

RISK ASSESSMENT OF HUMAN EXPOSURE TO RADIONUCLIDES IN SOIL OF URBAN AREAS (PUBLIC PARKS AND OPEN PLAYGROUNDS) IN KRUŠEVAC, SERBIA

by

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This study aims to obtain data on terrestrial radioactivity coming from naturally occurring (⁴⁰K, ²²⁶Ra, ²³²Th, and ²³⁸U) and artificial (¹³⁷Cs) radionuclides in surface soil (0-10 cm) of selected eighteen public-access urban areas in Kruševac city, Serbia, and to assess the corresponding health effects for citizens using those areas for recreational purposes. The specific activities of investigated radionuclides were analyzed using HPGe gamma-ray spectrometry. The mean specific activity of ⁴⁰K, ²²⁶Ra, ²³²Th, ²³⁸U, and ¹³⁷Cs was found to be 353, 39.8, 38.9, 41.0, and 5.9, respectively, in compliance with their values in other parts of Serbia and neighboring countries reported in similar researches. To evaluate the human health risk associated with radionuclides, conservative exposure assumptions and models recommended by the United States Environmental Protection Agency were employed taking into account three exposure routes: ingestion, inhalation of soil, and external irradiation. The absorbed gamma dose rate in the air due to natural radionuclides in soil was calculated. The calculated indices suggested that the radiation risk arising from natural and artificial radionuclides was not significant. The total excess lifetime cancer based on the 95 % upper confidence limit of the specific activities mean was calculated to be $5.89 \cdot 10^{-6}$, lower than the tolerable risk for regulatory purposes (10^{-4}). Among investigated radiation exposure pathways, external exposure was the most contributing one for the health risk. The results obtained for the city's parks and playgrounds suggested their safe use for recreational purposes from the radioecological point of view.

Key words: public green area, urban soil, radionuclide, radiological risk, human health, gamma-spectrometry

INTRODUCTION

All living beings on Earth are continuously and unavoidably exposed to ionizing radiation. About 85 % of exposure is coming from natural radiation whose main contributors are terrestrial radionuclide ⁴⁰K and radionuclides of the ²³⁸U and ²³²Th series. The natural radioactivity of soils is inhomogeneously distributed and depends on the local geology, pedology, and geographical factors [1]. Man-made radionuclides have been globally released in form of fallouts to the environment after nuclear weapon testing and nuclear reactor accidents. Radioisotopes discharged from the anthropogenic sources after deposition were retained in environmental compartments including soil. The

¹³⁷Cs has been considered as one of the biologically the most important fission products carrying major potential hazard to human and non-human biota due to its relatively long half-life (30.2 years), a strong affinity for binding to the soil, and physicochemical and metabolic properties analogous to potassium [2].

Urban environmental quality is closely related to the human health and wellbeing of city residents, hence investigated soil, as a significant component of an urban environment, is of vital importance. Public parks and public-access playgrounds are specially designated green areas within cities to provide opportunities for recreation, sport, and relaxation. Citizens may spend much of their free time there, especially parents with children, sportsmen and older generation people. The soil surfaces of parks and playgrounds due to their large, exposed areas act both as a source

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and a sink for many types of pollutants, among them radionuclides. Besides being externally exposed to ionizing radiation from terrestrial radionuclides, park users may get easily into direct contact with the surface soil due to outdoor activities, and under those circumstances, radionuclides from soil can reach the human body through oral and inhalation pathways. Oral exposure is crucial for children who may ingest significant quantities of soil by hand-to-mouth activities. Moreover, soil can be easily resuspended by the wind or human movements (*e. g.*, walking, running), and generated airborne particles or dust can be readily inhaled [3]. Therefore, it is of great importance to affirm whether urban green areas are safe for the population from a health point of view.

The health risks posed by radionuclides can be quantitatively determined considering different habits of target receptors and comprehension of the relevant exposure scenarios. Most frequently used methods for radiological risk assessment due to radionuclides in soils have been based on various indices related mainly to external exposure to gamma radiation depending on their specific activities in the topsoil. Considering the high radiotoxicity and chemical toxicity of radionuclides, determination of the attendant health risk requires particular attention and the application of the modern methodology. That entails a multi-step procedure based on statistics compilation and evaluation, human exposure assessment, toxicological data, and risk characterization. One of the best-developed methodologies is the health risk assessment model for various exposure and site-specific scenarios proposed by the United States Environmental Protection Agency (US EPA) that has been used by environmental professionals and researchers worldwide. This model has been proved as a powerful tool for assessing human health risk and identifying the exposure pathways of most concern due to different chemicals, mostly heavy metals, persistent organic pollutants, polycyclic aromatic hydrocarbons, *etc.*, present in diverse environmental media [4]. Although the methodology includes radionuclide risk assessment tools, it has been rarely used to characterize human health risks resulting from a potential exposure to radionuclides via the exposure pathways determined appropriate for the source area [5, 6].

The increasing concern about the safety of soils in urban parks and playgrounds resulted in numerous studies carried out in many cities all around the world over the past years. Much concern has been addressed over the problem of soil contamination with heavy metals and organic pollutants due to rapid industrialization and urbanization [3, 7-10]. At the same time, various radioecological studies performed in Serbia have been aimed mainly to a determination of the content of radionuclides in urban soil of different land uses, not exclusively public green areas, estimating only risks and health hazards from the external radiation exposure of the population without taking into ac-

count specific exposure scenarios [2, 4, 11-18]. The literature review has shown that there are no data on the application of US EPA health risk assessment methodology due to radiation exposure of humans in Serbia. Even though Kruševac is one of the largest cities in Serbia, studies evaluating radionuclide content in urban soils and accompanying potential health risks have not been undertaken in this area up to the present time.

Owing to the significance of potential exposure to radionuclides and terrestrial radiation in urban areas, the present study was conducted with the following objectives: to determine the specific activity of natural (^{226}Ra , ^{238}U , ^{232}Th , and ^{40}K) and artificial (^{137}Cs) radionuclides in surface soil samples taken from designated urban green areas in Kruševac and to estimate potential health risk due to radionuclides according to the US EPA methodology for citizens and visitors (who use parks and playgrounds for recreational purposes only) due to exposure to the soil via different exposure pathways.

MATERIALS AND METHODS

Study area

Kruševac (N 43° 34' 59.6", E 21° 19' 29.0") is the administrative center of Rasina District and represents one of the largest urban areas in the central part of Serbia, extending to 75.4 km², with a total population of around 80000 inhabitants [19]. The climate is continental, with relatively cold winters and moderately hot summers. The coldest month is January (0.2 °C), and the hottest is July (21.8 °C), with an annual mean temperature of 11.4 °C. The average annual precipitation is relatively low (628.1 mm). The average wind speed is 1.4 ms⁻¹. Prevailing winds blow from the south (7.6%) and the east (7.0%), while calm occurs 48.1% of the time [20].

The altitude of the city is between 150 and 200 m a.s.l. The most important watercourses are the West Morava River and the Rasina River. The wider region of Kruševac was formed of rocks of Serbian-Macedonian Massif. Paleozoic rocks – crystalline schists, divided by multiple faults comprise this tectonic unit, forming mountainous horsts or depressions like West Morava's trench, where tertiary lake sediments are positioned over Paleozoic rock. These lake sediments are covered by alluvial deposits and river terraces that are especially well-formed. The most common soil types (WRB classification) represented in the study area are Haplic Fluvisol, Haplic Cambisol (Eutric), Podzol, Haplic Vertisol, Haplic Vertisol (Eutric) – Regic Anthrosol (Eutric, Clayic), Albic Luvisol (Endoeutric), and Colluvic Regosol (Stagnic). Nonetheless, urban soils are markedly different from the natural ones, and highly spatially variable due to diverse human activi-

ties. They are known to have peculiar characteristics such as unpredictable layering, degradation, altered soil reaction, low organic matter content, limited aeration, and water drainage, high content of foreign materials, and typically contain higher amounts of contaminants and trace elements than those from rural origins due to the higher density of anthropogenic activity [21].

Kruševac is considered a highly industrialized city especially in sectors of chemical, machinery, woodworking, and food industry. The industry related to Technologically Enhanced Naturally Occurring Radioactive Materials does not exist in the study area. The region where the study was conducted is both a historically rich region and one of the most active in terms of tourism. The surface of public green areas per capita is 11 m², but the General Urban Plan of Kruševac envisions the extension of those areas up to 65 m² until 2025. Convenient weather settings alongside the inhabitants' behavior governed by their socio-economic factors favor spending time outdoor for both children and adults.

