TRANSFER FACTORS FOR THE „SOIL-CEREALS" SYSTEM IN THE REGION OF PCINJA, SERBIA

by

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Scientific paper
DOI: 10.2298/NTRP1604376M

The aim of the paper was to estimate the values of transfer factors for natural radionuclides (^{40}K, ^{226}Ra, ^{232}Th, ^{235}U, and ^{238}U) and ^{137}Cs from soil to plants (cereals: wheat, corn and barley) as important parameters for the agricultures in the selection of the location and the sort of cereals to be planted on. The results presented in this paper refer to the „soil-cereals" system in the region of Pcinja, Serbia. Total of 9 samples of soil and 7 samples of cereals were measured in the Department of Radiation and Environmental Protection, Vinca Institute of Nuclear Sciences, using three high-purity germanium detectors for gamma spectrometry measurements. In all the samples, transfer factors for ^{226}Ra are significantly lower than for ^{40}K, but they are all in good agreement with the literature data. On the three investigated locations, the calculated values of transfer factors for ^{40}K were in the range of 0.144 to 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 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. Also, the absorbed dose rate and the annual absorbed dose from the natural radionuclides in the soil, were calculated. The absorbed dose rate ranged from 49-86 nSv/h, while the annual absorbed dose ranged from 0.061-0.105 mSv. 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 future measurements and monitoring.

Key words: transfer factor, natural radionuclide, ^{137}Cs, soil, cereal, environment

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) [1, 2]. Natural radionuclide concentrations in soil strongly depend on the type of the parent rock in soil genesis. The concentrations of uranium and thorium are significant in igneous rocks (granite) and also in some sedimentary rocks (shale and phosphate rock). According to the reports, the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K, in the soil in Europe, ranges from 2-330 Bq/kg, 2-190 Bq/kg and 40-1650 Bq/kg, respectively [1].

Besides the natural radionuclides, due to various human activities, different artificial radionuclides entered the environment. The most significant among them is ^{137}Cs ($T_{1/2}$ = 30 year), found in the environment mostly as a result of the nuclear tests in the 1960-ties and the Chernobyl nuclear plant accident in 1986. The ^{137}Cs is bound in the surface layers of soil and is washed out and redistributed in the ecosystem, for a prolonged period of time, due to its long half-life. The reported values of ^{137}Cs in 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 [1, 3].

Soil-plant-animal-human chain is recognized as one of the major pathways for transfer of radionuclides to humans. Radionuclides may be transferred to plants along with the nutrients, accumulate in various parts of the plant and finally reach the edible portions. These plants or their parts, when consumed by men or animals, lead to continuous radiation exposure [4].

Cereals such as wheat, corn and barley are an important component of everyday human diet. Most of the radionuclides absorbed by the cereals originate

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from soil, so the values of transfer factors are important in the studies of transport and distribution of radionuclides in the "soil-plant-animal-human" chain, as well in evaluation of the radiation risk [5]. Transfer factors (TF) are crucial in the radionuclides transport models in the environment as well as in evaluation of the level of the specific activities of radionuclides in agricultural crops [4]. The main factors that determine the value of TF are the radionuclide itself, type of plant, physical and chemical characteristics of soil, concentrations of stable chemical elements in soils, as well as the local climate [6].The values of transfer factors should provide the basis for theoretical analysis on the different uptakes of elements both involved and not involved in physiological and biochemical processes in plants [7]. The investigations of TF conducted up to date in Serbia, suggest the values of TF ranging from 0.105 to 1.100 for ^{226}Ra, from 0.021 to 1.304 for ^{323}Th, from 0.447 to 3.776 for ^{40}K, and from 0.011 to 0.307 for ^{137}Cs [8, 9].

Transport processes in the "soil-plant" systems for radionuclides ^{226}Ra, ^{40}K, ^{232}Th, ^{238}U, ^{235}U, and ^{137}Cs, in the region of Pcinja, have not been investigated previously. Since the phosphate fertilizers are used in the agriculture in this region and bearing in mind that heavy bombardment took place in the nearby region, the need arose for this type of measurement that can assess the impact on the food chain. Thus, this paper presents the preliminary results for the transfer factors for cereals mostly used in human diet in the region.

