

**Aleksandra TODOROVIK, Elizabeta DIMITRIESKA STOJKOVIK,
Risto UZUNOV, Zehra HAJRULAI-MUSLIU,
Biljana DIMZOVSKA, Slobodan BOGOEVSKI ¹**

¹³⁷Cs ANALYSIS IN CULTIVATED SOIL AND UNCULTIVATED SOIL SAMPLES FROM CITIES SKOPJE- MACEDONIA, USING GAMMA RAY SPECTROMETRY

SUMMARY

The goal of this study was focused on determination of the specific activity of ¹³⁷Cs in the samples of cultivated and uncultivated soil, as well as comparison with specific countries. The samples were taken from 14 locations in the surrounding of the city of Skopje. The spectral analysis of the radionuclides of these samples was conducted by applying a γ -ray spectrometer with high purity germanium (HPGe) detector with 30% relative efficiency.

The data show that the specific activities of this radionuclide vary within a range of 6,04 Bq/kg (measured in Dracevo at a depth of 15 cm) to a maximal value of 21,87 Bq/kg (measured in Radisani at a depth of 5 cm) for cultivated land. The specific activities of this radionuclide vary within a range of 5,01 Bq/kg (measured in Radisani at a depth of 10-15 cm) to a maximal value of 19,05 Bq/kg (measured in Bardovci at a depth of 0-5 cm) for non-cultivated land.

On the basis of the obtained results, an expressed variability is perceived regarding the specific activities of ¹³⁷Cs in the examined soil samples according to location and depth. The very research indicates that in both types of soil, the highest level of ¹³⁷Cs exists at a depth of 5 cm which is in accordance with the literature data. In the examined locations there is no need of special procedures for soil decontamination, however the familiarization with the methods related to the decrease of the radiological activity can be beneficial considering the fact that the soil is a part of the link soil-food-human.

Keywords: soil, gamma spectrometry, ¹³⁷Cs

INTRODUCTION

The radionuclides on the surface of the ground come in a form of solid particles or with the rains in dissolved or non-dissolved state. The ones that come in the form of solid particles, are mechanically retained on the surface, while the ones which are dissolved with the filtration process penetrate the soil and most of

¹ Aleksandra Todorovik, (corresponding author: mizasandra@fvm.ukim.edu.mk), Elizabeta Dimitrieska Stojkovik, Risto Uzunov, Zehra Hajrulai-Musliu, Biljana Dimzovska, University Ss. Cyril and Methodius, Faculty for Veterinary Medicine, Food Institute, Skopje, Macedonia, Slobodan Bogoevski, University Ss. Cyril and Methodius, Faculty for Veterinary Medicine, Institute for Technology and Metallurgy in Skopje, Macedonia

Notes: The authors declare that they have no conflicts of interest. Authorship Form signed online.

them attach to its surface layer. One can distinguish ^{90}Sr and ^{137}Cs from the isotopes that cause permanent radiological contamination of the pedosphere, which depending on the properties of the soil, they attach i.e. penetrate its layers (Bauman, 2004).

The presence of ^{137}Cs in the soil is very important and it is a clear indicator that the area under study may have received specific radioactivity from decay, however it is very difficult, even impossible to precisely determine the source of this contamination because there are no data about the supplies of radio-caesium in the environment of the selected area prior to this study. However, one may assume that this may be due to the Chernobyl disaster and also due to the slight deposition from various atmospheric tests of nuclear weapons in the neighboring countries. ^{137}Cs comes to the soil in a form which is soluble in water. Hence, after contamination, it has been determined that it is present on the surface layer with 85%, at a depth of 5-10 cm - 12%, and at a depth of 10-15 cm, it is 3%. The accumulation of cesium on the ground depends on many factors (the type of the soil), and numerous researches have shown that it stays mostly in the layer of 5cm from the surface of the ground, because its penetration speed through the ground is 1-3 cm g^{-1} . However, the speed will depend on the type of the land and the amount of atmospheric rains (Krstić *et al.* 2004). The transfer of ^{137}Cs in the ground is also affected by its form. If it is present as part of the anion, the soil will hardly absorb it, and for this reason it will intensively migrate in the plants. This migration ability is also affected by the potassium, as well as the stable cesium (their excess slows down the migration). According to specific data, it has been concluded that ^{137}Cs is mostly fixed permanently to the soil, whereby within the first year from its arrival, it retains on the surface of the ground, and the speed of its migration according to the depth is few cm per year, which also depends on the type and the properties of the soil (Mirkovic, 1987).