Soil sampling

Eighteen public urban green areas designated as parks and areas for sports and recreation most frequently visited by residents within the Kruševac city area were selected for the sampling campaign carried out in July 2018 (fig. 1, tab. 1). The selection of green areas was carried out using digital orthophoto images of 10 cm resolution provided by the Republic Geodetic Authority coupled with the authors' experience. Those areas (representing more than 80 % of the green areas in Kruševac) were chosen based on the following criteria: they are open city areas, and relatively evenly distributed within city boundaries. Among selected locations, some parks are major touristic sites, particularly the archaeological park *Prince Lazar's Town*, *Pioneers Park*, *Bagdala Park*, and memorial complex *Slobodište*. Other sites are located in squares,

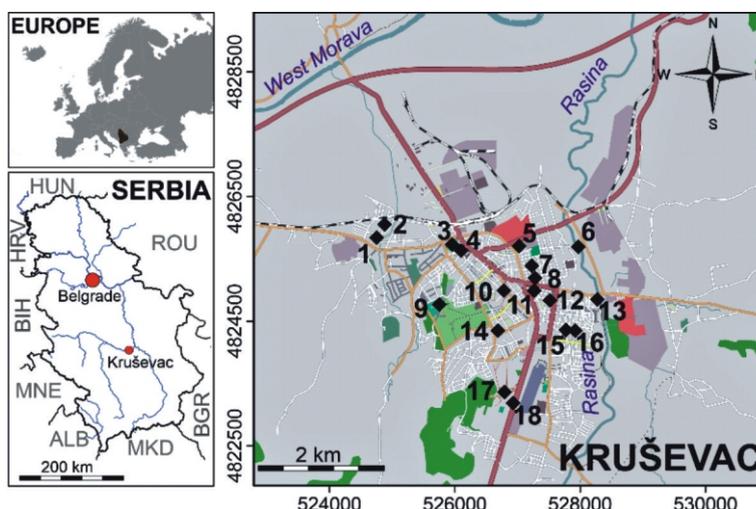
near schools, kindergartens, and day-care centers, or represent smaller neighborhood parks.

Each sampling site was georeferenced by a hand-held GPS device with a precision of 1 m. The surface layer covered by vegetation and debris was removed before soil sampling. The samples of 30 cm × 30 cm were taken from the topsoil to a depth of 10 cm [22]. To ensure the representativeness of the specimen, five subsamples (center and vertices of 2 m × 2 m rectangle) were taken from each location and thoroughly mixed to obtain a bulk composite sample. This sampling strategy was adopted to reduce the possibility of random influences from urban waste. Approximately, the weight of each subsample

Table 1. Brief description of the sampling sites

Location	Street	Land use
1	Drinska, Miletine bune	Public Park (Šljivak Park)
2	Luja Pastera, Železnička	Sports ground
3	Lazarev grad	Archaeological Park (Prince Lazar's Town)
4	Trg Despota Stevana	Public Park (Despot Stefan's Square)
5	Balkanska, Dušanova	Public Park (Pioneers Park)
6	Živorada Stankovića	Children playground
7	Trg slobode	Public Park (Freedom Square)
8	Svetog Save, Iločki trg	Public Park
9	Bagdala	Public Park (Bagdala)
10	Trg Kosturnica	Public Park (Kosturnica Square)
11	Jovana Dučića	Public Park
12	Kraljevića Marka	Children playground
13	Raševačka	Public Park (Rasina's promenade)
14	Dostojevskog	Children playground
15	Varšavska	Children playground
16	Petra Kočića	Sports ground
17	Slobodište	Memorial complex
18	Bruski put	Children playground

Figure 1. Simplified map of the study area with the position of sampling locations



involved in composite was 1 kg of soil. All samples were enclosed in a plastic bag and transported to the laboratory where the stones, visible roots, and other foreign material were removed from the soil samples and air-dried at room temperature for two weeks. For radionuclide analysis samples were brought to constant mass by drying in the oven at 105 °C. After that, samples were homogenized by mechanical mixing and were brought to uniform spread by sieving through a 2 mm mesh sieve. Part of the total sample for analysis was made by the quarter method. Samples were placed into 500 cm³ Marinelli beakers and sealed for 40 days before analysis to attain a secular equilibrium between ²²⁶Ra and its daughters. Due to topsoil layer investigations, this study does not deal with soil taxonomy.

Radionuclide analysis

The specific activities of ⁴⁰K, ²³⁸U, ²²⁶Ra, and ²³²Th were determined using a coaxial HPGe detector (ORTEC-AMETEK, GEM25P4-70) with an energy resolution of 1.67 keV at the 1.33 MeV of ⁶⁰Co and the relative efficiency of 28 %. The detector was housed in a 10 cm thick lead shield lined by a 0.5 cm copper layer. Energy and efficiency calibrations were done using standard reference material Source No. LR 320 certified by the Calibration laboratory for radioactivity measurement (Deutscher Kalibrierdienst, Braunschweig, Germany). The counting time for each sample was 100 ks, with an analytical precision of 10 %. The specific activity of ²³⁸U was calculated using gamma-rays of ²³⁴Th at 63.3 keV, 92.4 keV, and 92.8 keV. The specific activity of ²²⁶Ra was determined from the gamma-rays at 609.3, 1120.3, and 1764.5 keV of ²¹⁴Bi and 295.2 and 351.9 keV of ²¹⁴Pb. Gamma-rays with energies 911.2 and 969.0 keV (²²⁸Ac) and 238.6 keV (²¹²Pb) were used for the calculation of the specific activities of ²³²Th. Specific activities of ⁴⁰K and ¹³⁷Cs were determined using their own 1460.8 keV and 661.7 keV gamma-rays, respectively. The measured specific activity of ¹³⁷Cs was decay corrected to the sampling date. The output signal was processed by a multichannel analyzer 92x-II Spectrum Master, and the obtained spectra were analyzed by Gamma Vision 32 software (version 5.3).

Risk assessment and exposure parameters

Human health risk assessment is a tiered approach based on the source-pathway-receptor concept where both quantitative and qualitative assessments of potential health risks are made depending on site-specific characteristics. For this study, the receptor identified for health risk assessment due to radionuclides in soil was a recreator. The scenario assumes temporary occupancy by an individual, a recreator, who uses se-

lected areas for recreational purposes, relaxation, playing, and tourism. The following exposure routes were considered: incidental ingestion of soil, inhalation of particulates emitted from soil, and external exposure to ionizing radiation [23]. Environmental decisions concerning risk assessment are made based on mean concentrations of the contaminants of potential concern. Since the potential receptors are exposed to direct contact with topsoil, the input parameter was the specific activity of each radionuclide investigated in the surface soil. The potential risk for children and adults was estimated for exposures to the 95 % upper confidence level ($UCL_{0.95}$) of the arithmetic mean of specific activities of radionuclides as a health-protective value and a measure of reasonable maximum exposure of the receptor. A $UCL_{0.95}$ represents that value so that one can be 95 % confident that the population mean cannot exceed that limit.

US EPA risk assessment model, which is fully accessible through the Risk Assessment Information System (RAIS) compilation [23] was exploited for risk calculation. Based on US EPA classification, all radionuclides belong to the Group A – human carcinogens [24]. The initial calculation of excess lifetime cancer risk ($ELCR$) as a result of exposure to radionuclides is given by eq. (1)

$$ELCR = TI \cdot SF \quad (1)$$

where $ELCR$ represents the unitless probability of an individual developing cancer over a lifetime, TI is a total intake of radionuclides, SF is a cancer slope factor. The total cancer risk, $ELCR_{total}$, for simultaneous exposure to all radionuclides investigated was calculated as a sum of all $ELCR$ for all radionuclides across all exposure routes considered.

Slope factors for exposure to radionuclides are age-averaged, lifetime attributable radiation cancer incidence risk per unit activity inhaled or ingested (internal exposure), or per unit time-integrated specific activity in soil (external exposure). Their values were obtained from relevant US EPA guidance [24]. Slope factors for ²²⁶Ra, ²³²Th, ²³⁸U, and ¹³⁷Cs were chosen to account for short-lived decay products and include the parent radionuclide, as well as ingrowth of daughter isotopes out to 100 years (+P designation in tab. 3), assuming secular equilibrium with the parent nuclide [5].

