VERTICAL DISTRIBUTION OF ^{137}Cs IN CULTIVATED AND UNDISTURBED AREAS

by

Snežana S. NENADOVIĆ[1]***, Miloš T. NENADOVIĆ***[2*]***, Ivana S. VUKANAC***[3]***,***
Aleksandar R. DJORDJEVIĆ[4]***, Slavoljub S. DRAGIĆEVIĆ***[5]***, and Milutin A. LJEŠEVIĆ***[5]

[1]Laboratory for Material Sciences, Vinča Institute of Nuclear Sciences, Belgrade, Serbia
[2]Laboratory for Atomic Physics, Vinča Institute of Nuclear Sciences, Belgrade, Serbia
[3]Laboratory for Nuclear and Plasma Physics, Vinča Institute of Nuclear Sciences, Belgrade, Serbia
[4]Faculty of Agriculture,University of Belgrade, Belgrade, Serbia
[5]Faculty of Geography, University of Belgrade, Belgrade, Serbia

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This paper introduces the distribution background and unique characteristics of ^{137}Cs in cultivated and undisturbed areas. The samples were taken from three measuring points of the depth of 1 m. We examined all visible horizons and determined their classifications. There were four horizons in one profile. All four horizons had a different zone thickness. ^{137}Cs is an artificial radionuclide that has been produced primarily as a result of atmospheric thermonuclear weapon tests since the 1950. Also, the great amount of ^{137}Cs (85 PBq) was released in the atmosphere during the Chernobyl accident. So, ^{137}Cs has been globally distributed, with fallout rates generally related to latitude and precipitation depth. The movement of ^{137}Cs in soil is primarily controlled by soil erosion processes, such as processes caused by water, wind, and tillage. Thus, ^{137}Cs is a valuable tracer to study soil erosion. The specific activity of ^{137}Cs in soil and sediment samples was determined by using the gamma-spectrometric method.

Key words: ^{137}Cs, erosion, soil, gamma-spectrometric method

INTRODUCTION

Soil erosion is a serious environmental problem. Traditional methods used to measure soil erosion are time consuming and the results obtained for an experimental plot are usually mutually incomparable. Soil degradation by water and wind erosion is a serious problem in most countries. Furthermore, loessial soils are very susceptible to water erosion processes. Even a small inclination of few degrees may result in medium to large water erosion of loessial soil. Additionally, inappropriate soil tillage intensifies soil erosion. To choose an appropriate method of soil conservation the rate of soil erosion should be known. There are many methods to estimate the intensity of soil erosion, but traditional methods have many limitations. Having this background in mind, the method of soil erosion measurement based on the ^{137}Cs activity measurements overcomes many limitations of the traditional methods to measure soil erosion as well as the empirical models such as USLE or RUSLE, or Gavrilovic method [1], used in former Yugoslavia. The detailed discussion of the advantages and limitations of the method based on ^{137}Cs activity was presented by Ritchie and McHenry [2]. This isotope has been present in the environment since the beginning of nuclear weapon testing and nowadays ^{137}Cs is a part of many ecosystems. ^{137}Cs seems to be a valuable tracer for studying soil erosion and sedimentation because after its deposition on the land surface it is rapidly and strongly adsorbed by soil particles [3] and thus its redeposition in the soil environment goes together with the soil particles. In a few words, this method is based on the comparison of the ^{137}Cs inventories in sampling points with a reference inventory. The reference inventory presents the local input fallout of ^{137}Cs. This means that for a reference site neither erosion nor deposition of soil has occurred. The estimation of the local input fallout of ^{137}Cs is problematic especially in regions where undisturbed sites are uncommon. The simple comparison between ^{137}Cs inventory at a sampling point and the reference ^{137}Cs inventory allows recognizing erosion and deposition areas; to obtain quantitative estimation of soil erosion, however, one of the models should be used [4]. Those relationships allow converting ^{137}Cs data into erosion rate data.

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* Corresponding author; e-mail: milosn@vinca.rs

This paper describes the measurement of ^{137}Cs activity in valley soil. We measured the activities of ^{137}Cs in soil samples in a small agricultural catchment. The study area was contaminated by ^{137}Cs originating from Chernobyl accident [5]. The method based on ^{137}Cs activity was used as a tool to investigate soil erosion and accumulation for small agricultural catchment. The value of the erosion rate was obtained by applying the mass balance models as well as the proportional model. The aim of this study is to evaluate the soil erosion and accumulation rates for the agricultural field located on the loess slope as well as to evaluate the structure of soil redistribution for the study catchment.

