ORIGINAL ARTICLE



Assessment of radiation exposure to human and non-human biota due to natural radionuclides in terrestrial environment of Belgrade, the capital of Serbia

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Received: 26 October 2017 / Accepted: 31 March 2018 / Published online: 7 April 2018 © Springer-Verlag GmbH Germany, part of Springer Nature 2018

Abstract

The main focus of this study was to assess radiation exposure to human and non-human biota due to natural radionuclides in soil of the Serbian capital. For the first time, ERICA tool was employed for calculation of gamma dose rates to non-human biota in this area. In analyzed soils, the mean values of ²²⁶Ra, ²³²Th and ⁴⁰K specific activities were found to be 35, 43 and 490 Bq kg⁻¹, respectively. The distribution of analyzed natural radionuclides in soils was discussed in respect to its statistically significant correlations with sand, silt, clay, carbonates, cation exchange capacity and pH value. The annual outdoor effective dose rates to the population varied from 48 to 98 μ Sv, and the total dose rates to terrestrial biota, calculated by ERICA tool, varied from 9.84 × 10⁻² μ Gy h⁻¹ (for tree) to 5.54 × 10⁺⁰ μ Gy h⁻¹ (for lichen and bryophytes). The results obtained could serve as a baseline data for the assessment of possible anthropogenic enhancement of the total dose rate to human and non-human biota of the study area.

Keywords Natural radionuclides · Statistical analysis · Gamma dose rates · ERICA tool

Introduction

Human and non-human biota are exposed to ionization radiation from natural (such as cosmic and terrestrial radiation) and man-made sources of radiation (UNSCEAR 2010, 2011). Primordial gamma-emitting radionuclides such as ²³⁸U, ²³²Th along with their daughter products and ⁴⁰K, present in trace amounts in the soil, represent the main external sources of exposure (UNSCEAR 2010), and a number of surveys were carried out worldwide to determine specific activities of natural radionuclides in soils and associated gamma dose rate (Abdalhamid et al. 2017; Antovic et al. 2012; Baykara and Doğru 2009; Bouhila et al. 2017;

Dragović et al. 2006; Huy et al. 2012; Taskin et al. 2009). The data obtained in these surveys are dominated by dose assessment to humans. The need to protect the non-human biota from exposure to ionizing radiation led to the development a number of models and approaches (e.g., RESRAD-BIOTA, ERICA tool, DosDiMEco, LIETDOS-BIO) for the dose and risk assessment (Beresford et al. 2008a; IAEA 2012; Vives i Batlle et al. 2007, 2011). The ERICA tool has been widely used worldwide to assess the radiological risk to biota within terrestrial and aquatic (i.e., freshwater and marine) ecosystems (Botwe et al. 2017; Čern et al. 2012; Doering and Bollhöfer 2016; Hosseini et al. 2011; Li et al. 2015; Mazeika et al. 2016; Nedveckaite et al. 2011; Wood et al. 2008).

A few assessment studies dealing with radiological risk due to naturally occurring gamma-emitting radionuclides and Chernobyl-derived ¹³⁷Cs in soil were conducted so far on the territory of Belgrade urban area (Janković-Mandić et al. 2014; Janković Mandić and Dragović 2010; Petrović et al. 2013). However, their main focus was the radiation exposure to humans. In available literature, there are no data on application of ERICA tool for non-human biota of Belgrade urban area. Although previous studies conducted on the territory of Belgrade area did not indicate the increased

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specific activities of natural radionuclides in soil and corresponding dose rates to population, this study will be the first effort to establish reference levels of dose rates to both human and terrestrial non-human biota. The importance of knowing and monitoring these doses lies in the fact that the environment of Belgrade is the habitat of many plant and animal species, with a number of protected natural assets such as Miljakovačka šuma, Topčider, Košutnjak, Banjička šuma. (Đurdić et al. 2011) and also over million people live within urban area of the city of Belgrade. Accordingly, the objectives of this study were to determine the ²²⁶Ra, ²³²Th and ⁴⁰K specific activities in surface soil samples collected from the territory of Belgrade, to investigate correlations among radionuclides and soil properties by the Pearson correlation analysis and to estimate dose rates to the population and to terrestrial biota in the Belgrade area based on the measured radionuclides in soil.

Materials and methods

Study area and sample collection

Belgrade, the largest and capital city of Serbia, is situated in the northern part of Serbia (Southeastern Europe). The agglomeration of Belgrade covers the area at the contact of the Pannonian Basin and its southern rim, represented by the Sumadija beam of low mountains (Komatina and Komatina 1999). It is situated on the confluence of the two great rivers, the Sava and the Danube. The southeastern part of the city lies on the hilly terrains of Belgrade Posavina, with elements of the karst relief. Most of New Belgrade was developed in the lowland on sandy and alluvial terrains. Most of the city is covered by neogene deposits underlined by flysch sediments. The territory of Belgrade is a zone of contact between the two tectonic units, the Vardar zone in the south and the Pannonian depression in the north. Significant tectonic processes took place during the neogene and post-neogene phases. The area is characterized by a diverse geological structure with significant distribution of clay, sand, conglomerates, and quaternary sediments dominated by eolian, river and lake loess, as well as other sediments of Pleistocene age, especially in the area of New Belgrade and Zemun. In the southeastern parts of the city, there are magmatic rocks and serpentinized peridotites of Mesozoic age.