Total intakes of radionuclides present in the soil for ingestion TI_{ing} (Bq), inhalation TI_{inh} (Bq), and external exposure to ionizing radiation TI_{ex} (Bqkg⁻¹ per year) were calculated according to the eqs. (2)-(4).

$$TI_{ing} = A_s (RN) \cdot IFS_{adj} \cdot F_{kg/mg} \cdot (1 - e^{-\lambda t}) \cdot t^{-1} \cdot \lambda^{-1} \\ IFS_{adj} = EF \cdot ED_{child} \cdot IRS_{child} + EF \cdot ED_{adult} \cdot IRS_{adult} \quad (2)$$

$$TI_{inh} = A_s (RN) \cdot IFS_{adj} \cdot PEF^{-1} \cdot (1 - e^{-\lambda t}) \cdot t^{-1} \cdot \lambda^{-1} \\ IFS_{adj} = EF \cdot ED_{child} \cdot ET \cdot F_{d/h} \cdot IRA_{child} \\ EF \cdot (ED_{child} \cdot ED_{child}) \cdot ET \cdot F_{d/h} \cdot IRA_{adult} \quad (3)$$

$$TI_{ex} = \frac{A_s(RN) \cdot EF \cdot F_{a/d} \cdot ED_{adult} \cdot ET}{F_{d/h} \cdot GSF \cdot ACF \cdot (1 - e^{-\lambda t}) \cdot t \cdot \lambda^{-1}} \quad (4)$$

The Exposure Factors Handbook [25] was used as the main source for parameters for *TI* calculations. The meanings and values of specific and constant parameters used in the equations are given in tabs. 2 and 3. The total exposure time (*t*) of 26 years was defined as the mean age of the residents, taking into account mainly the changes in the future residence and living environment of residents of different ages.

Additionally, the level of radiological hazard has been estimated in terms of gamma absorbed dose rate in air, originating from radioactive sources in the soil. The absorbed gamma dose rate in air 1 m above the ground surface for the uniform distribution of the naturally occurring radionuclides was computed based on guidelines provided by UNSCEAR [1]

$$\dot{D} = 0.462A_s(^{238}\text{U}) + 0.604A_s(^{232}\text{Th}) + 0.0417A_s(^{40}\text{K}) \quad (5)$$

where A_s [Bqkg⁻¹] – the specific activity of respected radionuclide in soil.

The average contribution to the absorbed dose rate from the artificial radionuclide ¹³⁷Cs is minor compared to the contributions from the natural radionuclides, and it

was omitted from further calculations. In this study, the absorbed gamma-dose rate has been calculated to compare obtained results with other relevant investigations due to a lack of published data obtained using US EPA methodology.

Statistical analysis and calculation of upper confidence limit

Descriptive statistics were determined for each dataset using the IBM SPSS Statistics 25 software. Spatial distribution and risk values in the selected locations were charted and visualized on maps produced by the Golden Software Surfer 12. The exploratory data analysis and the 95 % upper confidence level calculation were carried out using the ProUCL (version 5.1) software [26, 27]. This specific software computes decision statistics and calculates upper statistical limits by exploiting several parametric and nonparametric methods covering a wide range of data variability, distribution, skewness, and sample size.

The US EPA's guidance address three kinds of data distributions. These include normal, lognormal, and gamma distribution. For the normally distributed dataset, a $UCL_{0.95}$ on the mean based on the Student's *t*-statistic has typically been used, following eq. (6)

Table 2. Exposure parameters used for the health risk assessment

Parameter	Meaning (value)	Unit*
<i>ACF</i>	Area correction factor for 1000 m ² , radioisotope specific – ¹³⁷ Cs (0.764), ⁴⁰ K (0.832), ²²⁶ Ra (0.818), ²³² Th (0.817), ²³⁸ U (0.811)	Unitless
<i>ED</i>	Exposure duration for children (6) and adults (26)	a
<i>EF</i>	Exposure frequency (75)	da ⁻¹
<i>ET</i>	Exposure time (1)	hd ⁻¹
<i>F_{d/h}</i>	Correction factor (24 ⁻¹)	dh ⁻¹
<i>F_{kg/mg}</i>	Correction factor (10 ⁻⁶)	kgmg ⁻¹
<i>F_{a/d}</i>	Correction factor (365 ⁻¹)	ad ⁻¹
<i>GSF</i>	Gamma shielding factor (1)	Unitless
<i>IFA_{adj}</i>	Inhalation fraction – age adjusted (1473.5)	m ³
<i>IFS_{adj}</i>	Ingestion fraction – age adjusted (240000)	mg
<i>IRA</i>	Inhalation rate for children (10) and adults (20)	m ³ d ⁻¹
<i>IRS</i>	Ingestion rate of soil for children (200) and adults (100)	mg ^d -1
<i>PEF</i>	Particulate emission factor (site- specific, 3.12 10 ²⁹)	m ³ kg ⁻¹
λ	Radioactive decay constant (radioisotope-specific)	a ⁻¹
<i>t</i>	Time spent by recreator in the area (26)	a

*d – day, a – year

Table 3. Radionuclide carcinogenicity slope factors (*SF*) and calculated total intakes (*TI*) for different exposure routes (*P* – short-lived decay products)

Isotope	Associated decay chain (terminal radionuclide)	<i>SF_{ing}</i> [Bq ⁻¹]	<i>SF_{inh}</i> [Bq ⁻¹]	<i>SF_{ex}</i> [kga ⁻¹ Bq ⁻¹]	<i>TI_{ing}</i> [Bq]	<i>TI_{inh}</i> [Bq]	<i>TI_{ex}</i> [Bqakg ⁻¹]
¹³⁷ Cs + P	¹³⁷ mBa (¹³⁷ Ba)	1.15 10 ⁻⁹	3.04 10 ⁻⁹	6.85 10 ⁻⁸	1.7	3.26 10 ⁻²⁶	1.2
⁴⁰ K	–	1.58 10 ⁻⁹	6.00 10 ⁻⁹	2.16 10 ⁻⁸	133	2.56 10 ⁻²⁴	103
²²⁶ Ra + P	²²² Rn to ²¹⁰ Pb (²¹⁰ Pb)	1.83 10 ⁻⁸	7.63 10 ⁻⁷	2.26 10 ⁻⁷	10.0	1.93 10 ⁻²⁵	7.6
²³² Th + P	²²⁸ Ra, ²²⁸ Ac (²²⁸ Th)	5.86 10 ⁻⁸	2.35 10 ⁻⁶	1.09 10 ⁻⁷	9.9	1.90 10 ⁻²⁵	7.5
²³⁸ U + P	²³⁴ Th, ^{234m} Pa, ²³⁴ Pa (²³⁴ U)	5.32 10 ⁻⁹	6.40 10 ⁻⁷	3.22 10 ⁻⁹	10.7	2.05 10 ⁻²⁵	8.0

$$UCL_{0.95} = \bar{A}_s + t_{0.95, n-1} SD(A_s) n^{0.5} \quad (6)$$

where \bar{A}_s [Bqkg⁻¹] is the mean specific activity of a particular radionuclide, $SD(A_s)$ [Bqkg⁻¹] – the sample standard deviation, n – the sample size (18), and $t_{0.95, n-1}$ – the 0.95 quantile of the Student's t distribution with $n-1$ degrees of freedom ($t_{0.95, 17} = 1.740$) [27].

Most often, environmental datasets are characterized by positive skewness, and consequently, a lognormal distribution is applied to model those data and compute the various upper limits. For lognormal data, EPA recommends the Land method using the H -statistic [27]. Despite computational ease of lognormal distribution, in practice when the dataset is skewed and is burdened with outliers the H -statistic could yield unstable and unacceptably large UCL values. Typically, positively skewed datasets beside a lognormal follow a gamma distribution. The main advantage of a gamma distribution over lognormal is its tendency to result in reliable and stable $UCL_{0.95}$ values, providing the specified 95 % coverage of the population mean. For datasets of small size ($n < 50$) following a gamma distribution with shape parameter, $k > 1$, the $UCL_{0.95}$ should be computed applying an adjusted gamma method given by eq. (7)

$$UCL_{0.95} = 2n\hat{k}^* \bar{A}_s \chi_{2n\hat{k}^*}^2 \quad (7)$$

where \hat{k}^* is a bias-corrected estimate of shape parameter k , β is adjusted probability level, which is used to

achieve the 95 % confidence level (for $n = 18$, $\beta = 0.0357$), and $\chi_{2n\hat{k}^*}^2$ denotes cumulative percentage point of the chi-square with $2n\hat{k}^*$ degrees of freedom at β probability level [28].

