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Chapter

Radioactive Isotopes in Soils and Their Impact on Plant Growth

Jelena Markovic and Svetlana Stevovic

Abstract

In 1999, Serbia was bombarded by NATO. One of the cities most affected by the consequences of bombardment with uranium is the city of Vranje, where the consequences are felt even today. Due to the influence of uranium, the mortality rate has increased. This paper presents the effects of some of the radionuclides that have contaminated the soil, as well as the connection between soil and plants that grow on that soil. The performed measurements of radionuclides (^{226}Ra, ^{40}K, ^{232}Th, ^{238}U, and ^{235}U). The results show that the content of each of these radionuclides has different concentrations, but what is important is that some values are even below the detection limit, corn <0.06 ^{235}U on the location Korbevac and wheat <0.04 ^{235}U on the location Bujkovac. On the three and all of these gated locations, the calculated values of the transfer factors for ^{40}K were in the range of 0.144–0.392, while in the case of ^{226}Ra, the transfer factors ranged from 0.008 to 0.074. Only one value (0.051) was obtained for the transfer factor of ^{232}Th. Specific activities of ^{137}Cs, as well as uranium isotopes, in all the investigated cereal samples, were below minimal detectable activity concentrations. The ratio of radionuclides in soil and plants is of great importance for human nutrition.

Keywords: soil, plants, radioactive isotopes, monitoring, mortality

1. Introduction

Natural radioactivity in the environment, originating from the naturally occurring radionuclides of ^{232}Th, ^{238}U, and ^{235}U radioactive series and ^{40}K, largely contributes to the natural irradiation of man and biota, which can be external and/or internal (ingestion and inhalation). Natural radionuclides land characteristic of α and β radioactive decay [1]. The biggest number of radionuclides belongs to a radioactive series, which naturally has three. These three series start as radioisotope, so-called parent: ^{238}U (series of 4n + 2), ^{235}U (series of 4n + 3), and ^{232}Th (string 4n). A series of successive radioactive decays occur from parents whose offspring core is also unstable and is subject to decay. The process of disintegration ends stable isotopes, and for those strings to the ^{206}Pb, ^{207}Pb, and ^{208}Pb, respectively, from the radionuclide which does not belong to any of the radioactive series, the most important is the soil radionuclide ^{40}K. Gamma radiation created during the radioactive decay of uranium and thorium series, as well as ^{40}K, largely contributes to the natural irradiation of alive council (man and biota), which can be external and/or internal (ingestion and inhalation) [2]. The concentration of natural radionuclides depends on the composition of the soil. According to the

report of the UNSCEAR, the medium activity concentration of ^{238}U, ^{232}Th, and ^{40}K in the soil in the world amounts 33.45 and 412 mCi kg-1, respectively. The ranges of concentrations of ^{238}U, ^{232}Th, and ^{40}K in the soil in Europe are 2–330, 2–190, and 40–1650 Bq kg^{-1} [3].

Besides the natural radionuclides, due to various human activities, different manmade radionuclides entered the environment. The most significant among them is ^{137}Cs ($T_{1/2}$ = 30y), found in the environment mostly as a result of the nuclear tests in the 1960s ties and the Chernobyl nuclear plant accident in 1986 [4]. ^{137}Cs is bound in the surface layers of soil and is washed out and redistributed in the ecosystem for a longer period of time due to its long half-life. Thus, only a small amount of it is present in plants today. It is well-known that ^{137}Cs isotopes take important part in the environment, due to their good assimilation by plants, which are used to feed the animals and finally human beings [5]. The reported values of ^{137}Cs in the agricultural soil in the north part of Serbia, on several locations near the city of Novi Sad, are in the range of 1.5–12.6 Bq/kg [6].

Soil is a complex material composed of mineral (inorganic) as well as an organic matter that originated from plant decomposition. It is a compact matter providing necessary micro- and macro-nutrition elements for plants to function and grow. Cereals as wheat, corn and barley are important component of everyday human diet [21]. Most of the radionuclide cereals are absorbed from soil, so the values of transfer factors are important in the studies of the transport and distribution of radionuclides in the "soil–plant–animal-human" chain, as well as in the evaluation of the radiation risk [7]. Transfer factors (TF) are crucial in the radionuclide transport models, in the environment as well as in the evaluation of the level of the specific activities of radionuclides in agricultural crops [8]. The main factors determining the level of TF are radionuclide itself, type of plant, type (physical and chemical characteristics) of soil, concentrations of stable chemical elements in soils [9], as well the local climate [10].The values of transfer factors should provide the basis for theoretical analysis on the different uptakes of elements not involved in physiological and biochemical processes in plants [11].