The territory of Belgrade is under the influence of the steppe-continental climate. According to the Köppen–Geiger climatic classification, it has the Cfa (humid subtropical climate), with characteristics of moderate-warm climate with a warm summer (Šegota 1988; Kottek et al. 2006). The coldest month is January with an average temperature of 0.1 °C, and the hottest July with an average perennial temperature of 22 °C (Dragović and Kićović 2001). The average insolation is 2096 h. Growth of average annual air temperature is not continuous, but the trend of longer growth periods is observed. The average atmospheric pressure in the territory of Belgrade is 1001 millibars. During the year, the silence conditions are dominated, especially in the summer months, caused by the anticyclone and isobaric conditions. The winds of the south and southeast are the most frequent. These winds, known as Kosava, most commonly occur in the second half of the year as a result of unequal air pressure. The winter Kosava usually lasts 2-3 days, but can be extended up to 15 days. A cozy and cold winter Kosava contributes favorably to the climate of the city, reducing the level of pollution. The average relative atmospheric humidity is about 70%, with the highest values in December and the lowest in July and August. The high relative humidity in the winter months in the absence of wind affects increased air pollution because humid air binds particles that are the product of pollution. The mean annual precipitation is 669 mm.

Simplified geological map, based on a geological map of Serbia at 1:500,000 scale (Federal Geological Survey 1970), of study area with sampling locations is presented in Fig. 1. A total of 83 uncultivated soil samples were collected up to 15 cm depth in 2011 according to International Atomic Energy Agency (IAEA) guidelines for environmental contaminants (IAEA 2004). The soil samples were dried to constant weight, mechanically homogenized and sifted through a 2 mm mesh sieve. The 0.5 L Marinelli beakers full of samples were then hermetically sealed and stored for about 4 weeks, in order to achieve radioactive equilibrium among radium-226 and its daughters, prior to radioactivity measurements.

Analytical methods

The radioactivity of soil samples was measured using highpurity germanium (HPGe) gamma-ray spectrometer ORTEC-AMETEK (34% relative efficiency and 1.65 keV resolution at 1332 keV for ⁶⁰Co) for 16.7 h in order to provide an analytical precision of about $\pm 5-10\%$ at the 95% level of confidence. The calibration (energy and efficiency) of the HPGe gammaray spectrometer was performed using MBSS2 standard in 500 mL Marinelli beaker, Cert. No: 9031-Ol-419/10 supplied by CMI-Czech Metrology Institute. For quality assurance and quality control purposes, i.e., to assess the possible influence of inhomogeneous distribution of the sample and other sources of the uncertainty, the reference materials (IAEA-RGU-1 and IAEA-RGTh-1) and intercomparison exercises were used on regular basis. The ²²⁶Ra specific activities were determined using the most intense gamma-ray lines of ²¹⁴Bi-609.3 keV and ²¹⁴Pb-295.2 and 352.0 keV. The ²³²Th specific activities were determined using the most intense gamma-ray lines of ²²⁸Ac-338.4; 911.1 and 968.9 keV. For determination



Fig. 1 Simplified geological map of study area with sampling locations

of ⁴⁰K specific activities, its gamma-ray line at 1460.8 keV was used. For spectra analysis, Gamma Vision 32 MCA emulation software was used (ORTEC 2001).

Standard methods were used for determination of the following soil properties: particle size distribution (sand, silt and clay), soil pH (in H_2O), specific electrical conductivity—SEC, organic matter content—OM, carbonate content—CaCO₃ and total cation exchange capacity of the sorptive complex—CEC (ISO 10390 2005; ISO 10693 1995; ISO 11265 1994; Kappen 1929; Rowell 1997; Simakov 1957; Walkley and Black 1934). The Statistical Package for the Social Sciences—SPSS 16.0—was employed for statistical data analysis (SPSS 2007).

Assessment of the doses from terrestrial exposure of population

Total absorbed dose rates D_{tot} (nGy h⁻¹) in air at a height of 1 m above the ground surface were calculated from ²²⁶Ra,

²³²Th and ⁴⁰K specific activities in surface soil samples. Outdoor annual effective dose $AED_{outdoor}$ (µSv) was calculated using the factor of 0.7 Sv Gy⁻¹ to convert the absorbed dose rate in air to the effective dose received by adults and 0.2 for the outdoor occupancy factor (the fraction of time spent outdoors). Details about equations used for calculations of D_{tot} and $AED_{outdoor}$ can be found in UNSCEAR (2000).

Radiation dose assessment for the biota of terrestrial ecosystems

ERICA tool (version 1.2.1) was used to estimate dose rates to default terrestrial reference organisms: amphibian (frog), annelid (earthworm), arthropod—detritivorous (woodlouse), bird (duck), flying insects (bee), grasses and herbs (wild grass), lichen and bryophytes, mammal—large (deer), mammal—small burrowing (rat), mollusk—gastropod (snail), reptile (snake (on soil)), shrub and tree (Brown et al. 2008, 2016; ERICA 2007). At the present study, the Tier 2 of the ERICA tool was applied. Maximum measured ²²⁶Ra and ²³²Th specific activities in soil were used as input data in the model, in order to ensure that maximum possible value of dose rates to non-human biota was below the screening dose rate criterion of 10 μ Gy h⁻¹. Default uncertainty factor (UF) of 3 was selected at Tier 2 to account for the uncertainties involved in the assessment method. According to the definition available in ERICA tool software system, an UF = 3will test for 5% probability of exceeding the dose screening value, assuming that the risk quotient distribution is exponential (ERICA 2007). The dose rate calculations in Tool are performed using the inputted data by applying DCCs, dose conversion coefficients in μ Gy h⁻¹ per Bq kg⁻¹ fresh weight and weighting factors of 10 for α , 3 for low β and 1 for (high energy) β and γ radiation. Details about ERICA tool and their uncertainties can be found in a number of studies (Beresford et al. 2008b; Brown et al. 2008, 2016; IAEA 2014; Larsson 2008; Oughton et al. 2008).