Something which is very important is that the specificity of ^{137}Cs is such so that in the organism it behaves same as potassium, which means that it is located in every cell of the organism i.e. it is being distributed in all organs evenly. It is also the cause for great radiological risks (NCRP, 19914).

The measurement of radioactivity in the soil provides information about the natural resources and thereby it is important for the measurement of the dose of radiation for the general population and observation of the radiation. The interaction of the ionizing radiation and the human body leads to various biological effects which can later be manifested as clinical symptoms. The nature and the seriousness of the symptoms depend on the absorbed dose, as well as on the dose rate and many diseases which should have been effectively managed if information about the level of radiation of an environment was available. The familiarity with the concentration of radioactivity in our environment is essentially important for the estimation of the dose which is being accumulated in the population and also for formation of the foundation for assessment of the level of radioactive contamination or environmental contamination in the future.

Taking into consideration the importance of the distribution and the transfer of ^{137}Cs to the soil, the goal of this work was to determine and compare the specific activities of ^{137}Cs in samples of cultivated soil at three different depths.

MATERIAL AND METHODS

In order to measure the specific activity of ^{137}Cs , soil samples were taken from 14 locations in the surrounding of the city of Skopje. Samples were taken from cultivated and uncultivated soil for each location in order to make their comparison. Each sample is taken by means of a special dosing container with limiters which enable taking of samples at a depth of 0-5cm, 5-10cm and 10-15cm, which enables sampling above these soil layers. The soil sampling was performed so that 3-4 samples have been taken from every location, for the indicated depths according to the recommendations by the IAEA (IAEA 295, 1989).

Attention was paid that the microlocation comprises of a flat terrain, which excludes the consequences for possible horizontal translocation of radionuclides. The collected samples were carefully cleaned from small stones and then they were dried in an electric oven at a temperature of 110°C to 48 hours depending on the depth of the soil until the sample obtains a constant weight. After drying, the samples were crushed, placed on a base and melted to a previously determined size of particles according to the analytical demands and then they passed through a sieve with a 200 μm mesh size. The homogenized samples of the soil were packed in plastic containers which had the same geometry as the one for the reference materials.

The spectral analysis of the radionuclides of these samples was conducted by applying a γ -ray spectrometer with high purity germanium (HPGe) detector with 30% relative efficiency and energy resolution (FWHM) of 1.8 keV for 1.33 MeV reference passage of ^{60}Co (Verdoya et al. 2009).

The detector was protected with 9cm-thick lead with an internal line with a 0,5cm-thin copper panel covered by 1mm aluminum in order to absorb the x-rays from the lead and the copper. The internal size of the cavity of the shell was 30 x 30 x 30 cm. The detector was given a high voltage through a preamplifier which was then connected to an amplifier with a computer based channel analyzer through an ADC (analogue to digital converter). The software used for obtaining the data is Canberra software package Genie-2000, including search of maximal value and modules for identification of nuclides. The system was regularly calibrated for energy and efficiency. The energy calibration was performed by obtaining a spectrum of approved calibration sources of known energies such as ^{60}Co , for $E_{\gamma}=1332.5$ and 1173 keV, and ^{137}Cs , for $E_{\gamma}=661.6$ keV. The gamma rays of interest were within a range of 50-3000 keV. The prepared Marinelli glasses (samples) were placed on a final detector at a distance of approximately 10 mm. Every sample was measured within a period of 65000s in order to get good statistics and the constant time was lower than 10%. The measurements with an empty Marinelli glass, in identical conditions were also conducted in

order to determine the basic recounts. Then they were deducted from the measured spectrums of every sample in order to obtain the net activities of the radionuclides.

RESULTS AND DISCUSSION

On the basis of the obtained results (Table 1), can perceive expressed variability of the specific activities of ^{137}Cs in the examined soil samples according to locations and depth.

Table 1. Mean values of the specific activities of ^{137}Cs taken from samples of cultivated and uncultivated soil at three different depths