SAMPLING AND METHODS

Sampling and sample preparation

The samples of soils and cereals were collected during 2014, in the area of the city of Vranje, on three locations: the villages of Bujkovac, Korbevac, and Suvi Dol. The type of soil was the same in all the locations (so called *gajnjaca*). This type of soil 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 soil is in the range of 2 to 5 %. This type of soil is neutral or low acetous, has a high capacity for 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 forestation.

The total of 9 samples of the cultivated and uncultivated soil samples, with a mass of about 1 kg, and 7 samples of cereals' grains, with a mass of about 0.5 kg, were collected. Sampling sites co-ordinates are presented in tab. 1 and the position of the sampling sites is presented in fig.1. The soil samples were taken at different depths that also differ from site to site. The depths that the soil samples were taken from were: 0-5 cm, 0-10 cm, and 0-20 cm in Korbevac and Bujkovac; 0-5 cm and 5-10 cm and 10-15 cm at the sampling site Suvi Dol.

Table 1. Sampling sites and co-ordinates

Site	Co-ordinates		Elevation [m]	Soil sampling date
	North latitude	East longitude		
Korbevac	42°23'06"	21°44'24"	441	05. 11. 2014.
Suvi Dol	42°33'07"	21°56'05"	359	11.11.2014.
Bujkovac	42°33'26"	22°00'35"	718	09.11.2014.

The samples of soils were cleaned from mechanical impurities, stones, and plant material, sieved and dried at 105 °C for 24 hours. After drying, the samples were put in Marinelli beakers, sealed with bee wax and stored for 30 days to achieve radioactive equilibrium.

Samples of cereals consisted of ripe grain, collected in harvest season 2014, from the respective parcels from which the soil samples were collected in November 2014. Therefore, each cereal sample corresponds to one soil sample. No other part of the plant was used for measurement, since the grain is the only part of measured cereals that is used in human diet. The grain was dried at room temperature and mineralized at 450 °C. The ash obtained by mineralization was placed in cylindrical geometry (volume 125 ml), sealed with bee wax and stored for 30 days until the radioactive equilibrium is achieved.

Standard gamma spectrometry

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

The gamma spectrometry was performed on three high purity germanium (HPGe) detectors (CANBERRA) with relative efficiencies of 18 % (*n*-type), 20 % (*p*-type), and 50 % (*n*-type). Resolution of all of the detectors was 1.8 keV at 1332 keV. Efficiency calibration for soil samples was performed using 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). For cereal samples the detectors were calibrated with a secondary reference radioactive material in cylindrical geometry (volume 125 cm^3) produced 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) [10].

The counting time was 60000 s. The results are presented with the expanded measurement uncertainty for the factor $k = 2$, with the level of confidence of 95 % for normal distribution [11].

Figure 1. Sampling sites in the region of Pcinja

Table 2. The energies used to determine the specific activity of the investigated radionuclides

Radionuclide	Daughters	Energy [keV]
^{226}Ra	^{214}Pb	295, 352
	^{214}Bi	609, 1120, 1764
^{232}Th	^{228}Ac	338, 911
^{40}K	–	1460
^{137}Cs	–	661.7
^{238}U	^{234}Th	63
^{235}U	–	186

Calculations

The TF was calculated according to eq. (1), defined as the ratio of specific activity of radionuclide in plant dry matter [Bqkg^{-1}] and specific activity in soil [Bqkg^{-1}]

$$TF \quad \frac{A_p}{A_s} \tag{1}$$

where A_p is the specific activity of the radionuclide in plant dry matter [Bqkg^{-1}] and A_s – the specific activity of the radionuclide in soil [Bqkg^{-1}].

