Arch. Biol. Sci., Belgrade, 62 (2), 301-307, 2010 DOI:10.2298/ABS1002301G

NATURAL AND ANTHROPOGENIC RADIOACTIVITY OF FEEDSTUFFS, MOSSES AND SOIL IN THE BELGRADE ENVIRONMENT, SERBIA

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Abstract – By gamma spectroscopic measurement a content of natural radio-nuclides (^{40}K, ^{238}U, ^{226}Ra, ^{232}Th) and ^{137}Cs was determined in samples of soil, alfalfa, maize and moss on six sites in the surroundings of Belgrade. Natural radio-nuclides in the soil were at the level characteristic for Serbia, whereas a relatively high level of activity of ^{137}Cs (around 30 Bq kg^{-1}) was determined. On the other hand, in plant samples mostly used as feed (such as alfalfa and maize) the concentration of natural radio-nuclide activity and ^{137}Cs was relatively low, i.e. below the range of detection. The content of natural radio-nuclides in moss was within the standard range of values specific for Serbia. However, the activity level of ^{137}Cs in moss gathered from the wider area around Belgrade, was high, the highest measured level being in the Avala-Zuce area (158-221 Bq kg^{-1}). Our results show that this radio-nuclide is still present in the living environment of Belgrade even 20 years after the Chernobyl disaster, and that moss is a good indicator of living environment ^{137}Cs contamination.

Keywords: Belgrade environment, feedstuffs, mosses, radio-nuclides, soil

UDC 504.5(497.111)

INTRODUCTION

The increased use of nuclear energy worldwide, along with nuclear weapon probes, coal combustion, the production and use of phosphorus fertilizers, the mining industry and the formation of radioactive waste dumps, all contribute to the distribution of natural radioactivity. The quantity of radio-nuclides from the cited sources has significant impact on the environment (Pietrzak-Flis et al., 2000; Pantelić et al., 2004; Bikit et al., 2005). One of the important anthropogenic sources of environmental pollution by the primordial radio-nuclides ^{226}Ra, ^{232}Th, and ^{40}K are phosphorus fertilizers (Adel et al., 2005). Uranium, which is of anthropogenic origin, is often present in soil in a form which is easily accessible to plants, which offers a greater possibility for its incorporation in a food chain (Bolca et al., 2007).

The partial meltdown of the reactor at Chernobyl in April 1986 released high amounts of radionuclides into environment. ^{131}I, ^{134}Cs and ^{137}Cs were the most important and most dangerous radionuclides released and spread over a large part of Europe.

After the Chernobyl nuclear accident, Hecht (1988), Nifontova (1995), Delfanti et al., (1999), Dragović et al., (2004), Dragović et al., (2007) concluded that mosses, lichens and mushrooms were typical representatives of bioindicator plants, for both radiocesium and natural radio-nuclides presented in the environment.

Physical decay, weathering and other major factors have contributed to the significant decline of radionuclides in the environment. The level of the long-lived fission product, ^{137}Cs, has been reduced to very low values (Pantelić et al., 2007).

Mosses have some advantages compared to other bioindicator species. The accumulating capacity of mosses is higher than that of other plants

Table 1. Species of mosses collected in the area of six regions around Belgrade classified according to their individual growth substrate

Regions	Mosees species	
	Epilitic (stone)	Epixilic (tree bark)
Opovo	*Grimmia pulvinata (Hedw.) Sm.*	*Hypnum cupressiforme Hedw.*
Vinča-Boleč	*Tortula muralis Hedw.*	*Pseudoleskeella nervosa (Bird.) Nyholm*
Kosmaj- Nemenikuće	*Pylaisia polyantha (Hedw.) Schimp.*	*Dicranella heteromalla (Hedw.) Schimp*
Avala-Zuce	*Ditrichum pusillum (Hedw.) Hampe*	*Hypnum cupressiforme Hedw.*
Barajevo	*Didymodon spadiceus (Mitt.) Limpr.*	*Anomodon attenuatus (Hedw.) Huebener*
Jakovo-Bojčinska šuma	*Brachythecium rutabulum (Hedw.) Schimp*	*Brachytheciastrum velutinum (Hedw.) Ignatov &Hutten*

(Elstner et al., 1987; Mishev et al., 1996). They capture airborne particulates by both passive and active processes through an extra cellular ion-exchange process (Knight et al., 1961).