Location	Sample Soil	Depth cm	Cultivated soil Specific activities (Bq/kg)	Uncultivated soil Specific activities (Bq/kg)
Petrovac	1	5	15.03±0.30	15.23±1.22
		10	14.92±1.55	11.92±1.20
		15	14.11±1.55	11.15±1.20
	2	5	17.65±1.53	18.77±1.20
		10	15.33±1.52	11.42±1.20
		15	14.20±1.55	10.10±1.22
Belimbegovo	1	5	13.22±1.42	10.98±1.50
		10	11.45±1.51	7.23±1.51
		15	11.16±1.45	6.01±1.48
	2	5	11.11±1.43	11.10±1.36
		10	13.86±1.38	9.63±1.18
		15	11.09±1.43	5.08±1.43
Aracinovo	1	5	17.78±1.30	17.60±1.37
		10	19.89±1.35	13.83±1.53
		15	16.46±1.28	8.27±1.28
	2	5	19.17±1.29	15.12±1.29
		10	18.52±1.30	12.44±1.43
		15	18.68±1.32	9.78±1.32
Radisani	1	5	21.87±0.40	16.22±0.52
		10	18.75±0.40	10.11±0.50
		15	17.52±0.40	7.56±0.50
	2	5	19.67±0.42	16.89±0.52
		10	18.58±0.40	9.65±0.52
		15	16.11±0.45	8.07±0.50
Cucer	1	5	11.01±1.80	14.20±1.75
		10	10.72±1.75	11.83±1.70
		15	12.36±1.80	6.27±1.70
	2	5	10.42±1.82	12.92±1.75
		10	11.44±1.80	9.44±1.75
		15	11.24±1.81	8.73±1.75
Vizbegovo	1	5	12.02±1.70	17.26±1.75
		10	11.19±1.70	15.22±1.75
		15	10.14±1.72	11.27±1.70
	2	5	13.76±1.75	18.92±1.75
		10	11.45±1.70	16.06±1.75
		15	10.04±1.70	9.71±1.75
Bardovci	1	5	16.23±1.50	17.26±1.75
		10	15.11±1.55	15.22±1.75
		15	14.56±1.50	11.27±1.70
	2	5	17.08±1.50	18.92±1.75
		10	16.72±1.55	16.06±1.75
		15	12.84±1.55	9.71±1.75

Saraj	1	5	15.22±0.90	13.56±1.40
		10	12.26±0.95	9.46±1.43
		15	13.11±0.90	7.14±1.45
	2	5	15.43±0.90	14.58±1.40
		10	13.01±0.92	10.66±1.45
		15	11.94±0.90	8.66±1.45
Nerezi	1	5	10.56±1.50	11.56±1.50
		10	10.29±1.50	8.21±1.55
		15	09.98±1.56	6.34±1.50
	2	5	11.88±1.54	10.28±1.50
		10	11.05±1.55	9.46±1.55
		15	10.77±1.50	6.00±1.50
Lisice	1	5	7.29±1.1	9.18±1.75
		10	7.12±1.1	6.12±1.75
		15	6.78±1.1	5.01±1.70
	2	5	8.49±1.1	7.69±1.70
		10	7.66±1.1	5.84±1.70
		15	7.18±1.1	5.58±1.75
Dracevo	1	5	8.27±1.50	7.67±1.50
		10	6.47±1.50	7.97±1.55
		15	6.04±1.50	5.34±1.50
	2	5	9.02±1.50	7.32±1.50
		10	7.76±1.50	8.66±1.55
		15	8.32±1.50	6.48±1.50
Pintija	1	5	11.27±1.50	11.87±1.45
		10	11.06±1.50	9.26±1.55
		15	10.44±1.50	6.84±1.50
	2	5	12.32±1.52	12.92±1.50
		10	11.75±1.50	8.06±1.55
		15	11.04±1.50	7.34±1.50
Batinci	1	5	12.22±0.50	11.87±1.45
		10	12.44±0.50	9.26±1.55
		15	10.56±0.50	6.84±1.50
	2	5	11.75±0.52	12.92±1.50
		10	10.99±0.52	8.06±1.55
		15	10.22±0.55	7.34±1.50
Volkovo	1	5	10.38±0.90	9.38±0.55
		10	9.77±0.90	7.77±0.50
		15	9.42±0.90	6.42±0.50
	2	5	11.89±0.90	10.89±0.55
		10	10.65±0.90	9.65±0.50
		15	9.80±0.90	7.80±0.50

The specific activities of this radionuclide vary within a range of 6,04 Bq/kg (measured in Dracevo at a depth of 15 cm) to a maximum value of 21,87 Bq/kg (measured in Radisani at a depth of 5 cm) for cultivated land. One can perceive higher mean values of this radionuclide which are measured at a depth of 10 cm. The specific activities of this radionuclide vary within a range of 5,01 Bq/kg (measured in Radisani at a depth of 10-15 cm) to a maximal value of 19,05 Bq/kg (measured in Bardovci at a depth of 0-5 cm) for cultivated land. Higher mean values of this radionuclide can be perceived, measured at a depth of 5-10 cm.

