ANALYSING RADIONUCLIDE CONTENT IN SOIL SAMPLES AND RADIOLOGICAL RISKS IN THE CLAYEY MATERIAL SURROUNDING OF THE "ZBEGOVI" DEPOSIT, DONJE CRNILJEVO, SERBIA

by

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This paper presents the results of analyses of radionuclide content in the samples of the surrounding soil and clayey material of "Zbegovi" open-pit mine in Donje Crniljevo, Serbia. Samples from 78 sites were collected and prepared. The activity concentrations were determined for radionuclides: ^{238}U, ^{232}Th, ^{40}K, ^{226}Ra, and ^{137}Cs. The mean values obtained are as follows: 23 Bqkg^{-1}, 89 Bqkg^{-1}, 372 Bqkg^{-1}, 56 Bqkg^{-1}, and 11 Bqkg^{-1}, respectively. Concentrations of ^{238}U, ^{40}K, and ^{226}Ra in the studied area do not deviate from the values obtained for the soil in Serbia. The concentration of ^{232}Th in the studied area is slightly higher relative to average values for soil, and slightly lower compared to similar deposits of clayey material in the world. Measurements performed showed that the open-pit mine of clayey material is completely uncontaminated surface as far as ^{137}Cs is concerned, while there are sites where measured ^{137}Cs concentrations are significantly higher, which is due to topographic differences and inhomogeneous surface contamination of land after the Chernobyl accident.
To assess the radiological risks in the observed area, the following indices were determined: absorbed dose rate, annual outdoor effective dose, absorbed dose for biota, excess lifetime cancer risk outdoors as well as external radiation hazard index. The mean value of the estimated absorbed dose rate in the given area amounts to 80.1 nGyh^{-1}, and the annual outdoor effective dose ranges from 46.9 to 134 Sv. Absorbed dose rate for biota in the studied area is 1.31 10^{-4} Gyd^{-1}. The mean excess lifetime cancer risk outdoors for the population is 3.8 10^{-4}, and the mean value of the external radiation hazard index obtained in this study is 0.48, which is consistent with the world average. A low dose of radiation will not pose a risk to the population and biota in the studied area.

Key words: clay, radioactivity, gamma-ray spectrometry, radiological risks

INTRODUCTION

Exposure to natural radioactivity is inevitable, originating primarily from three natural radioactive series, with progenitors ^{232}Th, ^{238}U, and ^{235}U, as well as from ^{40}K. Uranium and radium, as the most important natural radionuclides, occur in the lithosphere and hydrosphere in various geological objects: rocks, ores, soil, as well as underground and surface waters. The natural concentrations of radionuclides in soil mainly depend on the type of parent rocks from which these soils were created. Concentrations of uranium and thorium are higher in magmatic rocks (*e. g.* granites), but also in some sedimentary rocks (shales and phosphate rocks) [1-3].

The cyclical movement of the chemical elements and isotopes in the environment takes place continuously through geochemical processes. Various radionuclides of natural origin are found in soil, at very low concentrations, generally. Depending on the geochemical processes of the elements, these radionuclides can be concentrated locally in the soil to levels that pose a risk to the population health. In the lithosphere, there are medium-heavy and heavy radionuclides, most often at depths of 5-20 cm, where they build compounds with present organic substances. Subsequently, depending on their physicochemical properties and morphological characteristics of the root systems of present plant species, through the process of absorption, these ra-

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dionuclides reach and concentrate in plants, through which they reach animals and finally, humans. Therefore, radionuclides in the soil are controlled at a depth of 5 to 20 cm [4].

The distribution of artificial radionuclides in soil, primarily ^{137}Cs, is characterized by pronounced unevenness in profile depth. In regions with moderate atmospheric precipitations, in soils with heavier mechanical composition, most of the artificial radionuclides are retained in the surface layer (up to 10 cm deep) of soil over a long period. In lighter, sandy soils, especially those with leaching water regime, a considerable part of radioactive material migrates in 10 to 15 years to 40-50 cm depths but can also reach groundwater and by rivers further transported to seas [5]. The ^{137}Cs radioactive isotope has a half-life of 30.05 years and has reached the environment due to the Chernobyl nuclear power plant accident (1986), but also other nuclear accidents.

Clays are a widespread mineral resource of great industrial significance [6]. Due to their high ion exchange capacity, poor permeability, and retention abilities they are acting as a physical and chemical barrier for various environmental pollutants. They appear as a set of fine-grained clayey minerals or as a type of sedimentary deposit composed mostly of clayey minerals and non-aluminosilicates [7].

The chemical composition of clays is similar to that of primary rocks. By formation, clays can be divided into: residual (formed as a result of surface decomposition of rocks influenced by precipitation, temperature, frost or wind and are mainly found in the place of origin) and transported (sedimentary – formed as a result of chemical decomposition of granite containing silica (SiO_2) and aluminum(III)-oxide, (SiO_2), by the dissolution of rocks, or limestone, and decomposition and dissolution of shales) [7]. Transported clays are secondary because they are transported from the place of origin by erosion and deposition. By chemical composition, clays have more oxygen than silicon, aluminum, or magnesium and can, therefore, be considered hydroxides of silicon, aluminum, or magnesium. Apart from being used in construction, ceramics production, medicine, pharmacy, and paper industry, in recent decades clays have also been applied as adsorbents, catalysts or their carriers, ion exchangers and bleaching materials [8, 9]. All the aforementioned clay applications are determined by their specific properties defined by the presence and type of non-clay and clay minerals, organic matter, type and quantity of exchangeable cations, *etc*. Nowadays, clays are used as cheap, alternative adsorbents for reducing concentrations of various pollutants in the environment. Among many types of clay that can be found in the environment, bentonite clays, consisting predominantly of montmorillonites and related clay minerals from the smectite group stand out by their adsorption properties [8]. Bentonite clays are widely used in the purification of water from heavy metal and as a decontamination agent for radiological decontamination. Recent research from Turkey has shown a new use of clay as an admixture to cement and concrete [6].