The main goal of this paper was to investigate transfer factor, because it can give crucial information about the possible quantity of radioactive and other



Figure 1.
Pcinja District.

toxic materials endangered for human's health. Starting from the soil, through the plants, they enter in the food chain and consequently they reach in the human body and affect mortality. The transport processes in the "soil–plant" systems for radionuclides (^{226}Ra,^{40}K, ^{232}Th, ^{238}U, ^{235}U) and ^{137}Cs in the Pcinja District. Pcinja District is located in the southern part of the Republic of Serbia. It covers the city of Vranje and the municipalities Vladicin Han, Surdulica, Bosilegrad, Trgoviste, Bujanovac, and Presevo (**Figure 1**). The Pcinja District has not been investigated yet.

2. Methodology

The samples of soils and cereals were collected in 2014, in the area of the city of Vranje, on three locations: the villages of Bujkovac, Korbevac, and Suvi Dol. The type of soils was the same on all the locations (the so-called *gajnjaca*). *Gajnjaca* belongs to well-drained soils, its chemical characteristics depending on the level of utilization, degree of erosion, chemical characteristics of the main substrate, and level of development. The content of humus in the gajnjaca soils is in the range of 2–5%. This type of soil in neutral or low acetous has a high capacity of adsorption, and the dominant ions in it are Ca and Mg. Its color is brown, reddish, or red depending of the content of aluminum and iron. It is very suitable for farming, wine growing, and afforestation.

2.1 Sampling sites and sample collection

About 11 samples of cultivated and uncultivated soils and 7 samples of cereals were collected. Sampling sites coordinates are present in **Table 1**. The samples of soils were taken from different depths that also differ from one to another sampling site. The depths that the soils were sampled from were 0–5 cm, 0–10 cm, and 0–20 cm on the sites Korbevac and Bujkovac and 0–5 cm, 5–10 cm, and 10–15 cm at the sampling site Suvi Dol. The samples of grain were taken from plots where the soil samples were taken. First, soil samples were taken for testing, on the land-sown cereals that are taken when they are ripe for examination. The position of the sampling sites is presented in **Figure 2**.

The radioactivity of the samples of soils was determined by gamma spectrometry in the Institute for Nuclear Sciences "Vinca" in the Laboratory for Radiation and Environmental Protection.

The mass of cereal and soil samples for analysis is necessarily 1 kg. The samples of soils were cleaned of mechanical impurities, stones and plant material, and dried at 105°C for 24 h. Samples of cereals were dried at room temperature and mineralized at 450°C. Soils were measured in Marinelli geometry (volume 500 ml) and cereals in cylinder bottles (volume 125 ml). The radioactive equilibrium was

Site	Coordinates		Elevation (m)	Sampling date
	(North latitude)	(East longitude)		
Bujkovac	42°33′26"	22°00′35"	718	09.11.2014.
Korbevac	42°23′06"	21°44′24"	441	05.11.2014.
Suvi Dol	42°33′07"	21°56′05"	359	11.11.2014.

Table 1.
Coordinates of the location of soil samples.



Figure 2.
Sampling sites on the territory of the city of Vranje.

achieved in all the samples, as they have been sealed by bee wax and left for 30 days before measuring. The samples of grain were taken at the stage of full maturity of the technology and to hand. Samples of cereals (fruit cereal) were dried in air at room temperature for at least 3 weeks and then were crushed and mineralized at a temperature of 450°C for 24 hours, dry-ashing method [12].

The specific activity of natural ^{226}Ra was determined by analyzing the spectra of its daughters [13], ^{214}Pb and ^{214}Bi, at the energies of 295, 352, 609, 1120, and 1764 keV [14]. Radionuclide ^{232}Th was determined by its daughter ^{228}Ac at the energies of 338 and 911 keV. The activities of ^{40}K and ^{137}Cs were determined at the energies of 1460 and 661.6 keV, respectively. The activity of ^{235}U and ^{238}U is determined by establishing a radiochemical equilibrium between ^{226}Ra and ^{214}Bi using photo-peak at energies around 186 keV-a [15, 16].