The computation of UCL on the mean for distribution-free datasets, depending on the sample size and skewness, can be done by several proposed nonparametric methods. The US EPA suggests the use of the Chebyshev inequality method which should be appropriate for a variety of distributions as long as the skewness is not very large. Calculation of $UCL_{0.95}$ exploiting a one-sided version of this method based upon sample mean and sample standard deviation is given by eq. (8) [27].

$$UCL_{0.95} = \bar{A}_s (1/0.05)^{0.5} SD(A_s) n^{0.5} \quad (8)$$

RESULTS AND DISCUSSION

The specific activity of radionuclides in soil samples

Descriptive statistics of specific activities for radionuclides determined in soil samples are listed in tab. 4. Exploratory data analysis revealed the existence of no outliers. The Shapiro-Wilk (S-W) and Lilliefors tests were used to test the normality or lognormality of a dataset of specific activities, while gamma distribution was tested by the Anderson-Darling (A-D) and Kolmogorov-Smirnov (K-S) tests. Results of those goodness-of-fit tests at a 95 % significance level, summarized in tab. 5, showed that ²²⁶Ra

Table 4. Descriptive statistics of specific activities (A_s) of analyzed radionuclides in all soil samples

A_s [Bqkg ⁻¹]	Mean	Median	Minimum	Maximum	Range	SD	CV [%]	Mode	Skewness	Kurtosis
⁴⁰ K	353	444	60.4	629	568.6	196	55.48	NA	-0.58	-1.37
²²⁶ Ra	39.8	39.2	32.2	49.6	17.4	5.8	14.67	42.2	0.26	-1.48
²³² Th	38.9	36.8	30.8	49.1	18.3	5.5	14.10	NA	0.61	-0.54
²³⁸ U	41.0	38.4	31.0	67	36	8.4	20.37	38.3	1.92	4.70
¹³⁷ Cs	5.9	3.3	0.91	16.7	15.8	5.5	92.80	NA	0.93	-0.74

Table 5. Goodness of fit test for data sets of specific activities for radionuclides of interest

Distribution	Goodness of fit test	⁴⁰ K		²²⁶ Ra		²³² Th		²³⁸ U		¹³⁷ Cs	
		Test statistics	Critical value	Test statistics	Critical value	Test statistics	Critical value	Test statistics	Critical value	Test statistics	Critical value
Normal	S-W	0.822	0.897	0.910	0.897	0.907	0.897	0.830	0.897	0.814	0.897
	Lilliefors	$p = 0.003$		$p = 0.109$		$p = 0.085$		$p = 0.003$		$p = 0.002$	
Lognormal	S-W	0.752	0.897	0.912	0.897	0.926	0.897	0.909	0.897	0.918	0.897
	Lilliefors	$p = 2 \cdot 10^{-4}$		$p = 0.119$		$p = 0.182$		$p = 0.0767$		$p = 0.154$	
Gamma	K-S	0.331	0.206	0.185	0.203	0.172	0.203	0.171	0.203	0.157	0.208
	A-D	2.020	0.752	0.661	0.738	0.670	0.738	0.664	0.739	0.701	0.761
Distribution (0.05 significance)		Distribution-free		Normal		Normal		Approximate normal*		Gamma	
$UCL_{0.95}$ type		Chebyshev (Mean, SD), eq. (8)		Student's-t, eq. (6)		Student's-t, eq. (6)		Student's-t, eq. (6)		Adjusted gamma, eq. (7)	
$UCL_{0.95}$ [Bqkg ⁻¹]		554		42.1		41.1		44.4		9.4	

* When dealing with a small data set ($n < 50$), like data set in this research ($n = 18$), and Lilliefors test suggests that data are normal and the Shapiro-Wilk test suggests that data are not normal, ProUCL will suggest that the data set follows an approximately normal distribution, therefore, $UCL_{0.95}$ of the mean has to be derived based on Student's-t statistics

and ^{232}Th followed a normal distribution, ^{238}U fitted well lognormal distribution, while ^{137}Cs was both lognormally and gamma-distributed. The specific activity of ^{40}K did not follow any distribution tested.

The coefficient of variation (CV) is the most discriminating factor for describing variability. This factor for ^{137}Cs was 92.80 % pointing out its anthropogenic origin and uneven distribution over the study area. For natural radionuclides, CV fell in the range of 14.10 to 20.37 %, except for ^{40}K (55.48 %). Based on such a high CV value for ^{40}K , one can infer that this radionuclide is likely to be influenced by anthropogenic activities and sources like the application of potassium fertilizers. Mean $A_s(^{40}\text{K})$ was well below the average value reported for Serbian soils [29]. Specific activities lower than 75 Bqkg^{-1} were measured in the soil at four locations (6, 7, 15, 16), in soils with a predominant content of sand, therefore leaching may occur in those soils as sandy soils do not contain enough clay to hold the potassium [30]. Moreover, the common practice before constructing green areas is removing the topsoil and replacing it with foreign material rich with sand. The highest ^{40}K specific activity was measured at Trg Despota Stefana (location 4) which is in proximity to the green market, which has been located there since 1929. For this site, the potassium accumulation would match the probable use of fertilizers and manures for horticultural production in the early 1900, as observed by Rate [31] for parklands in Western Australia, and Mitrović *et al.* for suburban soil in Belgrade and Pančevo (Serbia) [32].

The distribution of ^{137}Cs specific activity from soil samples showed the greatest variability in the distribution typical for contamination after the fallout deposition, which complies with the study's results carried out for ^{137}Cs contamination of Belgrade, Serbian capital [33]. Since ^{137}Cs contamination in the soils of the Balkan peninsula was mainly caused by wet deposition after the Chernobyl accident, this inhomogeneous distribution was most probably due to general meteorological conditions (rainfall and wind direction) in May and June of 1986 [2, 34].

According to UNSCEAR [35], the worldwide average specific activities in the soil for ^{40}K , ^{226}Ra , ^{232}Th , and ^{238}U are 412, 32, 45, and 33 Bqkg^{-1} , respectively. The average specific activities of ^{226}Ra and ^{238}U were slightly higher than corresponding worldwide averages, while those of ^{40}K and ^{232}Th were lower. Overall ranges for specific activities of primordial radionuclides were within their ranges obtained for the entire territory of Serbia, but only $A_s(^{232}\text{Th})$ closely matched the reported mean value of 37.8 Bqkg^{-1} . The mean specific activity for ^{238}U and ^{40}K notably departed averages for Serbia, *i. e.*, 32.8 and 550 Bqkg^{-1} , respectively [29].

Due to similarity in use and maintenance of the soil in urban green areas and yards and playgrounds of schools and kindergartens, to put results obtained in this

study in perspective, they have been compared with the results of available researches whose subject was radioactivity of such soil. The specific activities of radionuclides in this study were within the reported ranges in the study carried out for public schools and kindergartens in the city of Kragujevac, Serbia [4], with the slightly higher mean value of $A_s(^{226}\text{Ra})$ (34.6 Bqkg^{-1}). The specific activities of ^{226}Ra and ^{232}Th measured in our research complied with the results published for high schools' yards in Trabzon, Turkey (average values of 35 and 38 Bqkg^{-1} , respectively), but specific activities of ^{40}K (587 Bqkg^{-1}) and ^{137}Cs (38 Bqkg^{-1}) were significantly higher than in Kruševac [36]. The study analyzing soil radioactivity of ten elementary school playgrounds in Gwangju Metropolitan City (South Korea) revealed significantly lower specific activities of ^{226}Ra , ^{232}Th , and ^{137}Cs , but $A_s(^{40}\text{K})$ in Gwangju was twice as high in Kruševac [37].

The mean specific activities determined in Kruševac parks and playgrounds were higher than values obtained for 49 green public sites in Lima soils (Argentina) for ^{226}Ra (22 Bqkg^{-1}), and well below ^{40}K (597 Bqkg^{-1}). Radioactivity of ^{232}Th and ^{137}Cs was comparable but should be highlighted that nuclear weapon tests carried out in the southern hemisphere were the sole source of ^{137}Cs around the La Plata region [38].