Results and discussion

Specific activities of natural radionuclides in soils and their relationship with soil properties

The values of radionuclides specific activities in analyzed soils varied from 19 to 51 Bq kg⁻¹ (mean: 35 Bq kg⁻¹) for ²²⁶Ra; from 23 to 58 Bq kg⁻¹ (mean: 43 Bq kg⁻¹) for ²³²Th; and from 310 to 650 Bq kg⁻¹ (mean: 490 Bq kg⁻¹) for ⁴⁰K (Table 1). The great majority of soil samples are taken from areas where quaternary continental deposits and limestones with marine clastics are dominant geological rock types, 36 and 26, respectively. The rest of the soil samples (25%) are from areas of neogene lake deposits—10 samples, upper cretaceous flysch and limestones—7 samples, and only 4 samples are from brackish sediments, limestones and coal (Fig. 1). Accordingly, the obtained variability of radionuclide specific activities in soils can be related to the variety of geological composition of the investigated area. The mean values of ²²⁶Ra, ²³²Th and ⁴⁰K specific activities obtained

in this study are comparable to those obtained in different regions of the country as well as in countries in the region with similar geological composition and geotectonic structures (Dragović et al. 2014) (Table 2). For evaluation of relationships between ²²⁶Ra, ²³²Th and ⁴⁰K specific activities and soil properties, the Pearson correlation analysis was used. Basic descriptive statistics of the soil properties are presented in Table 3. The Pearson correlation coefficients among ²²⁶Ra, ²³²Th and ⁴⁰K specific activities and soil properties are shown in Table 4. The positive correlation coefficients were found between radionuclides specific activities (Table 4), which indicate their common source in soil (parent material) (Navas et al. 2011; Ćujić et al. 2015). Radionuclides specific activities were found to be strong positively correlated with both the clay and silt soil fractions, but negatively correlated with sand soil fraction (Table 4). These correlations indicated that radionuclides are associated with the finer soil fractions (clay and silt), which was expected since it is known that soils contain a number of radionuclide adsorbing components in the silt and clay fractions (Kumar et al. 2013). The correlations obtained in this study are in accordance with those reported in similar studies worldwide (Belivermis et al. 2010; Ćujić et al. 2015; Dragović et al. 2012a; Navas et al. 2002). Blanco Rodríguez et al. (2008) observed that the ²³⁸U, ²³⁰Th and ²²⁶Ra activity concentration in soil decreases as the particle size increases. Caridi et al. (2016) also found that the ²²⁶Ra, ²³²Th and ⁴⁰K concentrations increase with decreasing sediment particle size, and highest concentrations of radionuclides were obtained in pelite ($< 65 \mu m$) fraction. The carbonate content was negatively correlated with radionuclides specific activities (Table 4), which agrees with results reported in similar studies (Ćujić et al. 2015; Dragović et al. 2012a; Navas et al. 2011; Tomić et al. 2011). According to Ćujić et al. (2015), Dragović et al. (2012a) and Elejalde et al. (1996) negative correlations between radionuclides and carbonates suggest their binding in soils with minerals other than calcite, probably in silicates derived during weathering processes from parent rocks. In analyzed soils, radionuclides specific activities were negatively correlated with the cation exchange

Table 1 Descriptive statistics
of ²²⁶ Ra, ²³² Th and ⁴⁰ K specific
activities in surface soils from
Belgrade and estimated gamma
dose rates and annual outdoor
effective doses

	Specific	activity (Bq l	(g^{-1})	Gamma	a dose rate	AED _{outdoor}		
	²²⁶ Ra	²³² Th	⁴⁰ K	$\overline{D_{\mathrm{Ra}}}$	D_{Th}	D _K	D _{tot}	(µSv)
Mean	35	43	490	16	26	21	63	77
Median	36	43	497	16	26	21	63	77
Mode	33 ^a	49	314	14 ^a	23 ^a	24	61	75
SD	6.3	8.4	69	2.9	5.1	2.9	9.7	12
Range	31	35	340	14	21	14	41	50
Minimum	19	23	310	9.0	14	13	39	48
Maximum	51	58	650	23	35	27	80	98

^aMultiple modes exist. The smallest value is shown

Table 2 Comparison of the 226 Ra, 232 Th, and 40 K specific activities (Bq kg⁻¹) in soils obtained in this study with values reported for different parts of the Serbia and countries in the region

City/country	²²⁶ Ra (Bq kg ⁻¹)	²³² Th (Bq kg ⁻¹)	40 K (Bq kg ⁻¹)	References
Belgrade/Serbia	34 (12–55)	39 (11–64)	508 (285–797)	Janković Mandić and Dragović (2010)
Belgrade/Serbia	-	38 (11-64)	501 (232-814)	Janković Mandić et al. (2010)
Kragujevac/Serbia	34 (20–55)	50 (30-73)	426 (167–559)	Milenkovic et al. (2015)
Subotica/Serbia	20 (12-33)	18 (13–23)	290 (260-390)	Janković Mandić et al. (2016)
Serbia	33 (8-88)	36 (9.5-84)	508 (45-900)	Dragović et al. (2012b)
Serbia and Montenegro	31 (14–55)	41 (18-83)	567 (271–919)	Dragović and Onjia (2006)
Montenegro	40 (11-216)	44 (18–107)	438 (245–711)	Antovic et al. (2012)
Bulgaria	45 (12-210)	30 (7-160)	400 (40-800)	UNSCEAR (2000)
Hungary	33 (14–76)	28 (12-45)	370 (79–570)	UNSCEAR (2000)
Romania	32 (8-60)	38 (11–75)	490 (250-1100)	UNSCEAR (2000)
Albania	_	24 (4–160)	360 (15-1150)	UNSCEAR (2000)
Croatia	54 (21–77)	45 (12-65)	490 (140-710)	UNSCEAR (2000)
Slovenia	41 (2–210)	35 (2-90)	370 (15-1410)	UNSCEAR (2000)
Belgrade/Serbia	35 (19–51)	43 (23–58)	490 (310-650)	Present study