In some types of clay, a high ^{232}Th and ^{238}U content was found exceeding permissible limits prescribed by law [9, 10].

In the process of exploiting clay for construction purposes, it is necessary to carry out appropriate analyses of the type, quality, and quantity of mineral raw material as well as of radionuclide contents in clay.

Mass exploitation of clay material not only disrupts the existing relief and ecosystems of a given space but also deposit large quantities of by-products (tailings) in the vicinity that may contain radionuclides in elevated concentrations. A major problem in open-pit mines is the impact of wind and consequent dust scattering (aeolian distribution). Surface exploitation of clay in Serbia is considered to have degraded about 850 ha of land while less than 20 % has been recultivated [11].

EXPERIMENTAL PART

Geographical location, boundaries, and characteristics of the studied area

The studied area is a wider area of the settlement of Donje Crniljevo, Serbia, or the surroundings of the clayey material deposit "Zbegovi". In addition to the above deposit, there are two more deposits nearby: Jovanovića brdo and Galović (fig. 1). The settlement itself is in the municipality of Koceljeva, which is surrounded by the Kolubara basin in the east, Mačva, and Pocerina in the west, Srem in the north, mountain up-

Figure 1. Studied area and soil sampling sites

lands of Vlašić and Valjevo Podgorina in the south. It is in the southern part of Posavotamnava which, unlike other geographical wholes, is not clearly defined geographically. It is located on the M21 main road connecting Eastern Europe and the Southern Adriatic. It is 31 km away from Valjevo and 37 km from Šabac. The administrative, economic, and cultural center of the municipality of Koceljeva is the town of Koceljeva on the banks of the rivers Tamnava and Rasnica, on an average altitude of 200 meters [11].

The studied area is in the form of a rectangle (fig. 1.), measuring 17.8 km^2 in area and 17 km in circumference. It is southeast-northwest oriented, bordered by four points: A: 44°26 16.66 north latitude and 19°37 29.59 east longitude (far southern), B: 44°27 19.40 north latitude and 19°38 58.72 east longitude (far eastern), C: 44°29 27.08 north latitude and 19°36 57.33 east longitude (far northern), and D: 44°28 05.06 north latitude and 19°38 58.72 east longitude (far western).

In the studied area there are Donje Crniljevo settlement, the Tamnava river, M141 road, "Zorka" brick factory, Galovic kaolin drying plant as well as two prominent peaks of Vlašić (Jankov vis – 447 m above sea level and Razbojište – 465 m above sea level).

The area has a temperate continental climate. The average annual air temperature is 11.2 °C. The average annual rainfall is 720 mm^3 [11]. The average annual humidity is 76 %, with 1708 hours of sunshine a year and average annual cloudiness of 53 %, which has a very favorable effect on the richness of flora and fauna. Almost the entire municipality belongs to the Kolubara River basin, through its largest tributary Tamnava [11].

Geomorphology of the Zbegovi deposit, Donje Crniljevo, Serbia

In the 1950 s, in the area of the West Tamnava Tertiary Basin (Western Serbia), a large number of ceramic clay deposits were discovered and identified in cost-effective quantities [12]. The investigated deposits of plastic ceramic clays are small, and most are in the wider area of the village of Donje Crniljevo and represent the whole raw material base of quality ceramic clays in Serbia (fig. 2). Clays appear at depths of 2-35 m [12]. The productive Miocene series ends in different sands, gravels, and conglomerates. The lenticular appearance of clay at various levels of the Mio-

