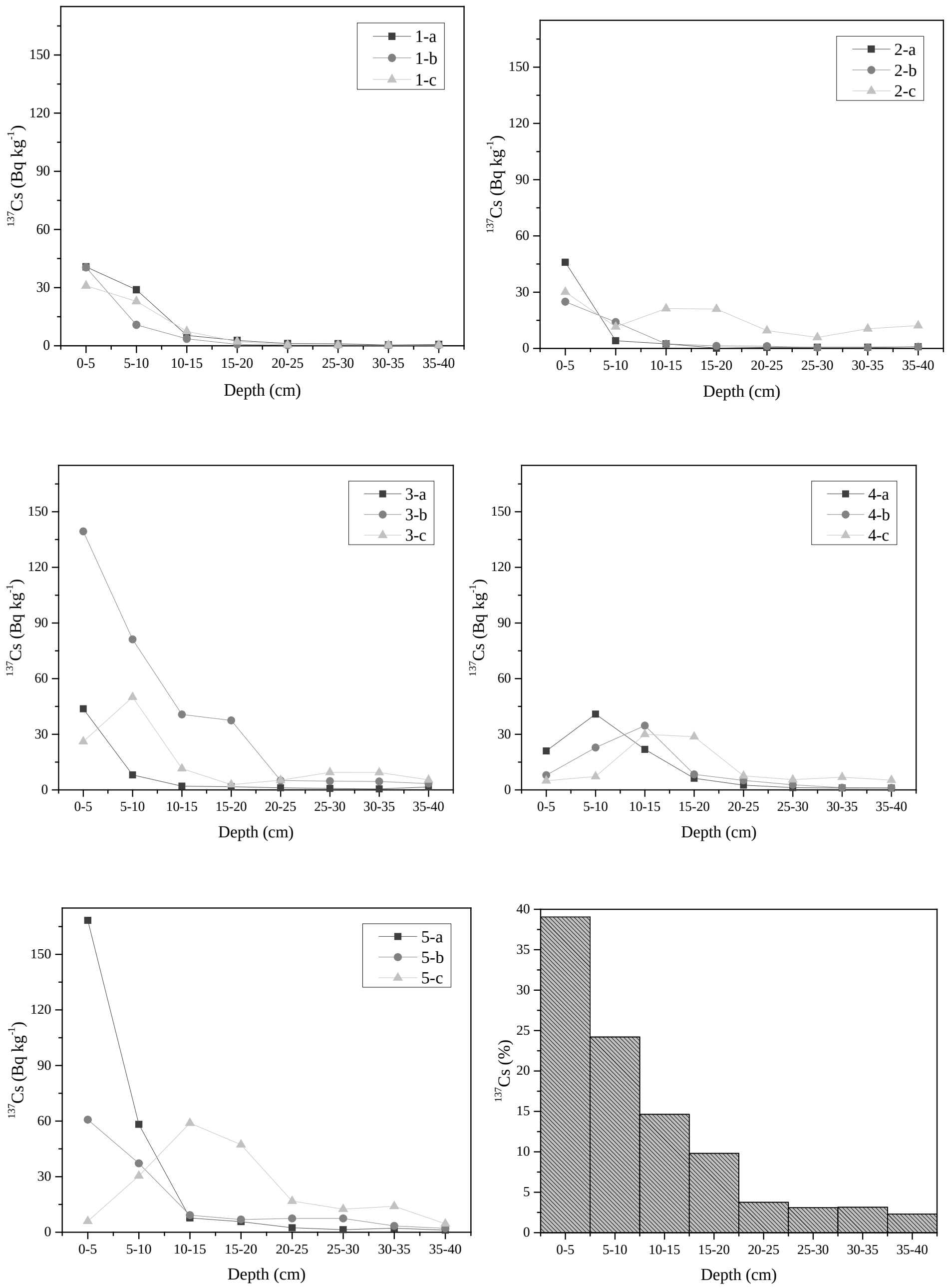

Fig. 2. Depth distribution of ^{137}Cs in soil profiles

The relations between ^{137}Cs content in soils and different soil parameters were determined (Table 2).

Table 2. Correlation coefficients between ^{137}Cs and soil properties.

Parameters	^{137}Cs (Bq kg^{-1})
Coarse sand 2-0.2 mm (%)	0.024
Fine sand 0.2-0.05 mm (%)	-0.196*
Silt 0.05-0.002 mm (%)	0.181*
Clay <0.002 mm (%)	-0.137
Carbonates (%)	-0.156
pH (H_2O)	-0.483**
Spec. el. cond. (μS cm^{-1})	-0.131
Org. matter (%)	0.422**

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

The ^{137}Cs activity concentration was negatively correlated with the fine sand content and positively correlated with silt content (see Table 2) which agrees with the findings of Dragović et al. (2012a). Clay minerals, particularly illites, have a strong affinity for cesium due to its small hydration energy and the presence of frayed edge sites (FES) in the minerals (Dumat et al., 1997; Dumat and Staunton, 1999; Staunton et al., 2002). Nevertheless, no significant correlation was found between ^{137}Cs values and clay content of analyzed soils (see Table 2). According to results of Gaspar and Navas (2013) and Navas et al. (2011) the lack of significant correlation can be a consequence of homogenous depth distribution of clay in soils and its limited range of variation. A negative correlation was observed between the ^{137}Cs activity concentration and the pH (see Table 2), which agrees with the findings of Gaspar and Navas (2013) and Iurian et al. (2014). The positive correlation between ^{137}Cs activity concentration and organic matter content was found (Table 2), which is in accordance with results obtained in several studies (Iurian et al., 2014; Milenkovic et al., 2015; Navas et al., 2011; Petrović et al., 2013). Ritchie and McCarty (2003), Ritchie et al. (2007) and Martinez et al. (2010) found strong and statistically significant correlations between ^{137}Cs and soil organic carbon (SOC) in agricultural landscapes suggesting that ^{137}Cs and SOC are moving along similar physical pathways and by the same mechanisms. Iurian et al. (2014) reached similar conclusions, and they found a strong and statistically significant correlations between ^{137}Cs and soil organic matter in uncultivated landscapes. In contrast, Martinez et al. (2010) did not find relationship between ^{137}Cs and SOC in undisturbed landscapes suggesting the impact of biological factors (e.g. biological oxidation, mineralization) on SOC spatial distribution. Different configurations of ^{137}Cs depth profile reported in study of Iurian et al. (2012) have been attributed to the percolating water, growth conditions of microflora or biotic interactions within the soil.