 Table 3
 Basic descriptive statistics of the analyzed soil properties

	Sand (%)	Silt (%)	Clay (%)	Carbonates (%)	Cation exch. capac- ity (cmol kg ⁻¹)	Org. matter (%)	pН	Spec. electrical con- ductivity (μ S cm ⁻¹)
Mean	16	64	20	5.9	71	4.2	7.5	150
Median	13	65	19	5.6	93	4.0	7.6	120
Mode	4.7 ^a	56 ^a	22	0.07 ^a	95 ^a	2.7 ^a	7.2 ^a	106 ^a
SD	13	11	5.3	5.5	33	1.7	0.43	86
Range	59	57	25	26	96	8.5	2.0	529
Minimum	1.2	24	10	0.10	4.0	0.80	6.1	35
Maximum	60	81	35	26	100	9.3	8.2	564

^aMultiple modes exist. The smallest value is shown

Table 4 Pearson correlation coefficients between radionuclides specific activities and soil properties

	²²⁶ Ra	²³² Th	⁴⁰ K	Sand	Silt	Clay	CaCO ₃	CEC	ОМ	pН	SEC
²²⁶ Ra	1.000										
²³² Th	0.834**	1.000									
⁴⁰ K	0.505**	0.622**	1.000								
Sand	- 0.576**	- 0.713**	- 0.478**	1.000							
Silt	0.431**	0.555**	0.404**	- 0.911**	1.000						
Clay	0.471**	0.539**	0.293**	- 0.473**	0.067	1.000					
CaCO ₃	- 0.308**	- 0.547**	- 0.370**	0.264*	- 0.173	- 0.268*	1.000				
CEC	- 0.356**	- 0.538**	- 0.249*	0.429**	- 0.250*	- 0.503**	0.649**	1.000			
OM	- 0.030	- 0.098	- 0.059	0.055	0.046	- 0.230*	0.090	0.114	1.000		
pН	- 0.286**	- 0.302**	- 0.091	0.224*	- 0.093	- 0.343**	0.431**	0.541**	0.027	1.000	
SEC	- 0.059	- 0.111	0.023	0.302**	- 0.230*	- 0.240*	0.200	0.374**	0.380**	0.196	1.000

Data in bold indicates statistical significance

*Correlation is significant at the 0.05 level

**Correlation is significant at the 0.01 level

capacity (Table 4). This means that the cation exchange capacity decreases when the radionuclides specific activities in the soil increase which may be due to various factors, since cation exchange capacity of soils is mainly determined by clay content, the type of clay minerals present, and soil organic matter content as well as soil pH. Although it is known that organic matter influences the radionuclides mobility in the soil (Boggs et al. 1985), no correlations were found between the organic matter content and radionuclides specific activities in soils (Table 4). The lack of correlation between radionuclides specific activities and organic matter content indicates association of these radionuclides with the mineral soil fraction. The lack of correlation was also reported by Navas et al. (2002, 2011). Negative correlations between soil pH and both ²²⁶Ra and ²³²Th specific activities were obtained (Table 4), which agrees with the findings of Belivermis et al. (2010), Dragović et al. (2012a) and Milenkovic et al. (2015). Guo et al. (2008) indicated that soil pH influences the distribution and mobility of thorium fractions in soil. Chandrasekaran et al. (2015) found concentrations of ²³⁸U and ²³²Th and absorbed gamma dose rate to be mainly controlled by physicochemical properties such as sand, silt, clay, pH and electrical conductivity.

Population doses from terrestrial gamma exposure

In the study area, the values of the D_{tot} varied from 39 to 80 nGy h^{-1} , with the mean value of 63 nGy h^{-1} (Table 1). Variability of D_{tot} reflects the variability of radionuclides specific activities among sampling locations. The highest contribution due to 232 Th (26 nGy h⁻¹, i.e. 41.3%) to the total gamma dose rate was found, followed by contribution due to 40 K (21 nGy h⁻¹, i.e. 33.3%) and 226 Ra (16 nGy h⁻¹, i.e. 25.4%) (Table 1). Results of the extensive survey conducted in Serbia and Montenegro show that contribution of ²²⁶Ra, ²³²Th and ⁴⁰K to the D_{tot} varied with sampling location and reflected the geological origin of the analyzed soils (Dragović et al. 2006). The normal frequency distribution of D_{Ra} , D_{Th} and $D_{\rm K}$ was obtained (Fig. 2), which is similar to results of the extensive research in Serbia and Montenegro (Dragović et al. 2006). The spatial distribution of AED_{outdoor} on the territory of Belgrade is presented in Fig. 3. Proportional symbol map is used for visualizing quantitative data and illustrating differences of values between locations. The center of every single circle represents a sampling location and the size of each circle is proportional to the value of AED_{outdoor} (μSv) . The value of each circle could be estimated using a map legend. The data are unclassed, and the legend includes the smallest (48 μ Sv) and the largest value (98 μ Sv) as well as some intermediate reference values (70, 80 and 90 µSv). The presence of overlaps between the circle symbols is a result of an attempt to easily see the differences of values associated with a specific location and identify patterns in the data. The values of the AED_{outdoor} varied from 48 to 98 μ Sv, with mean value of 77 μ Sv (Table 1), i.e., below the value of 1 mSv recommended by the ICRP for members of the public (ICRP 2007) and similar to worldwide average value of 0.07 mSv (UNSCEAR 2000). The lowest values of AED_{outdoor} are obtained within Quaternary continental deposits, clastics, deposits rich of organic matter and the highest within limestones and marine clastics (Fig. 3). Results of AED_{outdoor} obtained in this study match the AED_{outdoor} values obtained in previous studies conducted in Belgrade (72 µSv) (Janković Mandić and Dragović 2010) and also in Serbia and Montenegro (81.9 µSv) (Dragović et al. 2006). However, the mean value of the AED_{outdoor} obtained in this study is 18% lower then the average value obtained around the largest coalfired power plants (CFPP) in Serbia, 42 km upstream from Belgrade, where there was an indication of the pollution at some sampling locations (Ćujić et al. 2015).