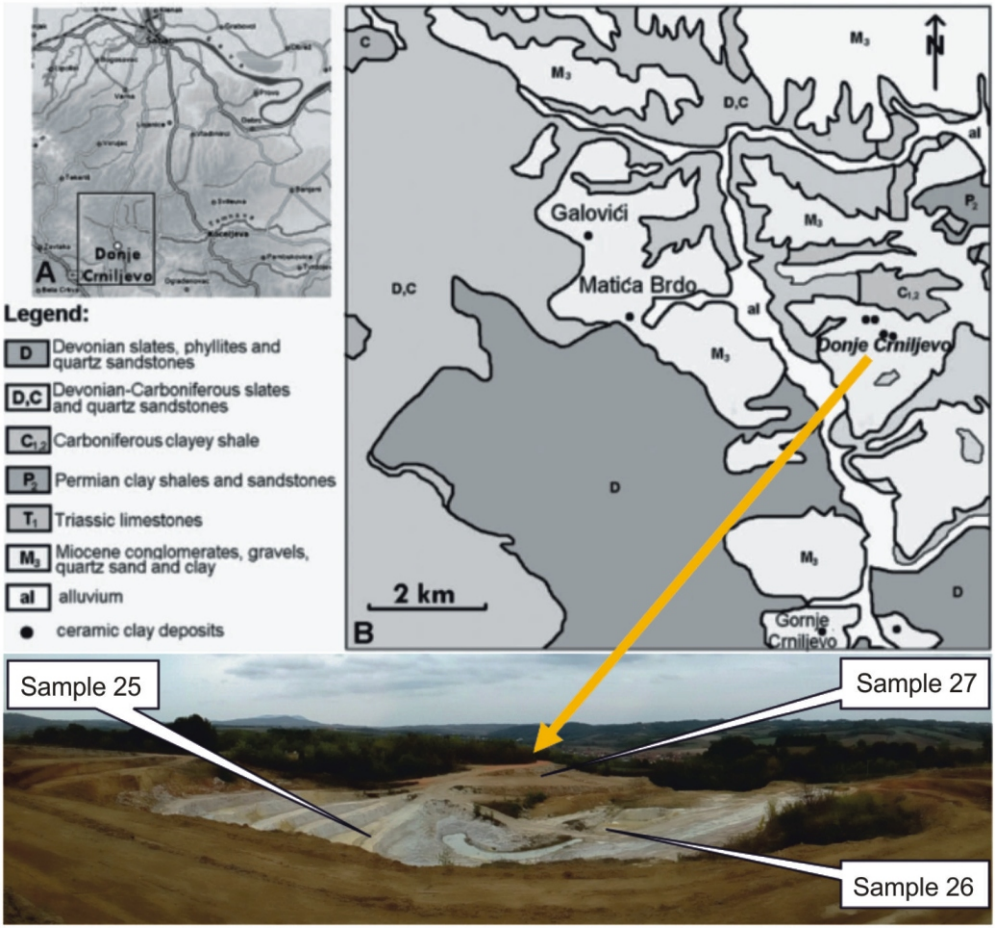

Figure 2. Geological map of the Tamnava tertiary basin and sampling site

cene series can be explained by the frequent oscillations of the Miopliocene waters, by basin bottom raising and deepening, *i. e*. by sediment deposition. Production of gray-white ceramic clay is about 100000 tonnes per year, with total reserves estimated at over 50 million tonnes [12, 13].

Based on mineralogical, physical, chemical and technological quality tests of a composite of excavated ceramic clays from active deposits in Donje Crniljevo [12]: Jovanovića Brdo, Bele Vode and Zbegovi (opened in 2007) four commercial clay types were separated:

– Red-yellow sandy-gravelly (brick clay).
– Gray-white poorly sandy (ceramic and partly refractory clay).
– Dark coaly (ceramic clay).
– Lamellar, greasy, and poorly sandy (highly aluminous and iron clay).

Brick clays are for internal use, the most important being gray-sandy ceramic clay, which was used to produce white facade bricks. All three types of ceramic clays from the Tamnava tertiary basin can be used for the wider non-metal processing industry, and these deposits give a full contribution to further economic development of this part of Southeast Europe [12]. By analyzing the obtained data on the mineralogical composition of the investigated ceramic clays from the Tamnava basin deposits, we may conclude that they differ from one another in granulometric composition (different sand, powder, and clay fraction ratios) and contain traces of other minerals (quartz, smectite clays, iron minerals, organic matter). Chemical analysis of clay samples shows a high content of SiO_2, moderate content of Al_2O_3, and low content of Fe, Ti, Ca, Mg, K and Na-oxide (tab. 1) [12].

The tailings pond of the "Zbegovi" open-pit mine in Donje Crniljevo is near the open-pit mine, occupying an area of several hectares and constantly increasing during exploitation.

Sampling and measurement methodology

Between August 2018 and the end of March 2019, within the limits of the studied space, 78 soil samples were collected and prepared [14, 15]. First samples of clay and clay-sand material were taken from the clay material deposit itself. The C25 sample of clayey material was collected from a depth of 20 m relative to the surrounding soil (fig. 2), and C26 sample of clayey-sandy material was taken from a depth of 25-30 m. Subsequently, tailing (C27) was sampled which lies north and continues to the open-pit mine forming a flat surface (fig. 2) and is flush with the surrounding land. Subsequently, 15 samples were taken nearby the open-pit mines, and then samples were collected from a wider area. The most characteristic sampling locations were (fig. 1): playground (C47), schoolyard (C46), churchyard (C48), the two highest peaks of Vlašić Mountain (C57 and C59), area near the M141 road (C19), the immediate vicinity of the brick factory (C28), the immediate vicinity of the Galović kaolin drying plant (C50), stream sediment below the deposit itself (C29) and sediment from the banks of the Tamnava River (C33). Of the 78 soil samples, 46 were samples from the location of non-arable land or the land assumed not to be contaminated with chemical preparations (artificial fertilizer, pesticides, herbicides, *etc*.), and the other 32 were from arable land. All samples were taken from a depth of 0-20 cm. Soil weight per sample was 2.5-3.5 kg.

The soil samples were dried at a temperature of 105 °C to constant weight and then all mechanical impurities were removed, mainly stone aggregate and parts of plant material, after which the samples were ground and homogenized [15]. For gamma ray spectrometric analysis, soil samples were sifted after homogenization through a 0.71 mm opening sieve and packed in Marinelli containers of 1000 ml in volumes which were, after measuring the sample's mass, hermetically sealed using a semi-transparent flexible film composed of a proprietary blend of waxes and polyolefins (parafilm). The typical weight of the samples ranged from 1020 g to 1560 g, and the mean density of the measured soil samples is 1.26 0.01 gcm^{-3}. Before gamma spectrometric measurements Marinelli containers were left for forty days to establish a radioactive balance between ^{226}Ra and its descendants.

Gamma spectrometric analysis of the samples was performed using standard methods according to ISO 18598 standard [16-18].