With the purpose of deeper insight, a detailed assessment of soil radioactivity in the study area was provided, and radionuclide contents were compared with the radionuclide specific activities in surface soil reported for other cities and regions in Serbia and the Balkan countries, tabs. 6 and 7. The ranges of specific activities for natural radionuclides determined in this paper have been found to lie nearly within the others, and their average specific activities did not differ to a great extent. The mean of ^{226}Ra specific activities were higher than in most other studies except for soil in North Kosovska Mitrovica [12]. Similar ^{40}K content was measured in surface soil in Western Serbia [11] and Priština [13]. Generally, $A_s(^{40}\text{K})$ in the soil of Kruševac soil was among the lowest, and only lower specific activity was reported for Subotica [15]. Janković Mandić *et al.* [15] and Manić *et al.* [16] determined notably lower specific activities of ^{232}Th for Subotica and Niš, respectively, compared to results reported here. Average $A_s(^{232}\text{Th})$ measured in Central and Western Serbia [2, 11] were appreciably higher which is not surprising since natural radioactivity in those regions originates from the magmatic rocks. In comparison with Balkan countries, specific activities for ^{226}Ra measured in Kruševac, concurred well with the values of specific activities for this radionuclide reported for North Macedonia [39, 40]. The specific activities of ^{226}Ra in this study were higher than those obtained in Mojkovac, Montenegro [41], but below mean values reported for soil in Nikšić, Montenegro [42], Slovenia [43], Istria County (Croatia) [44] and

Table 6. Mean values and ranges (in parenthesis) of specific activities (A_s) of natural radionuclides and ^{137}Cs and absorbed dose rate (\dot{D}) due to natural radionuclides in surface soil in other Serbian cities and regions

City	A_s [Bqkg ⁻¹]					\dot{D} [nGyh ⁻¹]
	⁴⁰ K	²²⁶ Ra	²³² Th	²³⁸ U	¹³⁷ Cs	
Vojvodina province [34, 46]	569 (238-1000)	35 (10-43)	43 (12-71)	42 (5-80)	8.57 (2.73-18.9)	59
Toplica region [18]	492 (7.2-1053)	29.9 (3.3-48.2)	36.6 (0.9-58.9)	–	56.4 (2.4-99.8)	56.4 (2.4-99.8)
Western Serbia [11]	379	33.2	49.1	60.4	36.4	73.4
Belgrade [17, 33]	490 (310-650)	35 (19-51)	43 (23-58)	–	29.9 (1.00-180)	63 (39-80)
Subotica [15]	290 (260-390)	20 (12-33)	18 (13-23)	–	–	32 (24-46)
Kuršumljija [14]	420 (263-700)	22 (9-37)	31 (15-53)	–	8.2 (BDL-38)	47.0 (23.9-73.5)
Kragujevac [2]	425.8 (167-559)	33.5 (20.4-55.2)	50.3 (29.9-73.4)	–	40.2 (0.5-90.5)	63.7 (43.7-83.2)
Niš [16]	414 (137-715)	21 (6.0-139)	26 (4.4-72)	–	4.7 (1-76)	39 (13-110)
North Kosovska Mitrovica [12]	743.2 (365.1-1148.7)	40.6 (21.2-91.1)	48 (20.3-103.8)	–	81 (6.8-385)	78.7 (38.9-149)
Priština [13]	375.4 (172-630)	23.7 (10.4-39.2)	35.1 (11.9-58.2)	–	–	47.8 (19.2-63.8)

Table 7. Mean values and ranges (in parenthesis) of specific activities (A_s) of natural radionuclides and ^{137}Cs and absorbed dose rate (\dot{D}) due to natural radionuclides in surface soil in Balkan countries

City, Country	A_s [Bqkg ⁻¹]					\dot{D} [nGyh ⁻¹]
	⁴⁰ K	²²⁶ Ra	²³² Th	²³⁸ U	¹³⁷ Cs	
Nikšić, Montenegro [42]	491.5 (284.9-724.6)	60.2 (26.3-136.8)	4.6 (33.9-78.6)	–	172.2 (2.91-475.9)	102.6 (61.3-171)
Mojkovac, Montenegro [41]	620.8 (515.6-753.4)	28.6 (20.5-38.3)	43.1 (31.2-56.3)	–	55 (1.26-276)	65.18 (52.32-74.47)
North Macedonia, entire territory [39]	584 (80-1390)	37 (9-123)	39 (7-145)	39 (9-111)	–	–
Veles, North Macedonia [47]	607 (30-1001)	–	53 (22-101)	28 (3.7-43)	71 (1.6-358)	–
Kavadarci, North Macedonia [40]	546 (286-801)	38.8 (21.7-91.6)	43.7 (25.0-131.8)	–	41.5 (4.0-220.3)	67 (42-148)
Slovenia, entire territory [43]	800 (98-2600)	63 (12-270)	77 (9-170)	–	–	110 (15-260)
Istria county, Croatia [44]	418 (11.2-1200)	69 (21.1-127)	60 (3.18-101)	61 (14.5-279)	–	–
Republic of Srpska, Bosnia and Herzegovina [45]	536 (60-821)	47 (5.1-128)	41 (6.8-72)	64 (6.5-228)	26 (2.1-68)	69 (9-118)
Bihać, Bosnia and Herzegovina [53]	367 (193-424)	–	41 (22-53)	54 (43-65)	–	65.10 (49.08-73.94)

Republic of Srpska (Bosnia and Herzegovina) [45]. The mean specific activity of ⁴⁰K and ²³²Th was lower than the mean values reported for the neighboring states. These differences in terrestrial radioactivity could be attributed to the geological and geographical specificities of investigated areas.

The mean and maximal values of ¹³⁷Cs specific activity were significantly lower than those obtained from studies previously conducted in Serbia and the Balkan region with exception of Niš [16], tabs. 6 and 7. Comparable ¹³⁷Cs specific activities were found recently in Vojvodina province [34, 46] and Kuršumljija [14]. Low ¹³⁷Cs specific activity in this research can be a consequence of pedology as the alluvial soil is the predominant type of soil in the study area, which is supported by the study of Dimovska *et al.* [47] who measured the lowest ¹³⁷Cs specific activity in alluvial sediments. Additionally, all studied soils have been disturbed due to construction and maintenance works

and consequent mixing with a deeper soil layer with lower $A_s(^{137}\text{Cs})$ [38]. On the other hand, it was found that ¹³⁷Cs activities in the soil samples collected from locations 5, 9, and 17 were notably higher than others (15.0, 16.9, 14.4 Bqkg⁻¹, respectively). These samples were collected from the three biggest and most forested parks. This feature can be explained by the high ¹³⁷Cs activity of the forested land that is rich in organic matter due to decomposed plant materials, with the ability to adsorb cesium onto humic substances that slows down the migration of ¹³⁷Cs in the soil [18, 33].

Radiological risk assessment

The total intakes of radionuclides and their progenies through ingestion and inhalation of soil, as well as external exposure for the whole period of exposure for a recreator in the area under the study, are summa-

Table 8. Total excess lifetime cancer risk for recreator in the study area stratified by exposure route and radionuclide calculated from $UCL_{0.95}$ of the mean of the radionuclide specific activities (P – short-lived decay products)

Isotope	$ELCR_{ing}$	$ELCR_{inh}$	$ELCR_{ex}$	$ELCR_{total}$
$^{137}\text{Cs} + \text{P}$	$1.95 \cdot 10^{-9}$	$9.92 \cdot 10^{-35}$	$8.24 \cdot 10^{-8}$	$8.44 \cdot 10^{-8}$
^{40}K	$2.10 \cdot 10^{-7}$	$1.53 \cdot 10^{-32}$	$2.21 \cdot 10^{-6}$	$2.42 \cdot 10^{-6}$
$^{226}\text{Ra} + \text{P}$	$1.84 \cdot 10^{-7}$	$1.47 \cdot 10^{-31}$	$1.72 \cdot 10^{-6}$	$1.91 \cdot 10^{-6}$
$^{232}\text{Th} + \text{P}$	$5.78 \cdot 10^{-7}$	$4.45 \cdot 10^{-31}$	$8.16 \cdot 10^{-7}$	$1.39 \cdot 10^{-6}$
$^{238}\text{U} + \text{P}$	$5.67 \cdot 10^{-8}$	$1.31 \cdot 10^{-31}$	$2.58 \cdot 10^{-8}$	$8.25 \cdot 10^{-8}$
$ELCR_{total}$	$1.03 \cdot 10^{-6}$	$7.39 \cdot 10^{-31}$	$4.86 \cdot 10^{-6}$	$5.89 \cdot 10^{-6}$

rized in tab. 3. The total intakes of radionuclides were calculated based on the $UCL_{0.95}$ presented in tab. 5. The current soil content of radionuclides of interest in the study area led to a total intake of 165 Bq for the 26 years of exposure through ingestion and inhalation. More than 80 % of this figure came from ingestion of ^{40}K , 19 % from ingestion of U and Th series radionuclides, and only 1 % from ^{137}Cs . These values seem to be negligible since reference values for annual dietary intake for adults are 22, 1.7, and 5.7 Bq for ^{226}Ra , ^{232}Th , and ^{238}U , respectively [1]. Calculated $TI_{ing}(^{40}\text{K})$ for the recreator in the study area for the total exposure time was found to be two orders of magnitude lower than the total estimated annual intake of ^{40}K in a diet for Pakistani adults ($2.88 \cdot 10^4$ Bq), and for the Chinese adult man ($1.8 \cdot 10^4$ Bq) [48]. This implies that ^{40}K intake is not pertinent from a radiation protection point of view, whereas the potassium content of the body is under strict homeostatic control and is not influenced by variations in its environmental levels [49].