The radiological investigation of a certain area involves the collecting of samples which represent potential danger for the health of the population. The goal of this paper was to determine the level of activity and presence of natural radionuclides (^{40}K, ^{238}U, ^{226}Ra, ^{232}Th) and the anthropogenic radionuclide ^{137}Cs in the samples of soil (arable and uncultivated), feedstuffs (maize and alfalfa) and mosses on the territory of six regions around Belgrade from which most food for human consumption originates.

The goal of this paper was to determine the level of activity of radio-nuclides ^{40}K, ^{238}U, ^{226}Ra, ^{232}Th and ^{137}Cs in the soil, feedstuffs and mosses (bioindicator species) in the Belgrade environment, Serbia.

MATERIALS AND METHODS

The samples of soil, feedstuffs and mosses were collected during the period of 2007 – 2008 in six regions around Belgrade: Avala-Zuce (a village near the mountain Avala), Kosmaj-Nemenikuće (a village near the mountain Kosmaj), Barajevo, Grocka - Vinča (a village near the river Danube), Opovo (a village near the town of Pančevo and the river Tamiš) and Jakovo-Bojčin forest (a village near the Bojčin forest and the Sava river). Each sample was taken in triplicate from each sampling site.

These sampling sites were selected because they represent significant arable land for the production of food (and feed) for the Belgrade population.

On each of the six sampling sites two types of soil samples were taken: cultivated and uncultivated. From the arable land (cultivated soil) samples of maize and alfalfa were collected, from uncultivated soil samples of mosses were taken.

Soil samples in amounts of 3-5 kg were collected from a depth of 10-20 cm, homogenized, dried at a temperature of 105°C and put in 1l Marineli beakers. Alfalfa and maize samples were collected in amounts of 2-5 kg, ground and burned to mineral ash. Plastic 200 ml vessels were used for measurements. Moss samples collected in an amount of 2 kg (in fresh form) from uncultivated soil were sorted by their individual growth substrate, into two groups: epilithic (stone) and epixilic (tree bark). Radioactivity was measured in the mixture of moss

Table 2. Average activity concentration of radionuclides in the soil and plants (Bq kg^{-1}) from the environment of Belgrade

Samples	^{40}K	^{238}U	^{226}Ra	^{232}Th	^{137}Cs
Soil cultivated	608 ± 15	47 ± 7	51 ± 5	50 ± 4	25 ± 2
Soil uncultivated	560 ± 16	41 ± 5	48 ± 2	47 ± 2	39 ± 1
Alfaalfa	538 ± 19	< 5	< 9.6	< 1.6	< 0.4
Maize	95 ± 5	< 1.9	< 1.5	< 0.3	< 0.1

mean ± standard deviation

species. The moss samples were first cleaned of soil and other impurities and dried at a temperature of 80°C until they became a constant mass, were ground and packed in 200 ml plastic vessels.

All samples were kept in the laboratory (at room temperature) for 40 days prior to measurement in order to achieve a radioactive balance. The radioactivity of the samples was determined by gamma spectrometric measurements on an HPGe detector (Ortec, USA) with a relative efficiency of 30% and energy resolution 1.85 keV (1332.5 ^{60}Co). Radioactive measurements were performed according to the procedure described by Mitrović et al., (2009).

All obtained results were expressed as means ± standard deviation (MEAN ± SD) and are shown in tables (Snedecor and Cochrain, 1971).

RESULTS AND DISCUSSION

Species of mosses collected in each region are shown in Table 1.

Average values of activity of natural and artificial radio-nuclides in samples of cultivated and uncultivated soils, alfalfa and maize, gathered from six sites around Belgrade are shown in Table 2.

In the environment radionuclides of natural (primordial) and artificial (anthropogenic) origin are present. Primordial radionuclides are formed as a result of cosmic and terrestrial radiance and have relatively low concentration. The most significant representatives of natural radionuclides are ^{40}K, ^{238}U, ^{226}Ra and ^{232}Th. The level of uranium in arable land can be higher as a result of the increased use of phosphorous fertilizers, whose production is directly dependent on the use of phosphate minerals which can often carry significant levels of radioactive uranium. This implies the importance of control of the raw materials used in phosphorous fertilizer production. The presence of anthropogenic radionuclides in the environment is generally a result of nuclear probes and accidents in nuclear plants.