The prepared soil samples were measured on a semiconductor HPGe spectrometer, manufactured by "AMETEK-ORTEC", 1.78 keV resolution (FWHM), and 56.2 % relative efficiency for ^{60}Co at 1332 keV.

The detector was housed in a special low-background lead protection with 12 cm thick walls and 5 mm thick copper inner layer. All samples were measured for 60000 seconds to obtain the appropriate statistics of counting speed in the spectrum.

Detector efficiency calibration was performed using the radioactive standard in 1000 ml Marinelli container, MBSS type, Inspectorate for ionizing radiation Czech metrological institute (Cert. No: 9031-OL-159/08) whose matrix is silicone resin with radionuclides: ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{133}Ba, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y, ^{54}Mn, and ^{60}Co. Spectrum analyzes were done based on the present gamma lines.

Table 1. Chemical composition of ceramic clay sample [12]

Component	Annealing loss	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	Na_2O	K_2O
Content [%]	10.7	61.9	21.6	1.17	1.01	1.05	0.46	0.07	2.05

Gamma lines ^{234}Th at 63 keV and 92.3 keV were used to determine the specific activity of ^{238}U; for ^{226}Ra, ^{214}Bi gamma lines at 609.3 keV and ^{214}Pb at 351.9 keV were used. For determining the specific activity of ^{232}Th, ^{228}Ac gamma lines at 911.2 keV and 969.1 keV and ^{212}Pb line at 238.6 keV were used. The specific activity of ^{40}K was determined based on the line at 1460.8 keV, and the specific activity of ^{137}Cs was determined based on the line at 661.66 keV, using the Gamma Vision 32 software package [19]. Obtained values of counts below the peaks of the observed lines were adjusted to the background measured at 250000 seconds.

To assess the radiation risk to the population living in Donje Crniljevo in the vicinity of the open-pit mines the strength of absorbed dose, annual effective dose, cancer risk factor, and radiation risk index were calculated. The radiation risk to biota was also assessed.

Absorbed gamma dose rate

The external absorbed dose rate ($\dot{D}$ (nGyh^{-1})) outdoor at 1 m above the ground level was calculated using the eq.

$$\dot{D} \quad 0.462A_{\mathrm{U}} \quad 0.604A_{\mathrm{Th}} \quad 0.042A_{\mathrm{K}} \qquad (1)$$

where the indexes: 0.462, 0.604, and 0.0417 are the dose conversion factors for ^{238}U, ^{232}Th, and ^{40}K respectively in soil (nGyh^{-1} per Bqkg^{-1}) [20], and A_{U}, A_{Th}, and A_{K} are the activity concentrations (Bqkg^{-1}) of ^{238}U, ^{232}Th, and ^{40}K.

Annual outdoor effective dose

To estimate the health effects of the absorbed dose, the annual effective dose rate (H(Sv)) should be determined taking into account the conversion coefficients from the absorbed dose in the air to the effective dose (0.7 SvGy^{-1}) and the outdoor occupancy factor (the fraction of time spent outdoors) of 0.2 both proposed by The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). The annual effective dose rate in units of Sv was estimated using the following formula [20]

$$H \quad \dot{D} \; N_{\mathrm{h}} \; 0.2 \; 0.7 \qquad (2)$$

where $\dot{D}$ is the absorbed gamma dose rate in the air (nGyh^{-1}) and $N_{\mathrm{h}} = 8760$ is the number of hours in one year.

Excess lifetime cancer risk outdoors

Excess lifetime cancer risk outdoors, F_{rk}, was calculated as follows

$$F_{\mathrm{rk}} \quad H \; DL \; P \qquad (3)$$

where H is the annual outdoor effective dose (Sv), DL the life expectancy (70 years), and P the nominal probability coefficient for detriment-adjusted cancer risk of 0.055 Sv^{-1} for the whole population [21].

External radiation hazard index

The external radiation hazard index, H_{ex}, is used to estimate the external radiation hazard due to the emitted gamma radiation. It was calculated [20] according to the following equation

$$H_{\mathrm{ex}} \quad A_{\mathrm{U}} / 370 \quad A_{\mathrm{Th}} / 259 \quad A_{\mathrm{K}} / 4810 \qquad (4)$$

where A_{U}, A_{Th}, A_{K} are the activity concentrations (Bqkg^{-1}) of ^{238}U, ^{232}Th, and ^{40}K, respectively, from soil samples.

The value of this index must be less than unity to keep the radiation hazard insignificant, *i. e*. to keep the radium equivalent activity and annual dose under the permissible limits of 370 Bqkg^{-1} and 1 mSv, respectively.

Assessment of radiation risk to biota using the RESRAD-BIOTA computer code

The concentration of radionuclides in the environment increases with performing industrial activities (air, water, as well as their accumulation in soil and sediment). The commercially available RESRAD-BIOTA code [22] provides a complete range of possibilities for estimating the impact of ionizing radiation on biota, from methods for general review to a comprehensive assessment of the receptor-specific absorbed dose.