2.2 Standard gamma spectrometry

The gamma spectrometry was performed on three HPGe detectors (CANBERRA) with relative efficiencies of 18, 20, and 50%; the resolution of all of the detectors was 1.8 keV at 1332 keV. For the samples of soils, the detectors were calibrated by a reference radioactive material—a silicone resin matrix, Czech Metrological Institute, Praha, 9031-OL-420/12, total activity 41.48 kBq on 31.08.2012 (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{203}Hg, ^{88}Y, ^{113}Sn, ^{85}Sr ^{137}Cs). The gamma-spectrometric measurements of radioactivity in soil samples was used and the ultra-low-background germanium detector-type GMX (extended energy range from 10 keV to 3 MeV-a manufacturer of ORTEC, the nominal efficiency of 32% in passive and active protection). Passive safety lead is made up of a thickness of 12 cm in the form of a cylinder and coated with a layer of tin and copper. The active protection (veto detectors) is the five plastic scintillation detectors which are anticoincidence mode working with HPGe detector and completely cover passive protection. Active protection lowers integral countdown in the background of a factor of three for the range from 50 to 2800 keV, which lowers the threshold of detection and is suitable for the measurement of environmental samples [17]. For cereal samples the detectors were calibrated with a secondary reference radioactive material in plastic boxes (volume 125 cm^3) obtained from the primary reference radioactive material—Czech Metrological Institute, Praha, 9031-OL-427/12, type ERX, total activity 72.40 kBq on 31.08.2012 (^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{203}Hg, ^{88}Y, ^{113}Sn, ^{85}Sr ^{137}Cs, ^{210}Pb) [19].

The counting time was 60,000 s. The results are presented with the expanded measuring uncertainty for the factor k = 2, level of confidence for normal distribution 95%.

2.3 Transfer factor calculations

Transfer factor (TF) was calculated according to Eq. (1), defined as the ratio of specific activity of radionuclide in plant (Bq/kg dry matter) and specific activity in soil (Bq/kg) [18]:

$$TF = \frac{A_p}{A_s} \quad (1)$$

where A_p is the specific activity of the radionuclide in plant [Bq/kg dry matter], and A_s is the specific activity of the radionuclide in soil [Bq/kg].

The change absorbed dose intensity into absorbed dose rate of gamma radiation from the natural radionuclides in soil was calculated according to Eq. (2).
The annual effective dose was calculated according to Eq. (3).

$$D(nGyh - 1) = 0.462 \times CRa + 0.604 \times CTh + 0.0417 \times CK \quad (2)$$

where CRa is the specific activity of ^{226}Ra in soil, CTh is the specific activity of ^{232}Th in soil, and CK is the specific activity of ^{40}K in soil.

$$DE(mS\nu) = 0.7S\nu Gy - 1 \times 0.2 \times 365 \times 24 \times D \quad (3)$$

3. Results and discussion

The results of the gamma spectrometry analysis of soils at different locations (sampling sites) are presented in **Table 2**.

The results of the calculated absorbed dose intensity and the annual effective doses from natural radionuclides in soils are presented in the table and in **Table 3.**

There are no significant differences among the specific activities of natural radionuclides in soils regarding the sampling depth of the soil at the specific location, i.e., the differences are within the measuring uncertainty. The same applies for the specific activities of ^{137}Cs—their values do not differ significantly regarding the sampling depth of the soil at the specific location. As it has been detected only in traces, it does not present a risk of being accumulated in plants and human diet [20].

For all of the locations, the specific activities of ^{226}Ra are in the range of 22–45 Bq/kg, while for ^{232}Th the values are in the range of 29–55 Bq/kg. For ^{40}K, the specific activities cover the interval from 460 to 730 Bq/kg, for ^{238}U the activities are in the range of 22–51 Bq/kg, and for ^{235}U in the range of 1.1–2.7 Bq/kg. The specific activities of ^{137}Cs cover the interval of 7.2–17 Bq/kg. The uneven distribution of cesium within the same area is mainly due to the relocation and washing out effects in the soil.