The results of the health risk assessed for each applicable exposure pathway stratified by radionuclide and its daughters are given in tab. 8. Among the three different plausible exposure pathways for the given exposure scenario, external exposure appeared to be the main route of exposure (82.5 %) to soil and thus posed the highest health risk to recreators at selected locations for all radionuclides, followed by ingestion (17.4 %). The total risk for external exposure was $4.86 \cdot 10^{-6}$, $1.03 \cdot 10^{-6}$ for ingestion, while $ELCR$ for inhalation of resuspended soil particles can be considered negligible ($7.39 \cdot 10^{-31}$). Furthermore, the total risk of each radionuclide (and respective progeny, if there any) was in the following order: ^{40}K ($2.42 \cdot 10^{-6}$) > ^{226}Ra ($1.91 \cdot 10^{-6}$) > ^{232}Th ($1.39 \cdot 10^{-6}$) > ^{137}Cs ($8.44 \cdot 10^{-8}$) > ^{238}U ($8.25 \cdot 10^{-8}$), with a portion in the total risk of 41.1 %, 32.4 %, 23.7 %, 1.4 %, and 1.4 %, respectively. When analyzing all ways of exposure and radionuclides combined, external exposure risk from ^{40}K in soil contributed most to the $ELCR_{total}$ with 37.6 %. External irradiation due to ^{226}Ra accounted for 29.3 % and due to ^{232}Th 13.9 % of the total risk. Incident ingestion of ^{232}Th contained in soil induced risk of $5.78 \cdot 10^{-7}$ that is more than half of the total $ELCR_{ing}$ (56.1 %).

Assuming cancer risk is linear at low doses (linear no-threshold model), US EPA considers risk below the target value of 10^{-6} to be negligibly small without the potential to provoke significant health effects. The

acceptable $ELCR$ threshold value is 10^{-4} , which equates to one lifetime excess radiogenic cancer being expected among 10000 exposed individuals. Risk surpassing 10^{-4} is viewed as unacceptable. Assuming exposure duration of 25-30 years, this risk yields an annual effective dose of approximately 100-150 Sv to a reasonably maximally exposed individual at a site containing radionuclides. Cancer risk lying between 10^{-6} and 10^{-4} (especially those in the 10^{-5} to 10^{-6} range) has been judged to be tolerable for regulatory purposes [50, 51]. The results presented in this study indicate that the risk for cancer development due to recreational use of the selected areas exists but is very small. The value of the aggregate cancer risk was $5.89 \cdot 10^{-6}$ which is well below the threshold value of 10^{-4} . It is worth mentioning that risk posed by the content of ^{40}K , ^{226}Ra , and ^{232}Th in soil exceeded the target risk but was very close to the 10^{-6} . Moreover, no detrimental health effects are expected to occur through the inhalation of particulates in individuals exposed under the conditions of assessment, since for outdoor air exposure pathway the target health risk limit was not exceeded for any radionuclide. Also, for ^{137}Cs and its progeny, as well as for ^{238}U , the total risk value for investigated soils was lower than the target risk.

The Institute of Public Health of Serbia recorded the standardized cancer incidence rate in 2015 for Rasina District of $3.02 \cdot 10^{-3}$ for males and $2.73 \cdot 10^{-3}$ for females [52]. In comparison to these very high values, $ELCR_{total}$ obtained in this work made a portion of the real cancer incidence that was completely insignificant (approximately 0.2 %).

In assessing the risk of radionuclides and decision-making for polluted or potentially polluted sites aimed at protecting human health and the environment, it is beneficial to know human health risks at targeted locations. The spatial distribution of $ELCR_{total}$ in the study area estimated using site-specific values of radionuclide specific activities is presented in fig. 2. The proportional map symbol is used for visualizing quantitative data and illustrating differences between locations. The total risk varied from $3.41 \cdot 10^{-6}$ (location 16) to $5.81 \cdot 10^{-6}$ (location 10), and the mean value was $4.79 \cdot 10^{-6}$. As can be seen, the total risk for all investigated urban areas was higher than 10^{-6} , but never exceeded 10^{-5} , making them safe in terms of radioecology for use by visitors. There was no discernible trend of the $ELCR_{total}$ spatial distribution over the

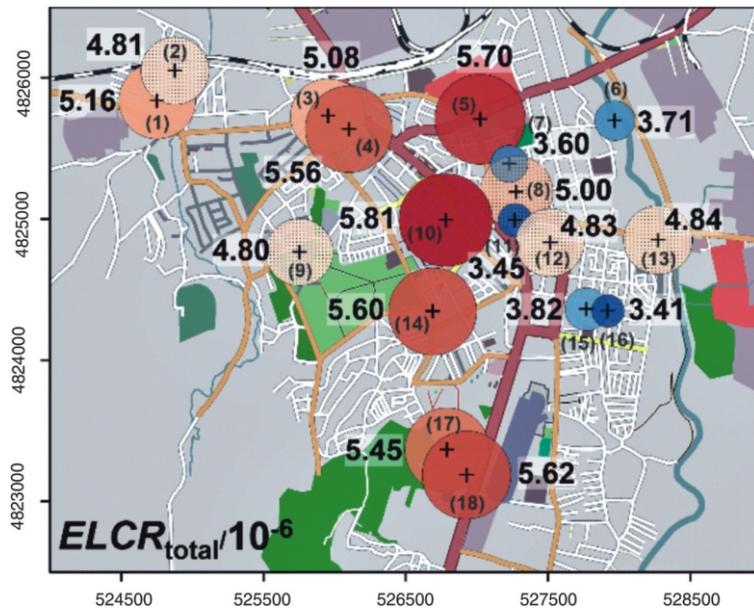


Figure 2. Estimated total lifetime cancer risk ($ELCR_{total}$) due to investigated radionuclides for recreator per individual location

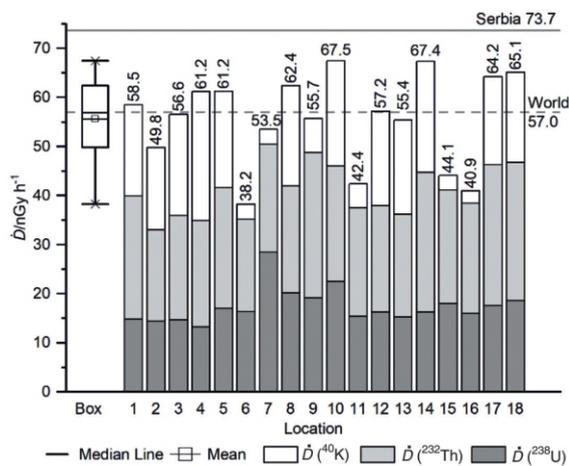


Figure 3. Total absorbed dose rate (\bar{D}) in the air due to natural radionuclides and contribution to the total dose rate from ^{40}K and radionuclides from ^{238}U and ^{232}Th series

study area, but it was perceptible that the factor most affecting total assessed risk was the content of ^{40}K . Namely, the sole significant correlation at the 0.01 level expressed as Spearman's correlation coefficient was found between total risk and specific activity of ^{40}K (0.81). Most compelling evidence of such ^{40}K influence was $ELCR_{total}$ being lower than $4 \cdot 10^{-6}$ for spots numerated with 6, 7, 11, 15, and 16 (fig. 2) where the lowest $A_s(^{40}\text{K})$ was measured (range 64.1 to 118 Bqkg^{-1}). Among other places (not including 9 – Bagdala public park) soil content of ^{40}K was appreciably higher (402-629 Bqkg^{-1}). The discrepancy from this pattern was recorded only for Bagdala with a total risk of $3.41 \cdot 10^{-6}$, which was the location with the higher radium and thorium content (48.0 and 49.1 Bqkg^{-1} , respectively) making 79.9 % of accompanying total cancer risk, while $A_s(^{40}\text{K})$ was 167 Bqkg^{-1} .

Having in mind that data on the radioactivity levels were obtained for the first time for the study area, some uncertainties could not be neglected. Only a rela-

tively small data set was considered in the current work. Furthermore, we used some exposure parameters provided by the US EPA because there is still no specified value of these parameters for Serbian people. Therefore, further investigations based on greater sample size and involving exposure parameters with site-specific exposure parameters that could reflect local human activity mode are recommended to accurately quantify the risk posed by radionuclides. Despite the uncertainties involved, the risk assessment model has proven to be a very useful tool to identify the radionuclide and exposure route of most concern in an urban environment and to reveal the true meaning and relevance of the radionuclide specific activity found in urban soil.