The code calculates the absorbed radiation dose due to external and internal exposure. Living space of the organism and the time it spends in the contaminated environment is considered when calculating the dose due to external exposure. When calculating a dose due to internal exposure intake of the radionuclide by inhalation of dust particles and ingestion of contaminated soil, sediment and various sources of food are considered. The code contains three levels of assessment. Level 1 – is the screening level at which default values for the bioaccumulation factor (Biv) are used to determine Biota Concentration Guide BCG screening. The BCG is the ratio of the limit dose (which for land plants and animals is 10 and 1 mGyd^{-1}, respectively) and dose conversion factor due to external and internal exposure [23, 24]. The bioaccumulation factor is used to determine, from the activity concentrations of a given environment, the activity concentration for biota (whole organism). In level 1, if the maximum value of the tested environment is less than the BCG screening value and the sum of all coefficients is less than 1, it is considered that there is no negative effect of radiation on the biota of the tested area. In levels 2

and 3, in addition to results for BCG, it is possible to estimate radiation dose strengths for organisms analyzed, at these levels, it is possible to perform sensitivity analysis for the used parameters, but an allometric approach can be included as well. The equations used in allometric calculations calculate biological half-lives, the amount of taken food, the amount of taken soil, inhalation, and maximum lifespan. Level 3 allows the user to create a new organism and perform a probabilistic assessment.

RESULTS AND DISCUSSION

Activity concentrations of radionuclides in soil samples

For all 78 soil samples, based on measurements of the spectrum obtained, activity concentrations of radionuclides ^{238}U, ^{232}Th, ^{40}K, and ^{137}Cs were calculated considering the count in the peak of a given energy (N), sample measurement time, t_m, sample mass, m, spectrometer efficiency at a specific peak energy, ε and yield of specific energy, p

$$A_S = \frac{N}{(p\varepsilon t_m m)} \quad (4)$$

For all samples, the count in the peak of given energy was adjusted to the background. ^{238}U, ^{232}Th, and ^{40}K radionuclide activity concentration results obtained for all 78 soil samples are presented graphically in fig. 3.

Using the Statistical Package for the Social Sciences software package (SPSS) [25], we calculated: mean, standard deviation, minimum and maximum values presented in tab. 2 for ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K,

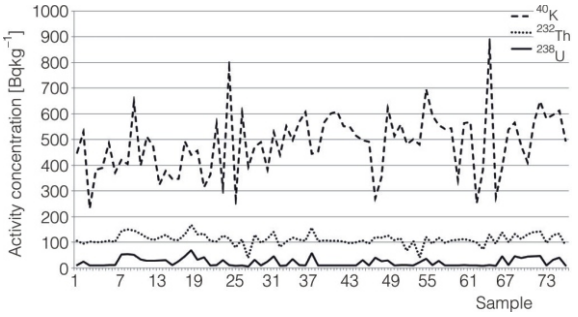

Figure 3. Activity concentrations of radionuclides in the studied area soil

Table 2. Descriptive radionuclide activity concentrations statistic in soil of the studied area for the confidence level $k = 1$

Radionuclide/ parameter	Activity concentration [Bqkg^{-1}]				
	^{238}U	^{232}Th	^{40}K	^{226}Ra	^{137}Cs
Middle value	23 8	89 3	372 7	56 2	11 1
Minimum	<7	18	129	<2	<0.01
Maximum	70	120	739	98	27

and ^{137}Cs activity concentrations in soil and clayey material samples.

The measured activity concentrations do not deviate from the average values for soil in Serbia [26]. Comparing the obtained values of concentrations with the average world values given by UNSCEAR (400 Bqkg^{-1} for ^{40}K, 35 Bqkg^{-1} for ^{226}Ra, and 30 Bqkg^{-1} for ^{232}Th [27]) it can be seen that these values are higher at ^{232}Th and ^{226}Ra. The highest activity concentrations of ^{232}Th (120 4 Bqkg^{-1}) and ^{40}K (739 13 Bqkg^{-1}) are measured at location 66 (Vlašić-Vinogradić), a site about 1.5 km southwest away from the open-pit mine. Comparing the obtained results with those of other studies, it may be concluded that the kaolin and sediment deposits of the studied area, as well as the surrounding land, are slightly richer in thorium [27]. The highest concentration of ^{226}Ra was 98 1 Bqkg^{-1} at location number 70, which belongs to arable land and at the time of sampling (March 17, 2019) was treated with artificial fertilizer.

As for the highest concentration of ^{137}Cs, it was at location number 78 and amounted to 27 1 Bqkg^{-1}, in forest covered land, at higher altitude and in the dominant wind direction, which coincides with the results obtained in other studies [5]. In the graphs (fig. 3.) showing the concentrations of radionuclides at sampling locations, the two locations with the lowest concentrations of ^{232}Th and ^{226}Ra (location 28 and 55) can be clearly seen. The first location is near the brick factory and the other at the perimeter of the Galović open-pit mine. At both sampling sites there was sandy material taken from the mine (now in the capacity of a terrain leveling embankment), with low vegetation and shrubs already beginning to emerge at location 28. The lowest value of ^{40}K (129 4 Bqkg^{-1}) was measured in the sample of arable land taken from location no. 3, which is in the immediate vicinity of the "Zbegovi" open-pit mine. Three times lower concentration of ^{40}K measured in the surrounding land of the "Zbegovi" mine which is reached by dust from the mine that deposits in it, shows that there is no increase in the natural radioactivity of the surrounding soil caused by the impact of dust raised during the surface exploitation of clay.

Artificial radionuclide ^{137}Cs had the lowest specific activity value in the sample No. 55 (the perimeter of the clayey material open-pit mine Galović), which was 0.6 0.1 Bqkg^{-1}. To further analyze the results obtained for samples of clay material taken on the "Zbegovi" open-pit mine and tailings (fig. 2.), activity concentrations of ^{238}U, ^{232}Th, ^{40}K, ^{226}Ra, and ^{137}Cs radionuclides of these samples are given in a separate table (tab. 3).