MATERIALS AND METHODS

The study area selected for the investigation is a small agricultural valley located at Rudovci tableland near Lazarevac, Serbia (44°22 N, 20°24 E). The average inclination of the slopes of the study area is 4-5° and the study area is 4 km^2. The average annual rainfall of the study area is 675 mm. Soil samples were collected at three measuring sites in December 2008.

The first measuring site was at 221 m above the see level, N 44° 22 46.2 , E 20°24 27.83 . This profile was divided into 4 zones. The samples were taken from the depth of 0-13 cm, 13-30 cm, 30-60 cm, and 60-100 cm.

The second measuring site was at 226 m above the see level, N 44°22 50.97 , E 20°24 56.51 . This site was also divided into 4 zones and the samples were taken from the following depths: 0-25 cm, 25-50 cm, 50-75 cm, and 75-100 cm.

The third measuring site was at 222 m above the see level, N 44°22 9.54 E 20°24 56.27 , divided into 4 zones and the samples were taken from the depth of 0-20 cm, 20-40 cm, 40-70 cm, and 70-100 cm.

The exact locations of each measuring site were determined by a GPS portable system (Garmin, 76S).

The samples in the undisturbed condition are used for the determination of mechanical, morphological and physical characteristics of the soil. Relative humidity and bulk density can also be determined. The samples in the undisturbed condition and in their natural state were taken from the same place where the samples for cultivated area had been taken from. Those samples were taken with special cylinder with the strictly defined volume of 100 cm^3, called Kopecky's cylinder. The cylinders have one sharp side for easier indenting the ground surface. The samples were taken from the surface to the horizon depth. The ground surface needs to be prepared for taking samples. It means that it needs to be clean and smooth, without roots and grass.

The bulk density of the soil was determined. Bulk density presents the ratio of soil mass dried at 105 °C and its volume in natural state defined by the Kopecky's cylinder. Data for 24 soil samples were taken into account in this research (tab. 1).

The vertical distribution of ^{137}Cs in the 12 soil samples was determined by gamma spectrometry measurements in the Laboratory for Nuclear and Plasma Physics, Vinča Institute of Nuclear Sciences, Belgrade. The samples were measured in Marynelly geometry, and the mass of each sample was approximately 0.5 kg. Radiological analysis was performed by means of a Schlumberger coaxial HPGe spectrometer (resolution:

Table 1. Bulk density of the samples

Sample	Profile	Sampling depth [cm]	Cylinder and sample mass [g]	Bulk density [gcm^{-3}]	Average bulk density [gcm^{-3}]
1	1	3-8	213.10	1.065	1.147
2			237.70	1.304	
3			212.00	1.073	
4	1	18-23	259.50	1.484	1.518
5			264.40	1.586	
6			256.30	1.484	
7	1	45-50	260.00	1.556	1.570
8			266.70	1.593	
9			261.80	1.561	
10	1	75-80	265.00	1.588	1.593
11			266.90	1.562	
12			271.20	1.631	
13	2	10.15	263.00	1.515	1.533
14			260.40	1.547	
15			256.10	1.538	
16	2	30-35	246.70	1.444	1.456
17			245.90	1.431	
18			255.90	1.493	
19	2	60-65	247.90	1.433	1.423
20			245.20	1.449	
21			248.40	1.387	
22	2	80-85	272.00	1.528	1.508
23			252.00	1.467	
24			267.90	1.528	
25	3	5-10	271.50	1.520	1.451
26			248.70	1.429	
27			244.10	1.403	
28	3	30-35	267.00	1.568	1.567
29			266.60	1.602	
30			258.90	1.529	
31	3	55-60	247.00	1.543	1.593
32			262.00	1.606	
33			266.90	1.630	
34	3	85-90	269.10	1.623	1.575
35			262.90	1.608	
36			257.80	1.494	

2.1 keV; relative efficiency: 15% at 1332.5 keV) associated with standard beam supply electronics units. The standardized solution of the common mixture of gamma-emitting radionuclides traceable to the Czech Metrological Institute was used to prepare the standards for the energy and efficiency calibration of the spectrometer in accordance with IAEA recommendations [6]. The measurement time ranged from 80 000 s to 230000 s. All spectra were recorded and analyzed using the Canberra's Genie 2000 software; net areas of the peaks were corrected for the background, dead time, and coincidence summing effects, applying the calculation method of Debertin and Schötzing [7]. The obtained specific activities are given in tab. 2 expressed in Bq/kg of dry soil mass. Quoted uncertainties (the confidence level of 1   ) were calculated by error propagation calculation. The combined standard uncertainties included the efficiency calibration uncertainty and the statistical uncertainties of the recorded peaks.