Dose rates to terrestrial organisms

Calculated external, internal and total dose rates to terrestrial reference organisms are presented in Table 5. As observed from the results (Table 5), the internal dose rates to reference organisms were found to be higher than external dose rates from both ²²⁶Ra and ²³²Th. The higher contribution to total dose rate in grass and mammals derived from the internal exposure by ²²⁶Ra and ²²⁸Th Sotiropoulou et al. (2016) attributed to the way by which these radionuclides (as alpha emitters) irradiate the organism. The total dose rates for all terrestrial reference organisms (Table 5) were found to be much higher due to ²²²Ra and then due to ²³²Th, and contribution of ²²⁶Ra to the total dose rates received by the reference biota from natural radionuclides range from 85.6 to 99.9%. As can be seen in Fig. 4, the highest total dose rate (derived from both ²²⁶Ra and ²²⁶Th) was obtained for lichens and bryophytes $(5.54 \times 10^{+0} \,\mu\text{Gy h}^{-1})$, but it is still below the screening level of 10 mGy h⁻¹ (Brown et al. 2008; ERICA 2007). Applying ERICA tool, Hosseini et al. (2011) found that lichen and bryophytes were most exposed organisms, but indicated they are not necessarily the most at risk bearing in mind that the mosses and lichens are least sensitive to radiation exposure comparing to other terrestrial animals and terrestrial plants. For all terrestrial reference organisms, the ²²⁶Ra exhibited higher contribution to the total, internal and external dose rates (Fig. 4, Table 5), which confirms the findings of previous studies (Černe et al. 2012; Oughton et al. 2013). The values of total dose rates obtained in this study were much lower than those found for terrestrial organisms from the uranium mine area Žirovski vrh in Slovenia (Černe et al. 2012), and this may be explained by much higher ²³⁸U, ²²⁶Ra and ²³⁰Th specific activities in area affected by uranium mining.



Fig. 2 The histograms and probability plots of gamma dose rates due to ²²⁶Ra, ²³²Th and ⁴⁰K in soils from the territory of Belgrade

Although there was no intention to analyze artificial radionuclides in this study, only for comparison reasons, the dose rates from ²²⁶Ra and ²³²Th were compared to those obtained by applying ERICA tool to ¹³⁷Cs data in soil of the same area published in our previous paper (Petrović et al. 2013). According to results presented in Table 5, for ¹³⁷Cs the internal dose rates were higher for grasses and herbs $(2.81 \times 10^{-2} \ \mu\text{Gy} \ h^{-1})$, lichen and bryophytes $(8.04 \times 10^{-2} \ \mu\text{Gy} \ h^{-1})$, large mammal $(2.09 \times 10^{-1} \ \mu\text{Gy} \ h^{-1})$, small-burrowing mammal $(1.04 \times 10^{-1} \ \mu\text{Gy} \ h^{-1})$ and shrub $(4.94 \times 10^{-2} \ \mu\text{Gy} \ h^{-1})$ in

comparison with external dose rates. The contribution of the dose rate by ¹³⁷Cs to the total dose rate per organism ranged from 2% for lichen and bryophytes to 40% for large mammal (Fig. 4, Table 5). In study conducted by Sotiropoulou et al. (2016), the contribution of natural radionuclides to the total dose rate was higher in comparison with the contribution from the artificial radionuclides. External, internal and total absorbed dose rates assessed from ²²⁶Ra, ²³²Th and ¹³⁷Cs obtained in this study were found to be similar to those obtained for terrestrial biota in the area around coal-fired power plant 'Nikola Tesla'





in Serbia (Ćujić and Dragović 2017), as a consequence of similar value of radionuclides specific activities in soil used as input data in these assessments.

Conclusions

According to results of the Pearson correlation analysis, sand, silt and clay fractions, carbonate content, cation exchange capacity and soil pH were found to be correlated with natural radionuclides in analyzed soil. The assessment of radiation exposure to human due to analyzed radionuclides showed that the mean value of external gamma dose rate was close to the Serbian average, with the highest contribution of ²³²Th. The dose rates for the terrestrial biota calculated by ERICA tool did not exceed the screening dose rate of 10 μ Gy h⁻¹. The ²²⁶Ra shows higher contribution to the total, internal and external dose rates for all terrestrial reference organisms compared to the ²³²Th. To our knowledge, this study is the first to investigate the radiation exposure of non-human biota of Belgrade applying ERICA tool.