Limits of radionuclide content in building materials where clay is used for interior, amount to: for radium (^{226}Ra): 3 10^2 Bqkg^{-1}, for thorium (^{232}Th): 2 10^2 Bqkg^{-1}, for potassium (^{40}K): 3 10^3 Bqkg^{-1}, respectively [10]. No measured radionuclide activity concentration exceeded the maximum permissible value and mean activity con-

Table 3. Activity concentrations of radionuclides in clayey material of "Zbegovi" Donje Crniljevo, Serbia

Sample	Activity concentration[Bqkg^{-1}]				
	^{238}U	^{232}Th	^{40}K	^{226}Ra	^{137}Cs
C25	<11	103 3	668 12	59 2	<0.01
C26	<9	70 2	190 5	59 2	<0.01
C27	<10	100 3	493 9	66 2	<0.01
Middle value	<10	91 3	450 9	61 2	<0.01

centrations do not deviate from the average values for clayey material (tab. 3).

Riekstina *et al.* [8] have shown that activity concentrations of ^{238}U, 2^{32}Th, and ^{40}K in clays of Lithuania amounted to: 35, 52, and 1078 Bqkg^{-1} respectively, while Todorović *et al*. [9] showed that different clays on the Serbian market had activity concentrations of 89 and 1303 Bqkg^{-1} for ^{232}Th and ^{40}K, respectively. Comparing the data for the clayey material of previous studies (tab. 4) with the data obtained in this paper (tab. 3), it can be concluded that the values are of the same order of magnitude. By activity concentrations of ^{238}U, ^{232}Th, and ^{226}Ra, this clayey material is closest to the clays of Serbia (Clay for indoor use "Riznica prirode"). Somewhat lower ^{40}K concentration values in this paper, compared to clays from the Serbian market, can be interpreted by different way of preparing a clayey material sample, or by measuring a sample that has not undergone industrial processing. Measured concentrations of radionuclides in the sample of clayey material from the mine correspond to the values measured in the ceramic tile as building material [28]. Artificial radionuclide ^{137}Cs was below the detection limit in all three clayey material samples (tab. 3) and it can be concluded that the openpit mines of clayey material, along with the tailings dump, are uncontaminated soil surfaces in terms of artificial radionuclides. No data were found in the available literature about concentrations of radionuclide activity in the surroundings of clay deposits.

In order to validate the results obtained for specific activities, activity concentration conversion was performed: ^{238}U, ^{232}Th, and ^{40}K to mass concentrations of uranium, thorium and potassium in soil [27, 28]. The following values were obtained: from 0.6 mgkg^{-1} to 5.7 mgkg^{-1} for uranium (mean value 1.9 mgkg^{-1}); from 4.4 mgkg^{-1} to 29.5 mgkg^{-1} for thorium (mean value 22 mgkg^{-1}) and from 0.4 % to 2.4 % for potassium (mean value 1.2 %). The obtained values are in agreement with previous investigations [29-35].

Assessment of radiation risk to population

Risk indices and absorbed dose rate of γ-radiation due to external exposure on the studied area were assessed using the mean of activity concentrations of ^{238}U, ^{232}Th, and ^{40}K soil samples. Descriptive statistics of the absorbed dose rate D, annual outdoor effective dose H, excess lifetime cancer risk outdoors F_{rk}, and external radiation hazard index H_{ex}, on the population of the studied area from 78 locations are shown in tab. 5.

Equation (1) was used to calculate the e absorbed dose rate of γ-radiation in air at a height of one meter above ground, which originates from ^{238}U, ^{232}Th, and ^{40}K. Calculated values of the absorbed dose rate range from 38.3 to 109 nGyh^{-1}, while the mean at 78 locations of the studied area is 80.1 nGyh^{-1}. According to data from the literature, the value of the absorbed dose rate at a height of one meter above ground in normal conditions is 60 nGyh^{-1}, while for different territories these values vary from 10 to 200 nGyh^{-1} [27]. The mean value of the absorbed dose rate for the territory of Belgrade is 60.5 nGyh^{-1} [20]. Higher values of absorbed dose rate obtained in this research can be interpreted by the diversity of the parent rocks from which the soil of the studied area originated relative to the soils in the above studies, that is, that slightly higher values of thorium were measured [3, 36]. Obtained absorbed dose rate values for all 78 locations of the studied area are presented in fig. 4.

Based on eqs. (2) and (3) and the measured activity concentrations of ^{238}U, ^{232}Th, and ^{40}K, annual outdoor effective dose and excess lifetime cancer risk out-

Table 4. Mean concentrations of radionuclide activity [Bqkg^{-1}] in clays in various parts of the world and in Serbia

S.N.	Country	Activity concentration [Bqkg^{-1}]				
		^{238}U	^{232}Th	^{40}K	^{226}Ra	Literature
1	Turkey	39.3	49.6	567	/	[6]
2	Nigeria	38.2	65.1	93.9	/	[3]
3	India	9.19	45.6	295	/	[3]
4	Lithuania (Devonian clay)	34.8	51.6	1078	/	[8]
5	Lithuania (Quaternary clay)	10	14.2	553	/	[8]
6	Iraq	101	87.5	305	/	[3]
7	Serbia, Clay for external use ("Riznica prirode")	/	72 1	1055 5	59 2	[9]
8	Serbia, Clay for internal use ("Riznica prirode")	/	89 2	1303 2	72 1	[9]
9	Serbia, Natural green clay for external use (YOGA glina)	/	71 6	962 9	65 4	[9]
Middle value		25.8	61.2	690	65.3	