Table 2. Measured ^{137}Cs inventory at the sampling point

Samples	Profile	Depth [cm]	^{137}Cs [Bqkg^{-1}]
1	1	0-13	37.2 2.6
2	1	13-30	7.0
3	1	30-60	0.09 0.02
4	1	60-100	0.19
5	2	0-25	38.1 2.7
6	2	25-50	1.0 0.1
7	2	50-75	0.09 0.01
8	2	75-100	0.28
9	3	0-20	30.8
10	3	20-40	0.42
11	3	40-70	0.13
12	3	70-100	–

RESULTS

The results of the ^{137}Cs vertical distribution at the three measuring sites have been shown in fig. 1. The values showed a vide range of specific activity, from 38.1 Bq/kg on the surface layer, to 0.028 Bq/kg at the maximum of the depth of 100 cm. This can be explained by massive surface contamination caused by Chernobyl accident. ^{137}Cs isotopes were captured on the surface layers, and 23 years later, their diffusion to the bulk is almost gone. The small trace has been found at the bottom, where the pollutant could get by a diffusion mechanism enhanced by water. Nevertheless, ^{137}Cs half-life is about 30 years, which means that this isotope has not yet achieved a half of its concentration. It could take 7 more years to get the half of the initial concentration caused by the accident (fig. 1).

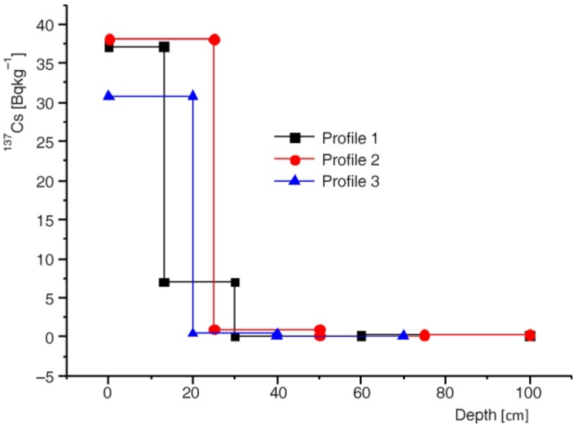

Figure 1. Dependence of ^{137}Cs radioactivity of sampling depth

As it is shown in fig. 1, the most significant migration of ^{137}Cs is at profile 2. It is in the range of 38.1   2.7 Bq/kg at the first horizon (0-25 cm) to 0.28   0.04 Bq/kg at the last horizon (75-100 cm).

The difference is smaller at profile 1, and it shows a step by step behavior. At the first horizon (0-13 cm), the measured value is 37.2   2.6 Bq/kg. At the second horizon (13-30 cm), this value is 7   0.6 Bq/kg, and this trend continues at the third horizon (30-60 cm) to 0.09   0.02 Bq/kg. At the last deepest horizon (60-100 cm), the value decreases to 0.019   0.03 Bq/kg.

Profile 3 shows a minor deviation for radioactivity values. The measured values for the horizons 0-20 cm, 20-40 cm, 40-70 cm, and 70-100 cm are 30.8   2.2 Bq/kg, 0.42   0,08 Bq/kg, 0.13   0,03 Bq/kg and 0 Bq/kg, respectively. Profiles 1 and 2 present cultivated areas and profile 3 presents an undisturbed area. There is a difference in calculating the erosion coeficient: for horizons 1 and 2 one model was applied, while for horizon 3 we used another model.

There are many relationships to obtain the quantitative estimates of soil erosion and/or deposition from ^{137}Cs measurements. In a few words, to obtain the quantitative value of soil erosion from cesium measurements, usually the initial input of the fallout of ^{137}Cs has to be known. Luckily, this input may be obtained for the area under study by relatively simple measurements of ^{137}Cs in soil in places without erosion or accumulation. Generally, the main idea of using ^{137}Cs measurements to study soil erosion is a comparison of the ^{137}Cs inventory for studying places with the local ^{137}Cs initial input. All approaches can be divided into two main groups: empirical relationships and theoretical models. Empirical equations have been established to explain the relationship between ^{137}Cs loss and soil loss. This approach demands the simultaneous measurement of ^{137}Cs loss from erosion plots (or the increase for accumulation plots) and soil loss. At present, a few empirical relationships exist [8]. Usually, the relationship between soil loss and ^{137}Cs loss is exponential. Although this approach is relatively easy to use, this kind of measurement of soil erosion has many limitations.