The total absorbed gamma dose rate in the air due to natural gamma emitters in the soil for each location investigated, together with a box-plot diagram is shown in fig. 3. The dose rate varied from 38.2 (location 6) to 67.5 nGyh^{-1} (location 10) with a mean value of 55.7 nGyh^{-1} that is slightly lower than the worldwide average value (57 nGyh^{-1}) [1]. All calculated values never exceeded the average dose rate for Serbia of 73.7 nGyh^{-1} [29]. These results are in the range typical for other Serbian cities and regions, tabs. 6 and 7, but some differences exist due to geological diversity. The calculated mean dose rate for Kruševac was two times lower than the dose rates reported for Nikšić, Montenegro [42], and Slovenia [43]. The reason lies in the geology of those environs since a number of samples with the highest specific activity of natural radionuclides were taken from areas where calcareous sediments, clastic sediments containing clay, and carbonate rock are dominant rock types having somewhat higher radiation background. Besides that, estimated dose rates did not significantly differ from external exposure rates from terrestrial gamma radiation for neighboring countries [40, 41, 45, 53] and South Europe [1, 35]. This shows that radiation in the study area was

within the natural limits. The contribution to the radiation dose from calculated radionuclides is non-uniform and reflects the variability of radionuclide-specific activities among sampling locations. Radionuclides from the ^{232}Th series were found to be the main contributor to the terrestrial gamma dose rate in the study area, contributing on average with 43 % (35-56 % range) to the total value, which is similar to results of research recently conducted in Belgrade [17]. The second contributor to the total dose was the U family with 32 % (22-53 % range), and the ^{40}K share was 25 %. The great variability of the dose rate among investigated sites was a consequence of different ^{40}K specific activity since the dose rate from this single radionuclide varied from 2.5 nGyh^{-1} (6 %) to 26.2 nGyh^{-1} (43 % of the total dose rate).

CONCLUSIONS

To our knowledge, this was the first radioecological study of urban soil for this region of Serbia with a focus on urban green areas. Targeted soil sampling and analysis of natural and man-made radionuclides were carried out to estimate associated health risks for users of selected locations through exposure to soils utilizing a proven US EPA risk assessment model for mainland use as recreational open space. Comparative analysis of the study results with the other regions in Serbia and other Balkan countries supported the assertion that the specific activities of selected radionuclides in the soil of parks and playgrounds of Kruševac were not elevated. The calculated cancer risk lay above the target risk value of 10^{-6} , but well below 10^{-4} above which the results are not acceptable. This reflected that exposure to naturally occurring radionuclides and ^{137}Cs carried low risk due to exposure to soils but did not mean any significant health concern for recreators and visitors in the area under evaluation.

The obtained results across the present research could be a reference for future radiological studies and radioactivity mapping in this region and as a basis for regular local and regional environmental quality and monitoring. Since urban parks and playgrounds are important outdoor resting and recreational places for urban dwellers, the health risk determined in this study could serve as a useful case study for the assessment of health risk posed by radionuclides in the urban environment in other cities and countries.

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AUTHORS' CONTRIBUTIONS

All authors contributed to the study's conception and design. Material preparation, data collection, and analysis were performed by Milan N. Tanić, Denis P. Dinić, Željko A. Mihaljlev, and Brankica D. Kartalović. All authors performed literature research, theoretical analysis, and discussion of the presented results. The first draft of the manuscript was written by Milan N. Tanić and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

REFERENCES

- [1] ***, Sources and Effects of Ionizing Radiation, Report to the General Assembly, with Scientific Annexes, United Nation Scientific Committee on the Effects of Atomic Radiation, New York, United Nations, 2000
- [2] Milenković, B., et al., Radioactivity Levels and Heavy Metals in the Urban Soil of Central Serbia, *Environ Sci Pollut Res*, 22 (2015), 21, pp. 16732-16741
- [3] Madrid, L., et al., Variability in Concentrations of Potentially Toxic Elements in Urban Parks from Six European Cities, *J Environ Monit*, 8 (2006), 11, pp. 1158-1165
- [4] Stajic, J. M., et al., Exposure of School Children to Polycyclic Aromatic Hydrocarbons, Heavy Metals, and Radionuclides in the Urban Soil of Kragujevac city, Central Serbia, *Chemosphere*, 146 (2016), Mar., pp. 68-74
- [5] Towle, K. M., et al., The Cancer Risk Associated with Residential Exposure to Soil Containing Radioactive Coal Combustion Residuals, *Risk Anal*, 38 (2017), 6, pp. 1107-1115
- [6] Nadal, M., et al., Human Health Risk Assessment of Environmental and Dietary Exposure to Natural Radionuclides in the Catalan Stretch of the Ebro River, Spain, *Environ Monit Assess*, 175 (2011), 1-4, pp. 455-468
- [7] De Miguel, E., Risk-Based Evaluation of the Exposure of Children to Trace Elements in Playgrounds in Madrid (Spain), *Chemosphere*, 66 (2007), 3, pp. 505-513
- [8] Mirzaei, R., et al., Comparative Study of Heavy Metals Concentration in Topsoil of Urban Green Space and Agricultural Land Uses, *Environ Monit Assess*, 187 (2015), 12, p. 741
- [9] Marija, P., et al., Evaluation of Urban Contamination with Trace Elements in City Parks in Serbia Using Pine (*Pinus Nigra* Arnold) Needles, Bark and Urban Topsoil, *Int J Environ Res*, 11 (2017), 5-6, pp. 625-639
- [10] Pavlović, D., et al., Fractionation, Mobility, and Contamination Assessment of Potentially Toxic Metals in Urban Soils in Four Industrial Serbian Cities, *Arch Environ Contam Toxicol*, 75 (2018), 3, pp. 335-350
- [11] Dugalic, G., et al., Heavy Metals, Organics, and Radioactivity in Soil of Western Serbia, *J Hazard Mater*, 177 (2010), 1-3, pp. 697-702
- [12] Gulan, L., et al., Correlation Between Radioactivity Levels and Heavy Metal Content in the Soils of the North Kosovska Mitrovica Environment, *Environ Sci Process Impacts*, 15 (2013), 9, pp. 1735-1742
- [13] Gulan, L., et al., Persistent Organic Pollutants, Heavy Metals, and Radioactivity in the Urban Soil of Priština City, Kosovo and Metohija, *Chemosphere*, 171 (2017), Mar., pp. 415-426