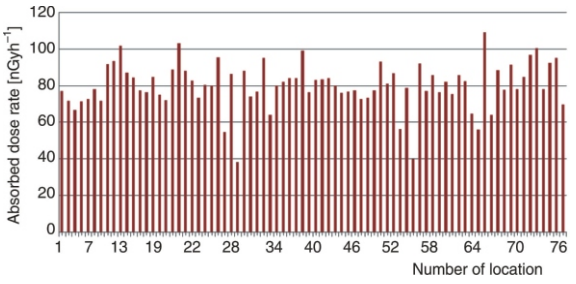

Figure 4. The absorbed dose rate at the sampling locations of the studied area

Table 5. Descriptive statistics of the absorbed gamma dose rate, annual outdoor effective dose, excess lifetime cancer risk outdoors and external radiation hazard index due to exposure to soil radionuclides

Parameter	Middle value	Standard deviation	Min	Max
$\dot{D}$ [nGyh^{-1}]	80.1	12.4	38.3	108.9
H [Sv]	98.2	15.3	46.9	133.5
F_{rk} (10^{-4})	3.8	0.6	1.8	5.1
H_{ex}	0.48	0.01	0.22	0.65

doors due to exposure to radionuclides from soil were calculated. Descriptive statistics of these parameters obtained based on the results of measuring the activity concentrations of samples from 78 locations is shown in tab. 5. Obtained values of annual outdoor effective dose in the studied area vary in the range 46.9-134 Sv. Worldwide annual outdoor effective dose, estimated on the basis of radionuclides from soil, is 70 Sv [37].

In the studied area, in the vicinity of Donje Crniljevo, the mean value of the estimated annual outdoor effective dose is 98.2 Sv, which is higher than the world average due to the characteristic of the soil with slightly higher thorium concentration. [3, 36]. The calculated mean value of the excess lifetime cancer risk outdoors is 3.8 10^{-4} and is consistent with the world average of 2.9 10^{-4} [38]. The resulting value can be interpreted in a way that the possibility of getting cancer, as a consequence of the negative impact of radionuclides from the soil, in the population in the vicinity and in the settlement of Donje Crniljevo itself, is slightly increased compared to the value for the territory of Belgrade, Serbia, which is 2.8 l0^{-4} [20] and is lower relative to the Kirklareli area, Turkey, where it amounts to 5.0 10^{-4} [38]. Based on activity concentrations of ^{238}U, ^{232}Th, and ^{40}K using eq. (4), the radiation risk index for the population due to natural exposure to radionuclides from soil was calculated. The mean of the external radiation hazard index obtained in this research is 0.48 and is not considered significant due to external exposure since it is less than one. The range of these index values of 0.22-0.65 indicates that in the studied area there is no significant radiation risk to the population due to exposure to natural sources of radiation from the soil. External radiation hazard index due to exposure to radionuclides from soil calculated based on radionuclide concentrations in soil in a study conducted at kaolin mines in Ifonyintedo, Nigeria, amounts to 0.48 [3] which is consistent with the results obtained in this study.

Assessment of radiation risk to biota

The legal framework prescribed by the US Department of Energy (DOE), rely on a technical standard-setting the limits for biota, the application of which limits the negative impact of ionizing radiation on the population of plants, animals, fungus and bacteria [23, 39]. The absorbed dose rate for aquatic animals and terrestrial plants should not exceed 10 mGyd^{-1}, and for terrestrial animals 1 mGyd^{-1} from exposure of radionuclides in soil. In this paper, analyzes were performed at Level 2 RESRAD BIOTA, version 1.8 (location-specific screening), using reference terrestrial organisms found in the software (terrestrial plants and terrestrial animals) [22-24].

Assessment of radiation risk to biota was done using RESRAD BIOTA software 1.8, Level 2, that is, location-specific screening (the studied area) were more realistically related parameters representative of the location are used [22, 23]. Calculated mean activity concentrations (Bqkg^{-1}) ^{238}U, ^{234}Th, ^{40}K, ^{226}Ra, and ^{137}Cs of soil (tab. 2) were the input data for estimating the absorbed dose rate for the reference terrestrial organisms: plants (lichens, bryophytes, grasses, shrubs, and trees) and animals (amphibians, reptiles, earthworms, flying insects, saprophyte invertebrates, birds, large and small mammals), which are by default found in the RESRAD BIOTA, in the studied area, also taking into account the time dependence and spatial extent of the contamination.

The resulting total ratio was 0.13 (<1), which means that the absorbed dose rate is below the allowed for terrestrial plants and animals and does not pose a threat to biota. After screening there was no need to move to the following level of analysis, that is, concentrations of radionuclides were below the recommended, *i.e*. irradiation of the biota in the studied area is within the dose limit.

Based on the radionuclide activity concentrations in the soil of the investigated area, using the computer code RESRAD-BIOTA, the total absorbed dose rate due to external and internal exposure was calculated, as well as absorbed dose rates for individual radionuclides for reference plants and animals. The obtained values are given in tab. 6 and graphically shown in fig. 5. The largest contribution to the total absorbed dose rate is given by ^{40}K and ^{226}Ra. For external exposure of biota originating from radionuclides from the soil, the obtained value of the absorbed dose rate was 5.89 10^{-6} Gyd^{-1} (tab. 6, fig. 5). The value of the absorbed dose rate for internal exposure for animals was 1.25 10^{-4} Gyd^{-1}, and for plants 1.22 10^{-4} Gyd^{-1} (tab. 6, fig 5). The total absorbed dose rate was 1.31 10^{-4} Gyd^{-1} for animals, or 1.28 10^{-4} Gyd^{-1} for plants (tab. 6, fig. 5.), which is within the recommended biota exposure limits (less than 1 10^{-3} Gyd^{-1} for terrestrial animals, or less than 1 10^{-2} Gyd^{-1} for plants) [22].