µGy h ⁻¹	Amphibian	Annelid	Arthro- pod—detri- tivorous	Bird	Flying insects	Grasses and herbs	Lichen and bryophytes	Mammal— large	Mam- mal—small burrowing	Mollusk— gastropod	Reptile	Shrub	Tree
External dose rate								5					
²²² Th	4.54×10^{-4} 8 12 × 10^{-6}	4.59×10^{-4} 8.12×10^{-6}	4.64×10^{-4} 8 70 × 10^{-6}	1.73×10^{-4}	1.79×10 ⁻² 25×10 ⁻⁶	1.68×10 ⁻² 6 38×10 ⁻⁶	1.76×10^{-4} 2.54×10^{-6}	9.18×10^{-3} 7.54×10^{-7}	4.34×10 ⁻² 6.96×10 ⁻⁶	1.79×10^{-4}	4.18×10^{-2} 6.38×10^{-6}	1.63×10^{-4} 2.90×10^{-6}	1.38×10 ⁻² 1 22×10 ⁻⁶
$^{137}Cs^a$	5.40×10^{-2}	5.40×10^{-2}	5.58×10^{-2}	1.98×10^{-2}	2.16×10^{-2}	1.98×10^{-2}	2.08×10^{-2}	1.01×10^{-2}	5.04×10^{-2}	2.16×10^{-2}	4.86×10^{-2}	1.98×10^{-2}	1.62×10^{-2}
Internal dose rate													
²²⁶ Ra	3.05×10^{-1}	3.01×10^{-1}	3.01×10^{-1}	2.64×10^{-1}	3.01×10^{-1}	$1.25 \times 10^{+0}$	$5.01 \times 10^{+0}$	3.14×10^{-1}	3.05×10^{-1}	3.32×10^{-1}	3.05×10^{-1}	$2.29 \times 10^{+0}$	8.29×10^{-2}
232 Th	5.19×10^{-4}	1.22×10^{-2}	6.77×10^{-3}	5.19×10^{-4}	6.77×10^{-3}	2.13×10^{-1}	5.08×10^{-1}	1.81×10^{-4}	1.81×10^{-4}	1.22×10^{-2}	2.90×10^{-3}	8.14×10^{-2}	1.69×10^{-3}
$^{137}Cs^a$	1.23×10^{-2}	2.04×10^{-3}	2.28×10^{-3}	1.93×10^{-2}	2.67×10^{-3}	2.81×10^{-2}	8.04×10^{-2}	2.09×10^{-1}	1.04×10^{-1}	1.02×10^{-3}	1.76×10^{-2}	4.94×10^{-2}	7.81×10^{-3}
Total dose													
rate													
²²⁶ Ra	3.50×10^{-1}	3.47×10^{-1}	3.47×10^{-1}	2.81×10^{-1}	3.19×10^{-1}	$1.27 \times 10^{+0}$	$5.03 \times 10^{+0}$	3.24×10^{-1}	3.49×10^{-1}	3.49×10^{-1}	3.47×10^{-1}	$2.31 \times 10^{+0}$	9.67×10^{-2}
232 Th	5.27×10^{-4}	1.22×10^{-2}	6.78×10^{-3}	5.21×10^{-4}	6.77×10^{-3}	2.13×10^{-1}	5.08×10^{-1}	1.82×10^{-4}	1.88×10^{-4}	1.22×10^{-2}	2.90×10^{-3}	8.14×10^{-2}	1.69×10^{-3}
$^{137}Cs^a$	6.63×10^{-2}	5.60×10^{-2}	5.81×10^{-2}	3.91×10^{-2}	2.43×10^{-2}	4.79×10^{-2}	1.01×10^{-1}	2.19×10^{-1}	1.55×10^{-1}	2.26×10^{-2}	6.62×10^{-2}	6.92×10^{-2}	2.40×10^{-2}
^a The dose ra	tes were calcul	lated from ¹³⁷	⁷ Cs data in soi.	l of the same a	area published	in Petrović et a	al. (2013)						

by ERICA tool	Flying
calculated	Bird
ence organisms	Arthro-
strial refere	Annelid
lose rates to terre	Amphibian
Table 5 D	µGy h ⁻¹

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Fig. 4 Contribution of each radionuclide to the total dose rate per organism calculated by ERICA tool

Acknowledgements This work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia (Project No. III43009).

References

- Abdalhamid S, Salih I, Idriss H (2017) Gamma absorbed radiation dose in Marrah mountain series, western Sudan. Environ Earth Sci 76:672. https://doi.org/10.1007/s12665-017-7009-7
- Antovic NM, Svrkota N, Antovic I (2012) Radiological impacts of natural radioactivity from soil in Montenegro. Radiat Prot Dosim 148:310–317
- Baykara O, Doğru M (2009) Determination of terrestrial gamma, ²³⁸U, ²³²Th and ⁴⁰K in soil along fracture zones. Radiat Meas 44:116–121
- Belivermis M, Kiliç Ö, Çotuk Y, Topcuoğlu S (2010) The effects of physicochemical properties on gamma emitting natural radionuclide levels in the soil profile of Istanbul. Environ Monit Assess 163:15–26
- Beresford NA, Barnett CL, Brown JE et al (2008a) Inter-comparison of models to estimate radionuclide activity concentrations in nonhuman biota. Radiat Environ Biophys 47:491–514
- Beresford NA, Barnett CL, Howard BJ, Scott WA, Brown JE, Copplestone D (2008b) Derivation of transfer parameters for use within the ERICA Tool and the default concentration ratios for terrestrial biota. J Environ Radioact 99:1393–1407
- Blanco Rodríguez P, Vera Tomé F, Lozano JC, Pérez-Fernández MA (2008) Influence of soil texture on the distribution and availability of ²³⁸U, ²³⁰Th, and ²²⁶Ra in soils. J Environ Radioact 99:1247–1254
- Boggs S Jr, Livermore D, Seltz MG (1985) Humic substances in natural waters and their complexation with trace metals and radionuclides: a review. ANL-84-78, Argonne National Laboratory, Argonne, Illinois