Dose rates to terrestrial organisms

The results of external, internal, and total dose rates to terrestrial reference organisms calculated using the ERICA assessment tool are presented in Fig. 3.

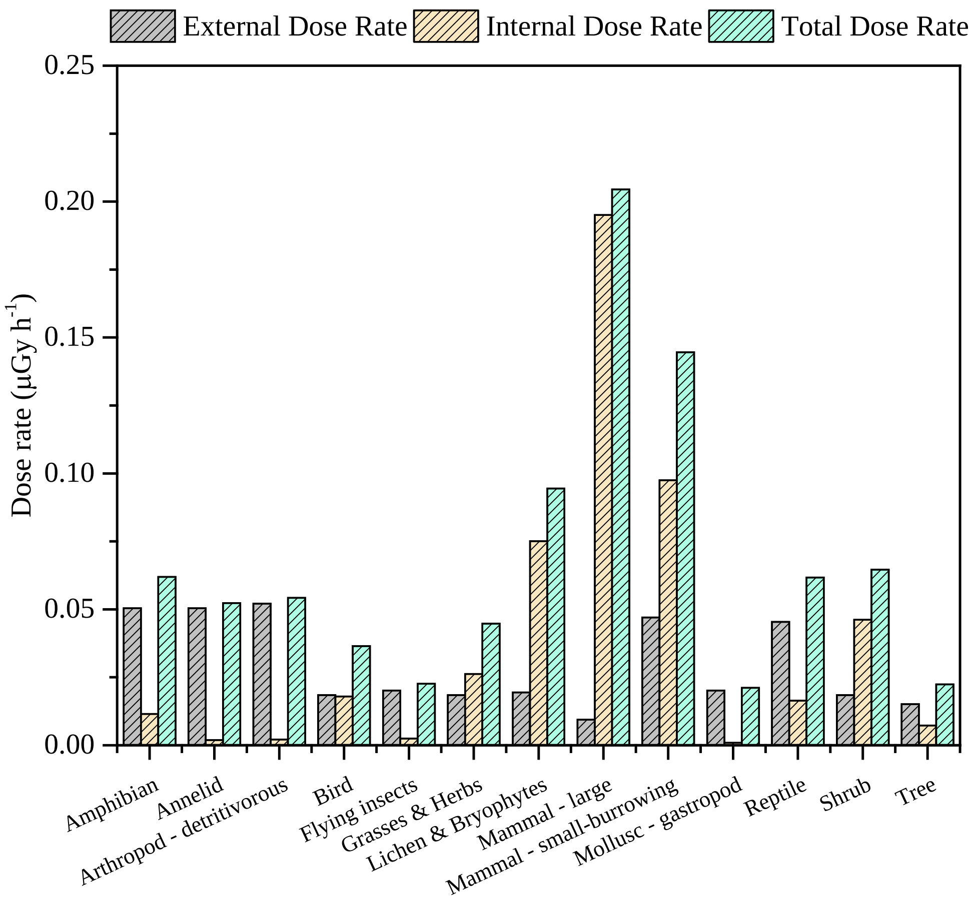

Fig. 3. Dose rates for terrestrial reference organisms calculated using ERICA Tool

The internal dose rates comparing to external dose rates were found to be higher for following terrestrial reference organisms: grasses and herbs, lichen and bryophytes, large and small burrowing mammal, and shrub (Fig. 3). According to the results of Ćujić and Dragović (2018) ^{137}Cs contributes mostly to the external dose rate. In the present study, the highest total dose rate calculated for a large mammal (Fig. 3) are still much below the value of the screening dose rate of 10 µGy h^{-1}. The obtained results indicate that terrestrial biota is exposed to low dose rates and that the risk form ^{137}Cs in the soil is insignificant. Similar dose rates due to ^{137}Cs in soils were found to terrestrial biota in the area around coal-fired power plant (CFPP) complex "Nikola Tesla" in Serbia (Ćujić and Dragović, 2018) and Belgrade area (Petrović et al., 2018). Nevertheless, it is important to stress that despite the low level of dose rates, this study confirms that terrestrial biota has been exposed to ^{137}Cs of Chernobyl origin. The data obtained in this study can be used as a baseline level for future radiological assessments in the study area.

4. Conclusions

The results of gamma spectrometry showed that ^{137}Cs of the Chernobyl origin is still present in soils of the special nature reserve Banat Sands. The vertical distributions of ^{137}Cs were found to be positively correlated with silt content and organic matter content, and negatively with fine sand content and soil pH. Results of the ERICA Tool show that terrestrial non-human biota is exposed to low dose rates from ^{137}Cs since all calculated dose rates are much below the value of screening dose rate. The presented results contribute to knowledge about ^{137}Cs distribution and factors affect its mobility in dune fields, as well as about radiation doses to terrestrial non-human biota in the protected area declared as Important Bird Area and Important Plant Area.

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