Pantelić et al., (2007) state that after the Chernobyl accident (1986) a high average activity concentration of ^{137}Cs was measured in alfalfa (791 Bq/kg) and in mosses (600-2000 Bq/kg) but not in maize (2.1 Bq/kg). Ten years later the activity concentration of ^{137}Cs has decreased to an average level of 0.5 Bq/kg in alfalfa and 0.1 Bq/kg in maize. Activity in mosses has decreased but was still relatively high, ranging between 90 and 210 Bq/kg . These results are in accordance with results obtained in our trial.

Even 20 years after the Chernobyl accident, the presence of ^{137}Cs can be determined in some samples due to its long half-life (27.7 years). Therefore, it is of great importance to investigate bioindicators because they present the most reliable indicator of whether the living environment was contaminated with ^{137}Cs as a result of the Chernobyl accident.

The ability of mosses to show the quality of air, water and soil, based on their presence, absence or floristic composition, presents an important trait and it enables their use as bioindicators of radioactive contamination. Mosses are, due to their anatomical, morphological and physiological characteristics,

Table 3. Average activity concentration of radionuclides in samples of epilithic mosses (Bq kg^{-1}) originating from the Belgrade environment

Regions	^{40}K	^{238}U	^{226}Ra	^{232}Th	^{137}Cs
Opovo	407 ± 17	< 20	18 ± 3	22 ± 4	41 ± 2
Vinča - Boleč	384 ± 12	53 ± 9	70 ± 8	18 ± 1.8	89 ± 3
Kosmaj - Nemenikuće	370 ± 17	56 ± 10	75 ± 10	18.4 ±3.8	99 ± 3
Avala - Zuce	178 ± 9	< 12.6	6.1 ± 1.3	8.4 ± 2.1	158 ± 5
Barajevo	154 ± 7	< 16	< 26	< 4.7	9 ± 0.4
Jakovo – Bojčin forest	270 ± 16	39 ± 9	11 ± 2	15 ± 3	14 ± 1

mean ± standard deviation

capable of accumulating pollutants from aero sediments, radionuclides and pollutants from the soil in their tissue. They represent good bio-indicators because they: have wide ecological and geological distribution, are present during whole year (and are in most cases perennial plants). Also, they exhibit a high degree of tolerance to pollutants, absorption of mineral matter is performed by the whole body surface and on leaves they do not have either cuticle or stomas. Thus, a clear correlation between the speed of accumulation of radioactive contaminants and moss growth can be observed (Grdović, 2005). Lately, and especially after the Chernobyl accident, mosses are used as reliable indicators of increased activity concentration of radionuclides in the environment (Stanković et al., 1997).

^{40}K causes the most dominant natural radioactivity. The activity level of this radionuclide was high in both soil and alfalfa (538-608 Bq/kg), while in maize it was significantly lower (95 Bq/kg). The activity levels of the other natural radionuclides (^{238}U, ^{226} Ra and ^{232}Th) were lower and within the limits of average values for Serbia (Pantelić et al., 2004).

The activity level of ^{137}Cs in the soil was in a range of 25-39 Bq/kg. These results indicate that even after 20 years since the Chernobyl nuclear accident, this radionuclide is present in the natural environment of Belgrade. In the cultivated soil lower levels of ^{137}Cs were found than in the non-cultivated soil, but the differences were not significant and was within the limits of the average values for Serbia (Pantelić et al., 2004). These results are in accordance with those of Bikit et al., (2005) and Bolca et al., (2007).

Measured specific activities of natural radionuclides in the plant samples, except for ^{40}K, were below detection limits for the applied measurement conditions (geometry of measurement, quantity of the sample and time of measurement). These plants are commonly used as feedstuffs (alfalfa, maize) and ^{40}K is determined as a major contribution to the total activity of this kind of sample. Also, all measured and reported specific activities for artificial radionuclides (just ^{137}Cs is reported) are below the minimum detectable activity (MDA) values for performed measurements. These results are in accordance with results presented earlier for some regions of Serbia (Pantelić et al., 2004 and 2007).