- Botwe BO, Schirone A, Delbono I, Barsanti M, Delfanti R, Kelderman P, Nyarko E, Lens PNL (2017) Radioactivity concentrations and their radiological significance in sediments of the Tema Harbour (Greater Accra, Ghana). J Radiat Res Appl Sci 10:63–71
- Bouhila G, Azbouche A, Benrachi F, Belamri M (2017) Natural radioactivity levels and evaluation of radiological hazards from Beni Haroun dam sediment samples, northeast Algeria. Environ Earth Sci 76:710. https://doi.org/10.1007/s12665-017-7061-3
- Brown JE, Alfonso B, Avila R, Beresford NA, Copplestone D, Pröhl G, Ulanovsky A (2008) The ERICA Tool. J Environ Radioact 99:1371–1383
- Brown JE, Alfonso B, Avila R, Beresford NA, Copplestone D, Hosseini A (2016) A new version of the ERICA tool to facilitate impact assessments of radioactivity on wild plants and animals. J Environ Radioact 153:141–148
- Caridi F, D'Agostino M, Marguccio S, Belvedere A, Belmusto G, Marcianò G, Sabatino G, Mottese A (2016) Radioactivity, granulometric and elemental analysis of river sediments samples from the coast of Calabria, south of Italy. Eur Phys J Plus 131:136. https:// doi.org/10.1140/epjp/i2016-16136-1
- Černe M, Smodiš B, Štrok M, Lj Benedik (2012) Radiation impact assessment on wildlife from an uranium mine area. Nucl Eng Des 246:203–209
- Chandrasekaran A, Rajalakshmi A, Ravisankar R, Vijayagopal P, Venkatraman B (2015) Measurements of natural gamma radiations and effects of physico-chemical properties in soils of Yelagiri hills, Tamilnadu India with statistical approach. Proc Earth Planet Sci 11:531–538
- Ćujić M, Dragović S (2017) Assessment of dose rate to terrestrial biota in the area around coal fired power plant applying ERICA tool and RESRAD BIOTA code. J Environ Radioact. https://doi. org/10.1016/J.JENVRAD.2017.09.014
- Ćujić M, Dragović S, Đorđević M, Dragović R, Gajić B, Miljanić Š (2015) Radionuclides in the soil around the largest coal-fired power plant in Serbia: radiological hazard, relationship with soil characteristics and spatial distribution. Environ Sci Pollut Res 22:10317–10330
- Doering C, Bollhöfer A (2016) A soil radiological quality guideline value for wildlife-based protection in uranium mine rehabilitation. J Environ Radioact 151:522–529
- Dragović R, Kićović DM (2001) Changes of climate of Belgrade caused by urban and industrial factors. In: Environmental protection of urban and suburban settlements, eco-conference Novi Sad, pp 369–374
- Dragović S, Onjia A (2006) Classification of soil samples according to their geographic origin using gamma-ray spectrometry and principal component analysis. J Environ Radioact 89:150–158
- Dragović S, Lj Janković, Onjia A (2006) Assessment of gamma dose rates from terrestrial exposure in Serbia and Montenegro. Radiat Prot Dosim 121:297–302
- Dragović S, Gajić B, Dragović R, Janković-Mandić LJ, Slavković-Beškoski L, Mihailović N, Momčilović M, Ćujić M (2012a) Edaphic factors affecting the vertical distribution of radionuclides in the different soil types of Belgrade, Serbia. J Environ Monit 14:127–137
- Dragović S, Janković-Mandić LJ, Dragović R, Đorđević M (2012b) Natural and man-made radionuclides in soil as sources of radiation exposure. In: Balenovic D, Stimac E (eds) Radiation exposure: sources, impacts and reduction strategies. Nova Science Publishers Inc., New York, pp 1–42
- Dragović S, Janković-Mandić LJ, Dragović R, Đordević M, Đokić M, Kovačević J (2014) Lithogenic radionuclides in surface soils of Serbia: Spatial distribution and relation to geological formations. J Geochem Explor 142:4–10

Đurdić S, Stojković S, Šabić D (2011) Nature conservation in urban conditions: a case study from Belgrade, Serbia. Maejo Int J Sci Technol 5:129–145

- Elejalde C, Herranz M, Romero F, Legarda F (1996) Correlations between soil parameters and radionuclide contents in samples from Biscay (Spain). Water Air Soil Pollut 89:23–31
- ERICA (2007) The ERICA assessment tool, version 1.2.1. http:// www.erica-tool.com/. Accessed 23 Oct 2017
- Federal Geological Survey (1970) Geologic map of SFRY, 1:500000. Belgrade, Serbia
- Guo P, Duan T, Song X, Xu J, Chen H (2008) Effects of soil pH and organic matter on distribution of thorium fractions in soil contaminated by rare-earth industries. Talanta 77:624–627
- Hosseini A, Brown JE, Szymanska M, Ciupek K (2011) Application of an environmental impact assessment methodology for areas exhibiting enhanced levels of NORM in Norway and Poland. Radioprotection 46:S759–S764
- Huy NQ, Hien PD, Luyen TV, Hoang DV, Hiep HT, Quang NH, Long NQ, Nhan DD, Binh NT, Hai PS, Ngo NT (2012) Natural radioactivity and external dose assessment of surface soils in Vietnam. Radiat Prot Dosim 151:522–531
- IAEA (2004) Soil sampling for environmental contaminants. IAEA-TECDOC-1415. International Atomic Energy Agency, Vienna
- IAEA (2012) Environmental modelling for radiation safety (EMRAS)—a summary report of the results of the EMRAS programme (2003–2007). IAEA-TECDOC-1678. International Atomic Energy Agency, Vienna
- IAEA (2014) Handbook of parameter values for the prediction of radionuclide transfer to wildlife. Technical reports series no. 479. International Atomic Energy Agency, Vienna
- ICRP (2007) The 2007 recommendations of the international commission on radiological protection. ICRP publication 103. Ann. ICRP 37. Oxford
- ISO 10390 (2005) Soil quality-determination of pH. International Standard Organization, Geneva
- ISO 10693 (1995) Soil quality-determination of carbonate contentvolumetric method. International Standard Organization, Geneva
- ISO 11265 (1994) Soil quality-determination of the specific electrical conductivity. International Standard Organization, Geneva
- Janković Mandić LJ, Dragović S (2010) Assessment of terrestrial gamma exposure to the population of Belgrade (Serbia). Radiat Prot Dosimetry 140:369–377
- Janković Mandić LJ, Dragović R, Dragović S (2010) Distribution of lithogenic radionuclides in soils of the Belgrade region (Serbia). J Geochem Explor 105:43–49
- Janković Mandić LJ, Dragović R, Pisanjuk S, Dragović S (2016) The natural radionuclides in soils of Subotica (Serbia): distribution and corresponding gamma dose rates. RAD Conf Proc 1:71–74
- Janković-Mandić LJ, Dragović R, Đordjević M, Đolić M, Onjia A, Dragović S, Bačić G (2014) Spatial variability of ¹³⁷Cs in the soil of Belgrade region (Serbia). Hem Ind 68:449–455
- Kappen H (1929) Die Bodenaziditaat. Springer, Berlin
- Komatina M, Komatina SM (1999) In: Chilton J (ed) Groundwater in the urban environment. Selected city profiles, Balkema, Rotterdam, pp 317–322
- Kottek M, Grieser J, Beck C, Rudolf B, Rubel F (2006) World Map of the Köppen–Geiger climate classification update. Meteorol Z 15:259–263
- Kumar A, Rout S, Ghosh M, Singhal RK, Ravi PM (2013) Thermodynamic parameters of U(VI) sorption onto soils in aquatic systems. SpringerPlus 2(530):1–7
- Larsson CM (2008) An overview of the ERICA Integrated Approach to the assessment and management of environmental risks from ionising contaminants. J Environ Radioact 99:1364–1370
- Li J, Liu S, Zhang Y, Chen L, Yan Y, Cheng W, Lou H, Zhang Y (2015) Pre-assessment of dose rates of ¹³⁴Cs, ¹³⁷Cs, and ⁶⁰Co for marine