First, these empirical equations have been established for relatively small experimental plots and thus the use for other plots is rather difficult. Second, this equation is the same for undisturbed and cultivated areas. This can lead to the underestimation of soil erosion for cultivated fields and an overestimation of soil erosion for undisturbed sites. This is due to the different depth distribution of ^{137}Cs in the soil profile for the undisturbed and cultivated field [9].

The second group of approaches is to convert the ^{137}Cs activity data into soil erosion value, and it consists of theoretical models. Generally, all existing theoretical models could be divided into two main groups: models developed for undisturbed and for agricultural areas. There are two kinds of theoretical models to convert the ^{137}Cs data for the agricultural fields: proportional models and mass balance models. The proportional models have been established and used by many scientists. The proportional models were summarised by Walling and Quine. Brown *et al.* established the gravimetric model but this model is a kind of a proportional model [10-12]. There are a few different proportional models for the calculation of the value of soil loss but the differences between them are rather small. Those models are based on the assumption that ^{137}Cs is completely mixed within the plough layer. The soil loss is directly proportional to the ^{137}Cs loss from the soil profile. The equation can be written as [13]

$$Y \quad 10\frac{BdX}{100T} \tag{1}$$

where:

Y – rate of soil loss, [tha^{-1}yr^{-1}],

B – bulk density of the soil, [kgm^{-3}],

d – depth of the plough layer, [m],

X – percentage reduction in total ^{137}Cs invertory, defined as $(A_{\mathrm{ref}} \quad A)/\,A_{\mathrm{ref}} \;\; 100$,

A_{ref} – local ^{137}Cs reference inventory, [Bqm^{-2}],

A – measured ^{137}Cs inventory at the sampling point, [Bqm^{-2}], and

T – time elapsed since initiation ^{137}Cs accumulation, [yr].

As it is shown in fig. 2(a), the rate of soil loss decreases faster in profile 1 than in profile 2. Differences between these values are not big due to its very similar chemical and physical composition. Also, the distance of these two measuring points is small and we can say that it is almost the same soil composition. In fig. 2(b), the rate of soil loss has a bigger value for 0.5 tha^{-1}yr^{-1} in profile 2 at the depth of 25 cm and has a constant value up to the depth of 75 cm. After this depth, the rate of soil loss decreases for 0.2 tha^{-1}yr^{-1}. Otherwise, the same values for profile 1 at the depth from 30 to 60 cm are 0.4 tha^{-1}yr^{-1} and they are constant. At the depth of 60 to 100 cm there is a slight increase of the rate of soil loss for 1 tha^{-1}yr^{-1}.

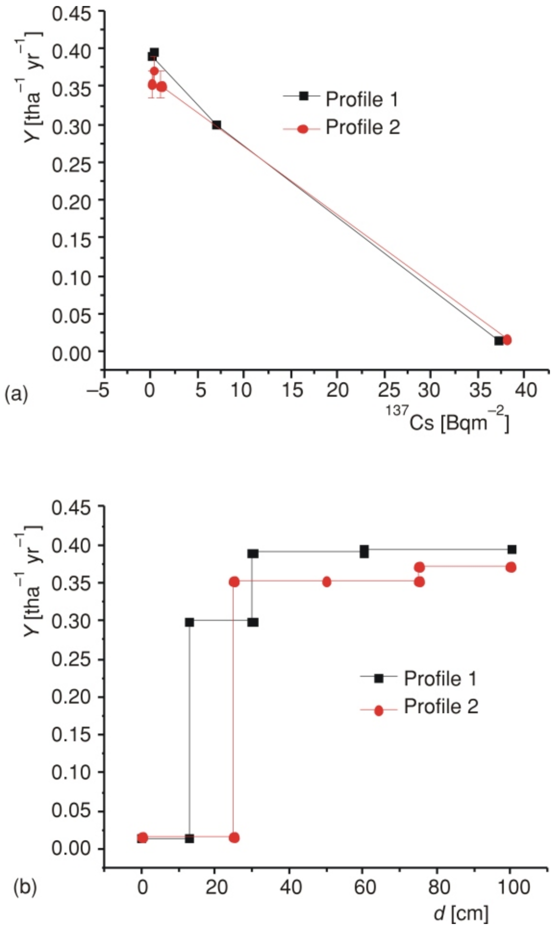

Figure 2. (a) Rate of soil loss dependence of activity at cultivated areas; (b) rate of soil loss dependence of sampling depth at cultivated areas