There are no significant differences among the specific activities of natural radionuclides between the locations (**Table 2**). The lowest values of the specific activities for ^{226}Ra, ^{232}Th, ^{238}U, and ^{235}U are obtained at Bujkovac, the highest ones at Korbevac. The values are within the range of the literature data of the specific

	Bq/kg					
Depth (cm)	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{238}U**	**^{235}U**	**^{137}Cs**
			Korbevac			
0–5	43 ± 3	55 ± 4	730 ± 50	47 ± 8	2.7 ± 0.2	16 ± 1
0–10	45 ± 3	54 ± 4	730 ± 50	51 ± 9	2.4 ± 0.2	16 ± 1
0–20	38 ± 3	51 ± 4	690 ± 40	40 ± 8	2.4 ± 0.2	15 ± 1
			Suvi Dol			
0–5	38 ± 3	52 ± 4	490 ± 30	35 ± 8	1.7 ± 0.1	10.1 ± 0.7
5–10	33 ± 2	48 ± 3	470 ± 30	34 ± 9	1.7 ± 0.2	7.9 ± 0.6
10–15	37 ± 3	50 ± 3	460 ± 30	34 ± 8	1.9 ± 0.2	7.2 ± 0.5
			Bujkovac			
0–5	22 ± 2	30 ± 2	500 ± 30	25 ± 8	1.6 ± 0.2	17 ± 1
0–10	23 ± 2	30 ± 2	510 ± 30	25 ± 7	1.5 ± 0.1	18 ± 1
0–20	25 ± 2	29 ± 2	520 ± 30	22 ± 8	1.1 ± 0.1	17 ± 1

Table 2.
Specific activity of radionuclides in soil samples at different depths and sampling sites [Bq/kg].

Depth (cm)	**D(nGyh^{-1})**	**D_E(mS$_v$)**
Korbevac		
0–5	83.53	0.102
0–10	85.85	0.105
0–20	73.89	0.091
Suvi Dol		
0–5	69.39	0.085
5–10	63.84	0.078
10–15	66.48	0.081
Bujkovac		
0–5	49.13	0.061
0–10	50.75	0.062
0–20	50.01	0.061

Table 3.
Absorbed dose intensity D(nGyh^{-1}) and the annual effective doses D_E(mS$_v$) from natural radionuclides in soils.

activities of natural radionuclides in soils reported for the region of former Yugoslavia [6]. Compared to the other locations, the specific activity of ^{226}Ra is lower only in soils sampled at Bujkovac.

The values of the calculated absorbed dose intensity are in the range of 49.13–85.85 nGy/h, while the annual effective doses range from 0.061 to 0.105 mSv/h and are within the values reported for other regions in the country [6].

The results of the levels of natural radionuclides and ^{137}Cs in cereals are presented in **Table 4**.

Table 5 presents the means of the specific activities of the radionuclides in cereals sampled on the investigated locations.

The specific activity of ^{232}Th (2.6 Bq/kg dry matter) presented in **Table 5** refers only to the sample of wheat from the village of Korbevac. In all the other samples of cereals, the specific activity of this radionuclide is under MDA. The specific activities of ^{238}U, ^{235}U, and ^{137}Cs in all investigated samples of cereals are under the MDA, too.

The values of calculated transfer factors are presented in **Table 6**. The values of the specific activity in soil used to calculate the transfer factors were the mean specific activity of the radionuclides for the different sampling depth at the location.

As some of the obtained values of the radionuclides, specific activities in cereals were under MDA; transfer factors were calculated only for ^{40}K, ^{226}Ra, and ^{232}Th. The calculated values of the transfer factors for cereals indicate that ^{40}K and ^{226}Ra are the main radionuclides transferred into the cereal grain. The TF for ^{40}K (0.144–0.392) are higher than the TF for ^{226}Ra and ^{232}Th by an order of magnitude (0.00–80.074 for TF (^{226}Ra)). The TF for ^{40}K can be rather high, as is known and reported in the literature [4]. Other radionuclides do not accumulate

	(Bq/kg)					
Sample	**^{226}Ra**	**^{232}Th**	**^{40}K**	**^{238}U**	**^{235}U**	**^{137}Cs**
Korbevac						
Wheat	2.2 ± 0.4	2.6 ± 0.8	150 ± 10	< 2	< 0.2	< 0.06
Corn	0.4 ± 0.1	< 0.2	108 ± 7	< 1	< 0.06	< 0.03
Suvi Dol						
Wheat	0.30 ± 0.07	< 0.1	106 ± 7	< 0.6	< 0.04	< 0.02
Corn	< 0.2	< 0.2	68 ± 5	<1	< 0.09	< 0.03
Bujkovac						
Wheat	0.37 ± 0.07	< 0.2	102 ± 7	< 1	< 0.04	< 0.02
Corn	1.4 ± 0.3	< 0.4	89 ± 7	< 2	< 0.1	< 0.06
Barley	1.7 ± 0.2	< 0.2	200 ± 10	< 2	< 0.1	< 0.07

Table 4.
Specific activity of radionuclides in grain samples [Bq/kg dry matter].