- [14] Gulan, L., et al., Environmental Radioactivity with Respect to Geology of Some Serbian Spas, *J Radioanal Nucl Chem*, 317 (2018), 1, pp. 571-578
- [15] Janković Mandić, L., et al., The Natural Radionuclides in Soils of Subotica (Serbia): Distribution and Corresponding Gamma Dose Rates, In: RAD Conference Proceedings, (Editor: Ristić, G.), 1 (2016), pp. 71-74
- [16] Manić, V., et al., Radioactivity of Soil in the Region of the Town of Niš, Serbia, *Radiat Prot Dosimetry*, 185 (2019), 4, pp. 456-463
- [17] Petrović, J., et al., Assessment of Radiation Exposure to Human and Non-Human Biota Due to Natural Radionuclides in Terrestrial Environment of Belgrade, the Capital of Serbia, *Environ Earth Sci*, 77 (2018), 7, p. 290
- [18] Stevanović, V., et al., Environmental Risk Assessment of Radioactivity and Heavy Metals in Soil of Toplica Region, South Serbia, *Environ Geochem Health*, 40 (2018), 5, pp. 2101-2118
- [19] ***, General Urban Plan of Kruševac 2014-2025, City of Kruševac, Kruševac, Serbia, 2015
- [20] ***, Climatolog – 30-year Averages 2019, 2019, [Online], Available, Republic Hydrometeorological Service of Serbia, Accessed 12 June 2019]
- [21] Smagin, A. V., et al., Criteria and Methods to Assess the Ecological Status of Soils in Relation to the Landscaping of Urban Territories, *Eurasian Soil Sci*, 39 (2006), 5, pp. 539-551
- [22] ***, Soil Sampling for Environmental Contaminants, IAEA-TECDOC-1414, International Atomic Energy Agency, Vienna, 2004
- [23] ***, US Department of Energy, RAIS The Risk Assessment Information System, 2009, (accessed July 11, 2017)
- [24] ***, Cancer Risk Coefficients for Environmental Exposure to Radionuclides, Federal Guidance Report No.13, Office of Radiation and Indoor Air, US Environmental Protection Agency, Washington, 1999
- [25] ***, Exposure Factors Handbook: 2011 Edition, National Center for Environmental Assessment, Office of Research and Development, US Environmental Protection Agency, Washington, 2011
- [26] ***, ProUCL Version 5.1 Technical Guide, Statistical Software for Environmental Applications for Data Sets with and without Nondetect Observations, US Environmental Protection Agency, Office of Research and Development, Washington, 2015
- [27] ***, Calculating Upper Confidence Limits for Exposure Point Concentrations at Hazardous Waste Sites, US Environmental Protection Agency, Office of Emergency and Remedial Response, Washington, 2002
- [28] ***, ProUCL Version 5.1 User Guide Statistical Software for Environmental Applications for Data Sets with and without Nondetect Observations, US Environmental Protection Agency, Office of Research and Development, Washington, 2015
- [29] Dragović, S., et al., Lithogenic Radionuclides in Surface Soils of Serbia: Spatial Distribution and Relation to Geological Formations, *J Geochemical Explor*, 142 (2014), July, pp. 4-10
- [30] Kolahchi, Z., Jalali, M., Effect of Water Quality on the Leaching of Potassium from Sandy Soil, *J Arid Environ*, 68 (2007), 4, pp. 624-639
- [31] Rate, A. W., Multielement Geochemistry Identifies the Spatial Pattern of Soil and Sediment Contamination in an Urban Parkland, Western Australia, *Sci Total Environ*, 627 (2018), June, pp. 1106-1120
- [32] Mitrović, B. B., et al., Radionuclides and Heavy Metals in Soil, Vegetables, and Medicinal Plants in Suburban Areas of the Cities of Belgrade and Pančevo, *Nucl Technol Radiat*, 34 (2019), 3, pp. 278-284
- [33] Petrović, J., et al., Spatial Distribution and Vertical Migration of ^{137}Cs in Soils of Belgrade (Serbia) 25 Years After the Chernobyl Accident, *Environ Sci Process Impacts*, 15 (2013), 6, p. 1279
- [34] Spasić-Jokić, V., et al., Effective Dose Estimation and Lifetime Cancer Mortality Risk Assessment from Exposure to Chernobyl ^{137}Cs on the Territory of Belgrade City and the Region of Vojvodina, Serbia, *Environ Sci Pollut Res*, 18 (2011), 5, pp. 708-715
- [35] ***, Sources, Effects and Risks of Ionizing Radiation, UNSCEAR 2008 Report to the General Assembly, with annexes, Volume 1, Annex B: Exposures of the Public and Workers from Various Sources of Radiation, New York, United Nations, 2010
- [36] Cevik, U., et al., Assessment of Radiological Levels at Schools in Trabzon, Turkey, *Indoor Built Environ*, 22 (2013), 2, pp. 376-383
- [37] Cho, J. H., et al., A Study on the Measurement and Analysis of Radioactivity Concentration and the Ambient Dose Rate in Soil on the Playgrounds of Elementary Schools in the Gwangju Area, *Environ Earth Sci*, 71 (2014), 5, pp. 2391-2397
- [38] Montes, M. L., et al., Activities of ^{232}Th , ^{226}Ra , ^{40}K , and ^{137}Cs in Surface Soil and External Dose Assessment at Two Zones of Buenos Aires Province, Argentina, *Environ Earth Sci*, 75 (2016), 4, p. 320
- [39] Stojanovska, Z., et al., Analysis of Specific Radionuclide Activity Variations in Soil within Geotectonic Units of Republic of North Macedonia, *Nucl Technol Radiat*, 34 (2019), 1, pp. 85-93
- [40] Dimovska, S., et al., Radioactivity in Soil from the city of Kavadarci (Republic of Macedonia) and its Environs, *Radiat Prot Dosimetry*, 148 (2012), 1, pp. 107-120
- [41] Antović, N. M., et al., Radioactivity in Soils from Mojkovac, Montenegro, and Assessment of Radiological and Cancer Risk, *Nucl Technol Radiat*, 27 (2012), 1, pp. 57-63
- [42] Antović, N. M., et al., Radioactivity Impact Assessment of Nikšić Region in Montenegro, *J Radioanal Nucl Chem*, 302 (2014), 2, pp. 831-836
- [43] Kovacs, T., et al., Systematic Survey of Natural Radioactivity of Soil in Slovenia, *J Environ Radioact*, 122 (2013), Aug., pp. 70-78
- [44] Radolić, V., et al., The Natural Radioactivity of Istria, Croatia, *Radiat Phys Chem*, 155 (2019), February, pp. 332-340
- [45] Janković, M., et al., Radioactivity Measurements in Soil Samples Collected in the Republic of Srpska, *Radiat Meas*, 43 (2008) 8, pp. 1448-1452
- [46] Todorović, N., et al., Radioactivity in Fertilizers and Radiological Impact, *J Radioanal Nucl Chem*, 303 (2014), 3, pp. 2505-2509
- [47] Dimovska, S., et al., Distribution of Some Natural and Man-Made Radionuclides in Soil from the City of Veles (Republic of Macedonia) and its Environs, *Radiat Prot Dosimetry*, 138 (2010), 2, pp. 144-157
- [48] Jun, T., et al., Potassium, In: Radionuclides in the Environment (Editor: Atwood, D. A.), John Wiley & Sons, Ltd, Chichester, 2010, pp. 65-71
- [49] ***, IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Volume 78: Ionizing Radiation, Part 2: Some Internally Deposited Radionuclides, International Agency for Research on Cancer, World Health Organization, Lyon, 2001
- [50] ***, Role of the Baseline Risk Assessment in Superfund Remedy Selection Decisions, OSWER9355030, Office of Solid Waste and Emergency Response, U.S. Environmental Protection Agency, Washington, 1991

- [51] ***, Supplemental Guidance for Developing Soil Screening Levels for Superfund Sites, OSWER 9355.4-24, U.S. Environmental Protection Agency, 2002
- [52] ***, Health Statistical Yearbook of Republic of Serbia 2017, Institute of Public Health of Serbia, Belgrade, 2018
- [53] Pehlivanović, B., et al., Measurement of Natural Environmental Radioactivity and Estimation of Popula-

tion Exposure in Bihac, Bosnia and Herzegovina, *J Radioanal. Nucl Chem*, 311 (2017), 3, pp. 1909-1915

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**ПРОЦЕНА РИЗИКА ИЗЛАГАЊА ЉУДИ РАДИОНУКЛИДИМА
У ЗЕМЉИШТУ УРБАНИХ ПОВРШИНА (ЈАВНИ ПАРКОВИ И
ИГРАЛИШТА НА ОТВОРЕНОМ) У КРУШЕВЦУ, СРБИЈА**

Циљ истраживања је било прикупљање података о радиоактивности природних (^{40}K , ^{226}Ra , ^{232}Th и ^{238}U) и вештачких (^{137}Cs) радионуклида у површинском слоју земљишта (0-10 cm) осамнаест јавних површина у Крушевцу и процена последичних здравствених ефеката за становнике који користе те површине у рекреативне сврхе. Специфична активност истраживаних радионуклида је анализирана применом HPGe спектрометрије гама-зрачења. Средње вредности специфичних активности ^{40}K , ^{226}Ra , ^{232}Th , ^{238}U и ^{137}Cs су биле 353, 39,8, 38,9, 41,0 и 5,9, респективно, и биле су сагласне резултатима објављеним у сличним истраживањима у другим деловима Србије и суседним државама. У циљу процене ризика по здравље људи услед експозиције радионуклидима, примењен је модел препоручен од стране Агенције за заштиту животне средине Сједињених Америчких Држава полазећи од конзервативних претпоставки о експозицији и узимајући у обзир три пута експозиције: ингестију, инхалацију земљишта и спољашње озрачивање. Израчуната је и јачина апсорбоване дозе гама зрачења које потиче од радионуклида у земљишту. Добијене вредности параметара указују да радиолошки ризик услед природних и вештачких радионуклида није значајан. Укупни канцерогени ризик током животног века који је процењен на основу вредности горње границе 95 % интервала поузданости средње вредности специфичних активности радионуклида је износио $5.89 \cdot 10^{-6}$, што је знатно мања вредност у односу на вредност толерантног ризика која се користи у регулаторне сврхе (10^{-4}). Од свих истраживаних путева експозиције, спољашње озрачивање је највише допринело вредности ризика. Резултати добијени за градске паркове и игралишта сугеришу да је њихова употреба за рекреацију безбедна са радиоэколошког становништва.

Кључне речи: урбана зелена површина, урбано земљиште, радионуклид, радиолошки ризик, људско здравље, гама-спектрометрија