biota from discharge of Haiyang Nuclear Power Plant, China. J Environ Radioact 147:8–13

- Mazeika J, Marciulioniene D, Nedveckaite T, Jefanova O (2016) The assessment of ionising radiation impact on the cooling pond freshwater ecosystem non-human biota from the Ignalina NPP operation beginning to shut down and initial decommissioning. J Environ Radioact 151:28–37
- Milenkovic B, Stajic JM, Lj Gulan, Zeremski T, Nikezic D (2015) Radioactivity levels and heavy metals in the urban soil of Central Serbia. Environ Sci Pollut Res 22:16732–16741
- Navas A, Soto J, Machín J (2002) ²³⁸U, ²²⁶Ra, ²¹⁰Pb, ²³²Th and ⁴⁰K activities in soil profiles of the Flysch sector (Central Spanish Pyrenees). Appl Radiat Isot 57:579–589
- Navas A, Gaspar L, López-Vicente M, Machín J (2011) Spatial distribution of natural and artificial radionuclides at the catchment scale (South Central Pyrenees). Radiat Meas 46:261–269
- Nedveckaite T, Filistovic V, Marciulioniene D, Prokoptchuk N, Plukiene R, Gudelis A, Remeikis V, Yankovich T, Beresford NA (2011) Background and anthropogenic radionuclide derived dose rates to freshwater ecosystem—nuclear power plant cooling pond—reference organisms. J Environ Radioact 102:788–795
- ORTEC (2001) Gamma vision 32, gamma-ray spectrum analysis and MCA emulation. ORTEC, Oak Ridge, version 5.3
- Oughton DH, Agüero A, Avila R, Brown JE, Copplestone D, Gilek M (2008) Addressing uncertainties in the ERICA Integrated Approach. J Environ Radioact 99:1384–1392
- Oughton DH, Strømman G, Salbu B (2013) Ecological risk assessment of Central Asian mining sites: application of the ERICA assessment tool. J Environ Radioact 123:90–98
- Petrović J, Ćujić M, Đorđević M, Dragović R, Gajić B, Miljanić Š, Dragović S (2013) Spatial distribution and vertical migration of ¹³⁷Cs in soils of Belgrade (Serbia) 25 years after the Chernobyl accident. Environ Sci Process Impacts 15:1279–1289
- Rowell DL (1997) Bodenkunde. Untersuchungsmethoden und ihre Anwendungen. Springer, Berlin
- Šegota T (1988) Climatology for geographers. Školska knjiga, Zagreb (in Croatian)
- Simakov VN (1957) Application of phenylanthranilic acid in determining humus, the method of Tyurin. Почвоведение 8:72–73
- Sotiropoulou M, Florou H, Manolopoulou M (2016) Radioactivity measurements and dose rate calculations using ERICA tool in the terrestrial environment of Greece. Environ Sci Pollut Res 23:10872–10882
- SPSS (2007) Statistical package for the social sciences 16.0. Chicago, Illinois
- Taskin H, Karavus M, Ay P, Topuzoglu A, Hidiroglu S, Karahan G (2009) Radionuclide concentrations in soil and lifetime cancer risk due to gamma radioactivity in Kirklareli, Turkey. J Environ Radioact 100:49–53
- Tomić ZP, Djordjević AR, Rajković MB, Vukašinović I, Nikolić NS, Pavlović V, Lačnjevac ČM (2011) Impact of mineral composition on the distribution of natural radionuclides in rigosol-anthrosol. Sens Transducers J 125:115–130
- UNSCEAR (2000) Sources and effects of ionizing radiation. UNSCEAR 2000 report to the general assembly with scientific annexes, vol 1: sources. United Nation, New York
- UNSCEAR (2010) Sources and effects of ionizing radiation. UNSCEAR 2008 report to the general assembly with scientific annexes, vol I. United Nation, New York
- UNSCEAR (2011) Sources and effects of ionizing radiation. UNSCEAR 2008 report to the general assembly with scientific annexes, vol II scientific annexes C, D and E. United Nation, New York
- Vives i Batlle J, Balonov M, Beaugelin-Seiller K et al (2007) Intercomparison of absorbed dose rates for non-human biota. Radiat Environ Biophys 46:349–373

Vives i Batlle J, Beaugelin-Seiller K, Beresford NA et al (2011) The estimation of absorbed dose rates for non-human biota: an extended intercomparison. Radiat Environ Biophys 50:231–251

Walkley A, Black IA (1934) An examination of the Degtjareff method for determining soil organic matter and a proposed modification of the chromic acid titration method. Soil Sci 37:29–38 Wood MD, Marshall WA, Beresford NA, Jones SR, Howard BJ, Copplestone D, Leah RT (2008) Application of the ERICA Integrated Approach to the Drigg coastal sand dunes. J Environ Radioact 99:1484–1495