Table 4. Average activity concentration of radionuclides in samples of epixilic mosses (Bq kg[1]) originating from Belgrade environment

Regions	^{40}K	^{238}U	^{226}Ra	^{232}Th	^{137}Cs
Opovo	451 ± 17	38 ± 0.9	41 ± 3	37 ± 4	68 ± 5
Vinča - Boleč	150 ± 9	35 ± 4	7.4 ± 1.1	7.2 ± 1.3	43 ± 5
Kosmaj - Nemenikuće	110 ± 7	32 ± 9	7.1 ± 1.4	7.7 ± 1.9	32 ± 9
Avala - Zuce	490 ± 19	80 ± 10	38 ± 7	45 ± 4	221 ± 13
Barajevo	322 ± 24	< 113	< 189	< 23	63 ± 3
Jakovo – Bojčin forest	247 ± 10	20 ± 3	7.7 ± 1.1	16 ± 2	16 ± 1

mean ± standard deviation

Tables 3 and 4 show the average values of activity levels of natural and artificial radio-nuclides in moss samples gathered from the investigated regions around Belgrade.

There were no significant differences in the activities of natural radio-nuclides between the moss species regarding growth substrate. For all measured samples, the arithmetic mean of ^{238} U activity was 43 Bq kg^{-1}, for ^{226}Ra it was 36 Bq kg, $^{-}$ for ^{40}K it was 294 Bq kg^{-1}, and for ^{232}Th it was 19 Bq kg^{-1}. Similar results were also found by Dragović et al., (2007),

^{137}Cs presence in the environment is due to atmospheric fallout from the Chernobyl accident which occurred in 1986. In the epilithic moss samples collected in the environment around Belgrade, ^{137}Cs activity ranged from 9 Bq kg^{-1} (Barajevo) and 14 Bq kg^{-1} (Jakovo) to 158 Bq kg^{-1} d.m. (Avala-Zuce). In the epixilic moss samples, ^{137}Cs activity ranged from 16 Bq kg^{-1} (Jakovo) and 32 Bq kg^{-1} (Kosmaj) to 221 Bq kg^{-1} d.m. (Avala-Zuce). Similar results were also found by Delfanti et al., (1999) but Dragovic et al., (2004) reported higher ^{137}Cs activities in the mosses collected in the environment of mountain regions of Serbia and Montenegro. Even 20 years after the accident the presence of ^{137}Cs can be detected in the environment of Belgrade. For that purpose mosses are a useful bio-indicator.

Based on the findings shown in Tables 2, 3 and 4 it can be seen that the activity concentration of measured radionuclides, except for ^{40}K, was significantly higher in mosses compared to maize and alfalfa. This was the case for both natural radionuclides (^{238}U, ^{226}Ra, ^{232}Th) and for ^{137}Cs, originating from the Chernobyl accident. It can be concluded that maize and alfalfa are plants that do not possess the ability to accumulate radionuclides and are safe as animal feed. The mosses present a more reliable indicator for radioactive contamination of the environment.

Acknowledgment - This work was supported by grant no. 19022 of the Ministry of Science and Technology of Serbia.

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ПРИРОДНА И ВЕШТАЧКА РАДИОАКТИВНОСТ СТОЧНЕ ХРАНЕ, МАХОВИНА И ЗЕМЉИШТА У ОКОЛИНИ БЕОГРАДА

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Гамаспектрометријским мерењима извршено је одређивање садржаја природних радионуклида (^{40}K, ^{238}U, ^{226}Ra, ^{232}Th) и ^{137}Cs у узорцима земљишта, луцерке, кукуруза и маховина, на шест локалитета у околини Београда. Природни радионуклиди у земљишту су били на нивоу граница специфичним за Србију, док је установљен релативно висок ниво активности ^{137}Cs (oko 30 Bq kg^{-1}). Насупрот томе, у узорцима биљака, које се најчешће користе као сточна храна (луцерка, кукуруз), концентрација активности природних радионуклида и ^{137}Cs је била ниска односно испод границе детекције. Садржај природних радионуклида у маховинама био је у стандардним границама детекције специфичним за Србију. Међутим, ниво активности ^{137}Cs у маховинама сакупљеним са широког подручја околине Београда, био је висок, а највиши са подручја Авала-Зуце (158-221 Bq kg^{-1}). Добијени резултати указују да је овај радионуклид и после 20 година од несреће у Чернобилу, присутан у животној средини Београда и да су маховине добар индикатор контаминације животне средине са ^{137}Cs.