Different factors may be responsible for the unequal distribution of ¹³⁷Cs. This variation of the data is not very significant, taking into consideration the

great geographical variations that may result from the difference of the soil characteristics, the environmental and meteorological factors, particularly rain in a period of deposition which is very important because it facilitates the deposition. However, from the very research one can perceive that in both soil types, the highest level of ^{137}Cs is present at a depth of 5 cm which is in accordance with the data from the literature (Clouvas *et al.* 2001) In all examined areas it is observable that the level of ^{137}Cs in composted soil for all depths has approximate values, and this is a result of the mechanical mixing of the soil, which is not the case with a soil that has not been composted.

From the Figure 1 may be concluded that the mean value of the specific activities of ^{137}Cs for cultivated soil is 7,58 Bq/kg (measured in Dracevo) to the highest value of 18,75 Bq/kg (measured in Radisani).

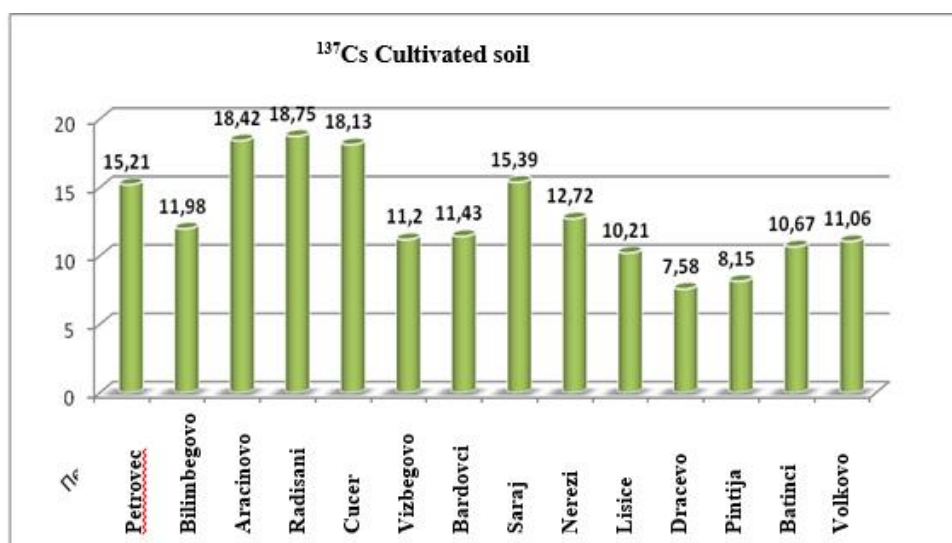


Figure 1. Mean values of the specific activities of ^{137}Cs for cultivated soil according to localities

Figure 2 shows that the mean value of the specific activities of ^{137}Cs for cultivated soil is 6,63 Bq/kg (measured in Dracevo) to the highest value of 14,94 Bq/kg (measured in Bardovci).

On the basis of the data from the literature, one can perceive that the mean values of the specific activities of ^{137}Cs are significantly higher in the areas which have been exposed to the radioactive cloud from Chernobyl (Bulgaria, Macedonia, Turkey, Republika Srpska and Serbia), (Karahana and Bayulken, 2000; Jankovic, 2001a; Janković, 2008b; Djingova and Kuleff, 2002), while they are significantly lower in Pakistan, Venezuela, China and these values are certainly a consequence of nuclear explosions and trials. Also one can perceive that the specific activities of ^{137}Cs are lower in sandy terrains (the Croatian islands and Saudi Arabia) (Kulic, 2008; Lu *et al.* 2006; La Brecque, 2005).