The absorbed dose rate of gamma radiation from the natural radionuclides in soil was calculated according to the [2]

$$\dot{D}[\mathrm{nGyh}^{-1}] \quad 0.462\ C_{\mathrm{Ra}} \quad 0.604\ C_{\mathrm{Th}} \quad 0.0417\ C_{\mathrm{K}} \tag{2}$$

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

The annual effective dose was calculated according to [2]

$$D_{\mathrm{E}}\,[\mathrm{mSv}] \quad 0.7\,\mathrm{SvGy}^{\ 1} \quad 0.2 \ \ 365 \ \ 24 \ \ \dot{D} \tag{3}$$

RESULTS AND DISCUSSION

The results of gamma spectrometry analysis of the soil samples at different sampling sites are presented in tab. 3. The results of the calculated absorbed dose rate and the annual effective doses from natural radionuclides in soils are presented in the tab. 4

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. The uneven distribution of cesium between different sampling locations can be due to the relocation and washing out effects in the soil. The ratio of the ^{235}U/^{238}U specific activities is used as an indication of the origin of these radionuclides and the eventual presence of contamination. This ratio in

Table 3. Specific activity of natural radionuclides and ^{137}Cs in the soil

Depth [cm]	Specific activity [Bqkg^{-1}]						
	^{226}Ra	^{232}Th	^{40}K	^{238}U	^{235}U	^{137}Cs	^{235}U/^{238}U
Korbevac							
0-5	43 3	55 4	730 50	47 8	2.7 0.2	16 1	0.057
0-10	45 3	54 4	730 50	51 9	2.4 0.2	16 1	0.047
0-20	38 3	51 4	690 40	40 8	2.4 0.2	15 1	0.06
Suvi Dol							
0-5	38 3	52 4	490 30	35 8	1.7 0.1	10.1 0.7	0.048
5-10	33 2	48 3	470 30	34 9	1.7 0.2	7.9 0.6	0.051
10-15	37 3	50 3	460 30	34 8	1.9 0.2	7.2 0.5	0.056
Bujkovac							
0-5	22 2	30 2	500 30	25 8	1.6 0.2	17 1	0.064
0-10	23 2	30 2	510 30	25 7	1.5 0.1	18 1	0.061
0-20	25 2	29 2	520 30	22 8	1.1 0.1	17 1	0.052

Table 4. Absorbed dose rate $\dot{D}$ and the annual effective dose DE from natural radionuclides in soil

Depth [cm]	$\dot{D}$ [nGyh^{-1}]	D_E [mSv]
	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 5. Specific activity of natural radionuclides and ^{137}Cs in cereals

Sample	Specific activity [Bqkg^{-1} dry matter]					
	^{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

investigated samples is close to natural ratio of 0.046 [12].

The values of the specific activities for ^{226}Ra, ^{232}Th, ^{238}U, and ^{235}U are within the range of the literature data for the specific activities of natural radionuclides in soils, reported for the region of ex-Yugoslavia [1] and for the region of Southern Serbia [9].

The specific activities of ^{137}Cs cover the interval from 7.2 to 17 Bq/kg, which is lower than previously reported values for Serbia [3, 9] and in the order of magnitude with the values reported in the other parts of the world [2].

The values of the calculated absorbed dose rates are in the range from 49.13 to 85.85 nGy/h, while the annual effective doses range from 0.061 to 0.105 mSv and are within the values reported for other regions in the country [13, 14].

The results of the levels of natural radionuclides and ^{137}Cs in cereals are presented in tab. 5. Although it is shown in the literature (for example [12]) that Cs is analogous to K and thus it takes part in metabolic reactions, our measurements indicate that this radionuclide is not accumulated in the grain in the concentration that can be detected. It would be necessary to perform gamma spectrometry of other parts of the plant to establish whether ^{137}Cs is retained in the root, or the stem of the plant.

Uranium concentrations in plants are generally several orders of magnitude lower than in soil [12], and in this study the activity concentration of ^{235}U and ^{238}U in cereals was below minimal detectable activity (MDA) concentration , which is in agreement with the findings in [8] and therefore, the transfer factors for ^{137}Cs, ^{235}U, and ^{238}U were not calculated.

Thorium is known to be relatively immobile in soils and plants. Transfer factor may be influenced by addition of phosphate that may reduce the bioavailability of thorium [12]. As it can be seen from tab. 5, in almost all cereal samples, the concentration of ^{232}Th was below MDA.

Table 6 presents the mean values of the specific activities of the radionuclides in cereals sampled at the investigated locations. The specific activity of ^{232}Th (2.6 Bq/kg of dry matter) presented in tab. 5, refers only to the sample of wheat from the village of Korbevac.