This approach has several limitations. First, this model assumes that cesium is uniformly distributed in the plough layer. Immediately after the fallout, the surface contained more cesium than the lower soil layer due to agricultural mixing, *e. g.* due to ploughing. It means that the results of the calculation of the soil loss may be overestimated [14]. Moreover, due to the selective removal of fine soil particles the results can be overestimated. This is because ^{137}Cs is more readily adsorbed on the fine soil fraction. In a nutshell, this approach has been widely used due to its simplicity although the results are sometimes inaccurate. To overcome the limitations of the proportional models (and also of the gravimetric approach), the mass balance model was proposed by Kachanoski *et al.* [15]. The authors supposed that the relationship between soil erosion and ^{137}Cs remaining was time-dependent.

They assumed that the relationship between the changing of ^{137}Cs in soil and soil erosion depended on the atmospheric deposition of ^{137}Cs and the tillage dilution of ^{137}Cs. The authors also considered the variation in soil erosion and ^{137}Cs deposition during a year.

The rate of soil erosion for undisturbed areas is usually much smaller than for cultivated areas. Moreover, due to the lack of mechanical mixing of soil during agricultural processes, the vertical distribution of ^{137}Cs in the soil profile is different than for cultivated areas [16]. Furthermore, soil erosion processes could be different for undisturbed soils and for cultivated soils.

For undisturbed soil, the models to convert ^{137}Cs measurement data into the soil erosion value can be divided into two main groups: the profile distribution models and the diffusion models. Both groups of models are classified as theoretical models. An empirical relationship similar to the relationships for cultivated soils also exists, but those relationships have many limitations and thus soil erosion estimation might be incorrect [17]. The profile distribution models are based on the assumption that generally for undisturbed soil the cesium activity decreases with depth exponentially

$$A(x) \quad A_{\text{ref}}(1 \quad e^{x/h_0}) \qquad (2)$$

where:

$A(x)$ – amount of ^{137}Cs above x depth, [Bqm^{-2}],

x – depth, [kgm^{-2}], and

h_0 – factor describing the profile shape, [kgm^{-2}].

For the soil erosion place, according to Walling *et al.* [11], and Walling *et al.* [18], the relationship could be expressed by the equation

$$Y \quad \frac{10}{t \quad 1963} \ln \quad 1 \quad \frac{X}{100} \quad h_0 \qquad (3)$$

where:

Y – rate of soil erosion, [t.ha^{-1}yr^{-1}],

t – years of sample collection, and

X – reduction of the ^{137}Cs inventory at the sampling point relative to the local reference inventory of ^{137}Cs [%] (fig. 3)

In fig. 3(b), it is clearly visible that the rate of soil erosion has a maximum value on the depth of 20 to 70 cm. This is confirmed at two horizons, the first horizon located at the depth of 20 to 40 cm and the second horizon located at the depth of 40 to 70 cm.

In fig. 3(a), it is confirmed once again, same as in cultivated area measurements, that with increasing ^{137}Cs activity, the rate of soil erosion decreases.

For this model, it is also possible to assume, same as for simplified mass balance model described in the previous chapter, that the total ^{137}Cs fallout occurred in 1963. This simplified version of the model was described by Walling *et al.* [18]. It should be mentioned that such a simplified mass balance model could be a source of the overestimation of the soil erosion rate. The models described above, although quite easy to use, might not provide correct values of soil erosion estimation.