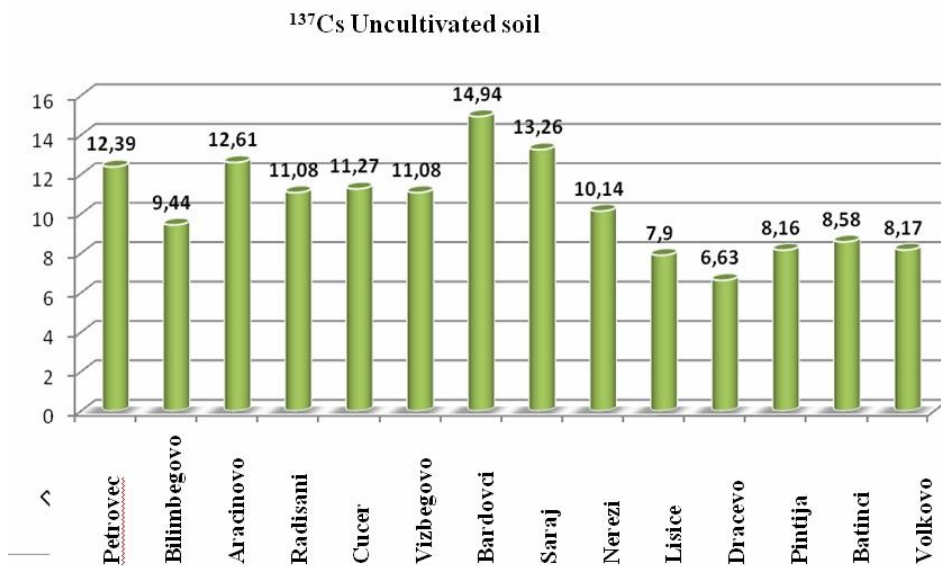


Figure 2. Mean values of the specific activities of ^{137}Cs from uncultivated soil according to localities

CONCLUSION

It may be determined that the specific activity of ^{137}Cs in uncultivated soil is significantly lower than measurements which are performed on cultivated soil. This is due to the application of different fertilizers which are applied to agricultural fields in recommendable amounts that may increase the radioactivity level in the soils (Akhtar and Tufail 2007).

The results of this study are useful as a basis with data, for the preparation of a radiological map of the studied area, as well as to enrich the world data bank.

REFERENCES

- Akhtar N, Tufail M. 2007. Natural Radioactivity Intake into Wheat Grown on Fertilized Farms in two Districts of Pakistan. *Radiation Protection Dosimetry*. 123; 103-112.
- Bauman A. 1965. The determination of ^{137}Cs in soil. *IRP*. 335-338.
- Clouvas A, Xanthos S, Antonopoulos-Domis M. 2001. Extended survey of indoor and outdoor terrestrial gamma radiation in Greek urban areas by in situ gamma spectrometry with a portable Ge detector. *Radiat Prot Dosimetry*. 94(3), 233-245.
- Djingova R, Kuleff I. 2002. Concentration of ^{137}Cs , ^{60}Co and ^{40}K in some wild and edible plants around the nuclear power plant in Bulgaria. *Journal of Environmental Radioactivity*. 59;61-73.
- IAEA 295. 1989. Measurement of radionuclides in food and the environment. A guide book, Technical Report 295.
- Jankovic, L. 2011. Distribucija prrodnih i procena radijacionog izlaganja stanovnistva stanovnistva regije Beograda. Belgrade University, Belgrade, Serbia. (Doctoral thesis).

- Janković M, Todorović D, Šavanović M. 2008. Radioactivity measurements in soil samples collected in the Republic of Srpska. *Radiation Measurements*. 43; 1448-1452.
- Karahan G, Bayulken A. 2000. Assessment of gamma dose rates around Istanbul. *Journal of Environmental Radioactivity*. 47; 213-221.
- Krstić D, Nikezić D, Šrećević N, Jelić M. 2004. Vertical profile of ¹³⁷Cs in soil. *Applied Radiation and Isotopes* 61; 1487-1492.
- Kulic O, Belivermis M, Tocuoglu S, Cotuk Y. 2008. Radioactivity concentrations and dose assessment in surface soil samples from east and south of Marmara region, Turkey. *Radiation Protection Dosimetry* 128; 324-330.
- La Brecque J, Cordoves J P R. 2005. Cesium 137 spatial activity in surface soils near and surrounding the Guri Reservoir (Venecuela). *Journal of Radioanalytical and Nuclear Chemistry*. 265; 91-94.
- Lu GJ, Huang YJ, Li F, Wang L, Li S, Hsia Y. 2006. The investigation of Cs-137 and Sr-90 background radiation levels in soil and plant around Tianwan NPP, China. *Journal of Environmental Radioactivity*. 90(2), 89-99.
- Mirković, G. 1988. Prilog ispitivanja uticaja meteoroloških faktora na akutnu radiokontaminaciju različitih zemljišta u BIH u toku 1986-1987. Veterinary Faculty, Zagreb, Croatia. (MA thesis).
- NCRP (National Council on Radiation Protection and Measurements). 1991. Report No 110,49.
- Verdoya M, Chiozzi P, De Felice P, Pasquale V, Bochiolo M, Genovesi I. 2009. Natural gamma-ray spectrometry as a tool for radiation dose and radon hazard modelling. *App. Rad. Isot.* 67(5), 964-968