The values of the calculated TF are presented in tab. 7. The value of the specific activity in soil used to calculate the TF, was the mean specific activity of the radionuclides, for the different sampling depths, at the given location.

Table 6. Mean values of the radionuclides specific activities in cereals

Radionuclide	Mean specific activity in cereals dry matter [$Bqkg^{-1}$]		
	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			

Table 7. The TF for different radionuclides

Cereal	Transfer factor		
	TF(^{226}Ra)	TF(^{232}Th)	TF(^{40}K)
	Korbevac		
Corn	0.009	–	0.151
Wheat	0.052	0.051	0.209
	Suvi Dol		
Corn	–	–	0.144
Wheat	0.008	–	0.224
	Bujkovac		
Corn	0.061	–	0.174
Wheat	0.016	–	0.200
Barley	0.074	–	0.392

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 transfer factors for cereals indicate that ^{40}K and ^{226}Ra are the main radionuclides transferred into the cereals grain. The TF for ^{40}K (0.144-0.392) are higher than TF for ^{226}Ra and ^{232}Th by an order of magnitude (0.008-0.074 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 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 % to 16 % is accumulated in the grain [15]. The addition of phosphate to soil reduces the availability of thorium for root uptake through 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]).

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 4 to 5 times lower due to the water content [7].

CONCLUSIONS

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 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 are the main radionuclides that are transferred in cereals. This evaluation is most important for 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 to 0.392, for ^{226}Ra the values of transfer factors were in the range of 0.008 to 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 4 to 5 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 where under the MDA.

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.

AUTHORS' CONTRIBUTIONS

Theoretical analysis was carried out by J. S. Marković, S. M. Stevović, and D. J. Todorović. Exper-

iments were carried out by J. S. Marković, M. M. Rajić, D. J. Todorović, and J. D. Krneta-Nikolić. All authors analyzed and discussed the results. The manuscript was written by J. S. Marković, S. M. Stević, M. M. Rajić, D. J. Todorović, and J. D. Krneta-Nikolić. The figure was prepared by J. S. Marković.

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Received on February 23, 2016
Accepted December 1, 2016

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ВРЕДНОСТИ ТРАНСФЕР ФАКТОРА ЗА СИСТЕМ ЗЕМЉА-ЖИТАРИЦЕ У ПЧИЊСКОМ ОКРУГУ У СРБИЈИ

Циљ овог рада је процена вредности трансфер фактора за природне радионуклиде (^{40}K, ^{226}Ra, ^{232}Th, ^{235}U, ^{238}U) и ^{137}Cs из земљишта у биљке (житарице: пшеница, кукуруз и овас) као параметра који је у пољопривреди важан приликом избора локације за сетву и врсте житарица које ће бити посејане. Резултати представљени у овом раду односе се на систем "земљиште-житарице" у Пчињском округу, у Србији. Укупно девет узорака земљишта и седам узорака житарица је испитивано у Лабораторији за заштиту од зрачења и заштиту животне средине Института за нуклеарне науке „Винча", гама спектрометријом на три HPGe детектора. Вредности трансфер фактора за ^{226}Ra у свим узорцима ниже су од вредности трансфер фактора за ^{40}K, али се налазе у оквиру вредности описаних у литератури. На три испитиване локације, израчунате вредности трансфер фактора за ^{40}K су биле у опсегу од 0.144 до 0.392, док су за ^{226}Ra биле у опсегу од 0.008 до 0.074. Само једна вредност (0.051) добијена је за трансфер фактор за ^{232}Th. Вредности ^{137}Cs као и изотопа уранијума у свим узорцима житарица биле су испод минималне детектабилне специфичне активности. Такође, израчунате су и јачина апсорбоване дозе и годишња апсорбована доза која потиче од природних радионуклида у земљишту. Јачина апсорбоване дозе је у опсегу од 49 nSv/h до 86 nSv/h, док је годишња апсорбована доза у опсегу од 0.061 mSv до 0.105 mSv. Истраживање представљено у овом раду је прво те врсте спроведено у Пчињском региону, стога резултати ових мерења могу да послуже као база за даља мерења и мониторинг.

Кључне речи: трансфер фактор, природни радионуклид, ^{137}Cs, земљиште, житарица, животна околина