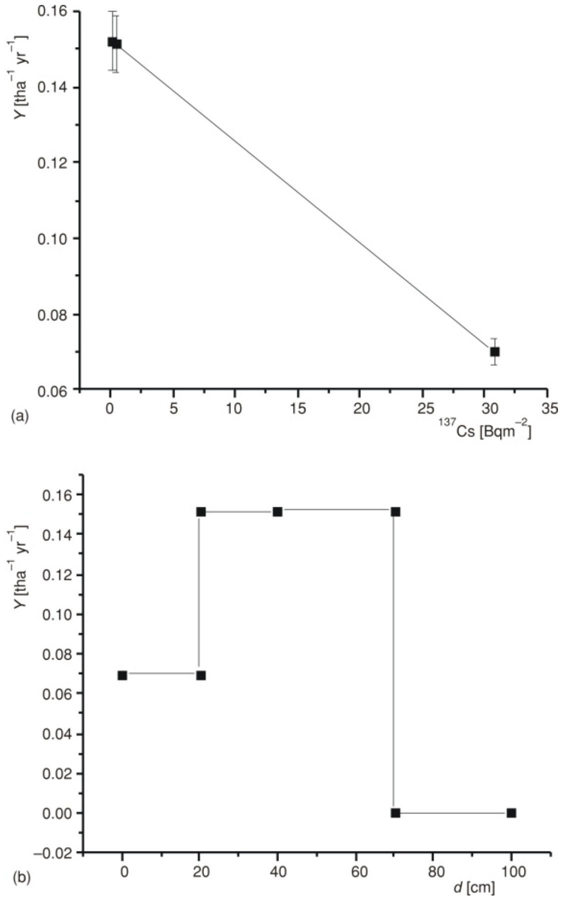

Figure 3. (a) Rate of soil erosion dependence of activity at undisturbed areas; (b) rate of soil erosion dependence of sampling depth at undisturbed areas

The results of soil erosion calculation depend on the distribution pattern of ^{137}Cs in the soil profile, as was shown by Yang *et al.* [19]. They performed a numerical simulation of the soil erosion rate by using different values of ^{137}Cs loss as well as the depth distribution of ^{137}Cs. They found that for a small percentage of the reduction in the total ^{137}Cs inventory, the relationship between soil loss and ^{137}Cs loss is almost linear. They concluded that the depth distribution patterns of ^{137}Cs are a major factor for estimating the rate of soil loss. To carry out more realistic studies, the diffusion models are better. The behaviour of cesium in soil after the deposition is quite complex and the depth distribution of ^{137}Cs in soil profiles is time-dependent. There are physical, chemical, and biological processes responsible for cesium redistribution within soil.

CONCLUSIONS

This study served as a preliminary investigation of the amount and variability of ^{137}Cs in the soil under two different land uses, cultivated and undisturbed soils. ^{137}C activity is substantially different at each 12 samples and reflects the erosion status of each site. Variability is not unusually high for a soil property, but the degree of variability found presents serious sampling problems because of the long counting times involved in ^{137}Cs analyses. Variability appears to be primarily dependent on the land use and sampling area.

The variability in ^{137}Cs activity in soil precludes the use of individual samples for soil loss estimates. The routine analysis of the large numbers of samples in erosion surveys is likely to be impractical. It is suggested that once an estimate of variability has been made, any further samples collected be bulked for analysis.

Caesium-137 method almost always gives good results, especially in defining erosion intensity in undisturbed areas. Otherwise, some errors could be present as a consequence of non-ideal distribution of radioisotopes, especially their concentration and distribution on dense wooden areas, cultivated areas, *etc*. From this reason, it is necessary that the obtained results should be compared with the results obtained using other methods and techniques. For example, this other method could be the well known Gavrilovic's and hydro-technical method.

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Снежана С. НЕНАДОВИЋ, Милош Т. НЕНАДОВИЋ, Ивана С. ВУКАНАЦ,
Александар Р. ЂОРЂЕВИЋ, Славољуб С. ДРАГИЋЕВИЋ, Милутин А. ЉЕШЕВИЋ

ВЕРТИКАЛНА РАСПРОСТРАЊЕНОСТ ЦЕЗИЈУМА–137 У ОБРАДИВИМ И НЕОБРАДИВИМ ЗЕМЉИШТИМА

У овом раду приказана је распрострањеност и јединствене карактеристике цезијума-137 како у обрадивим тако и у необрадивим земљиштима. Узорковање је вршено на три мерна места до 1 m дубине. Испитани су сви видљиви хоризонти и утврђено је постојање 4 хоризонта у једном профилу, који су различитих дебљина. Цезијум-137 је вештачки радиоизотоп који је настао као последица термонуклеарних реакција испитиваног оружја у атмосфери земље као и нежељени производ нуклеарних акцидената (Чернобил, 1986). Овај радиоизотоп је широко распрострањен у целом свету, где се његова миграција у земљишту прати помоћу интензитета ерозије потпомогнутим водом и ветром. Стога је цезијум-137 важан фактор при одређивању интензитета ерозије како у земљишту тако и у седиментима, те је одређена његова специфична активност коришћењем гама спектрометрије.

Кључне речи: цезијум-137, ерозија, земљиште, гама спектрометрија