Radionuclide	**Cereals (Bq/kg)**		
	Mean value	**Interval**	
		Min	Max
^{226}Ra	1.06	MDA*	2.2
^{232}Th	2.6	MDA	2.6
^{40}K	118	68	200
^{238}U	The values are under MDA		
^{235}U			
^{137}Cs			

**MDA—minimal detection limit.*

Table 5.
Mean values of the radionuclides' specific activities in cereals [Bq/kg dry matter].

	Transfer factor		
Cereal	$^{226}Ra_p/^{226}Ra_s$	$^{232}Th_p/^{232}Th_s$	$^{40}K_p/^{40}K_s$
	Korbevac		
Wheat	0.052	0.051	0.209
Corn	0.009	—	0.151
	Suvi Dol		
Wheat	0.008		0.224
Corn	—	—	0.144
	Bujkovac		
Wheat	0.016	—	0.200
Corn	0.061	—	0.174
Barley	0.074	—	0.392

Table 6.
Value of transfer factor for cereals.

in the plant in more significant amounts [12]. This is mostly due to the discrimination in uptake of essential and nonessential elements, exhibited by the plant [12]. Also, it is reported that small percentage of the total activity found in the plant is accumulated in the root system, while 1–16% is accumulated in the grain [15]. The addition of phosphate to soil reduces the availability of thorium for root uptake through the formation of phosphate salts that have low solubility [15]. Regression analysis, reported in [15], showed that thorium availability to wheat was negatively related to soil pH and positively related to soil organic matter, cationic exchange capacity, and clay content. In comparison to the literature, it can be seen that the obtained TF for cereals in Pcinja region are in agreement with the results obtained in other parts of the world [7, 12, 16], while they are lower by the order of magnitude in comparison to the TF reported for the plants that are principally grass pasture, where the stem and leaves were analyzed (TF(Ra) = 0.17, TF(Th) = 0.058, TF(K) = 1.3 [17]).

4. Conclusion and recommendation

The specific activities of the radionuclides in soil, at all the investigated locations, were in the range from 22 to 45 Bq/kg for ^{226}Ra, from 29 to 55 Bq/kg for ^{232}Th, 460 to 730 Bq/kg for ^{40}K, from 22 to 51 Bq/kg for ^{238}U, from 1.1 to 2.7 Bq/kg for ^{235}U, and from 7.2 to 17 Bq/kg for ^{137}Cs. The obtained specific activities for ^{236}Ra, ^{232}Th, and ^{40}K in "gajnjaca" soil are in good agreement with the values obtained for the other types of soils [4]. The differences between the specific activities of a radionuclide in soil samples from different depths are within the measuring uncertainties, and the ratio of specific activities for ^{235}U/^{238}U suggests the natural origin of uranium. The activities of radionuclides in cereals also do not differ from the values obtained by other authors. Distribution of radionuclides from the soil into the plant depends on the bioavailability of minerals in the soil, the root structure of the investigated plant and the processes in the plant tissue. The calculated values of TF for cereals indicate that ^{40}K and ^{226}Ra are the main radionuclides that are transferred in cereals. This evaluation is most important for the production of foodstuffs diet with low contents of radionuclides.

On the location of Korbevac, Suvi Dol, and Bujkovac the calculated values of TF for ^{40}K were in the range of 0.144–0.392; for ^{226}Ra the values of transfer factors were in the range of 0.008–0.074. It should be noted that the evaluated activities refer to the content of radionuclides in dry plant matter and that the activities in the fresh plants are on the average four to five times lower due to the water content. For other natural radionuclides and for ^{137}Cs, the TF have not been calculated as the specific activities of these radionuclides in cereals were under the MDA. The results presented in this paper are the preliminary investigations of the contents of radionuclides in soils and cereals in the region of Pcinja. As the transfer factors in the "soil-cereal" system were determined only for the specific type of soil, the investigations should continue for other types of soils and cereals mostly used in animal and human diet. The measurements presented in this manuscript are the first to be conducted in the region of Pcinja, thus providing the results that can be used as a baseline for the future measurements and monitoring.

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