Table 6. Absorbed dose rate due to external and internal exposure and the total absorbed dose rate due to exposure to ^{137}Cs, ^{40}K, ^{226}Ra, ^{234}Th, and ^{238}U from the soil for reference animal and plants on the studied area

Nuclide	$\dot{D}_{ext}$ [Gyd^{-1}]		$\dot{D}_{int}$ [Gyd^{-1}]		$\dot{D}_{tot}$ [Gyd^{-1}]	
	Plant	Animal	Plant	Animal	Plant	Animal
^{137}Cs	1.21 10^{-7}	1.21 10^{-7}	1.23 10^{-6}	1.42 10^{-5}	1.35 10^{-6}	1.43 10^{-5}
^{40}K	3.47 10^{-6}	3.47 10^{-6}	6.94 10^{-5}	8.12 10^{-5}	7.29 10^{-5}	8.47 10^{-5}
^{226}Ra	2.09 10^{-6}	2.09 10^{-6}	5.05 10^{-5}	2.78 10^{-5}	5.26 10^{-5}	2.99 10^{-5}
^{232}Th	1.48 10^{-8}	1.48 10^{-8}	1.01 10^{-6}	1.59 10^{-6}	1.03 10^{-6}	1.60 10^{-6}
^{238}U	2.90 10^{-7}	2.90 10^{-7}	1.05 10^{-7}	1.04 10^{-7}	3.95 10^{-7}	3.94 10^{-7}
Sum	5.98 10^{-6}	5.98 10^{-6}	1.22 10^{-4}	1.25 10^{-4}	1.28 10^{-4}	1.31 10^{-4}

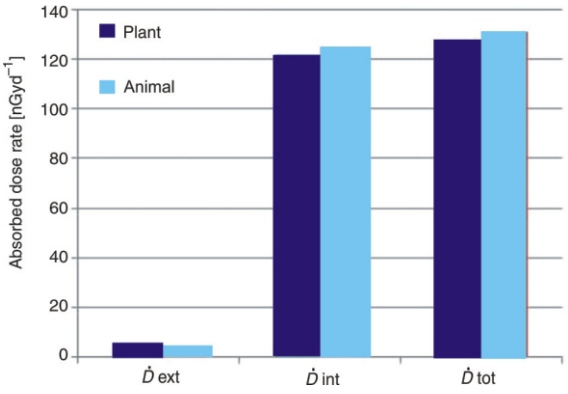

Figure 5. Graphical representation of internal, external and total absorbed dose rate (Gyd^{-1}) for plants and animals from radionuclides from soil

CONCLUSIONS

The results presented in this paper were obtained based on measurement and analysis of the content of radionuclides in soil collected at 78 locations at a depth of 0-20 cm, in the period from 2018 to 2019, in the vicinity of Donje Crniljevo, Serbia, or clayey material deposit "Zbegovi":

- Mean values of activity concentrations (Bqkg^{-1}) of the analyzed radionuclides determined by gamma-spectrometric method in the studied area are as follows: 23 8 Bqkg^{-1} for ^{238}U; 89 3 Bqkg^{-1} for ^{232}Th; 372 7 for ^{40}K; 56 2 Bqkg^{-1} for ^{226}Ra and 11 0.5 Bqkg^{-1} for ^{137}Cs.
- Obtained values of activity concentrations do not deviate from the average values for the soil in Serbia and the Republic of North Macedonia [10, 26, 40]. Comparing data from previous studies with data obtained in this paper for clayey material, it can be concluded that the values are of the same order of magnitude [3, 6].
- In samples of clayey material and soil from the open-pit mine itself, ^{137}Cs concentrations are below detectable concentrations.
- The absorbed gamma dose rate in the air at a height of one meter above the ground, which originates from ^{238}U, ^{232}Th, and ^{40}K is in the range 38.3-110 nGyh^{-1}, while the obtained mean at 78 locations of the studied area is 80.1 nGyh^{-1}. That value is slightly higher than the mean value of absorbed dose rate for the territory of Belgrade (60.5 nGyh^{-1} [20]) but is within the limits specified by absorbed dose rate values determined at different locations (10-200 nGyh^{-1} [27]). Higher absorbed dose rate values obtained in this study can be interpreted in that the soil has slightly more thorium [3, 38]. Obtained annual outdoor effective dose values in the studied area are in the range 46.9-133 Sv. The mean annual outdoor effective dose is 98.2 Sv and is slightly higher in relative to the world average, which is 70 Sv [27].
- The mean excess lifetime cancer risk outdoors obtained is 3.8 10^{-4}. In a population in the vicinity and the settlement of Donje Crniljevo, the excess lifetime cancer risk outdoors value is higher relative to the value for the territory of Belgrade (2.8 l0^{-4} [20]) and the world average (2.9 10^{-4} [39]), but significantly lower than that of the Kirklareli area, Turkey (5.0 10^{-4} [38]).
- The mean value of the external radiation hazard index obtained by this research is 0.48. All obtained values are in the range of 0.22-0.65 indicating that there is no significant radiation risk to the population on the studied area.
- Using the computer code RESRAD BIOTA version 1.8, level 2, absorbed radiation dose rate was calculated and radiation risk assessed for the biota of the studied area. The reference organisms (animals and plants) that are by default found in the program were used in the assessment. External exposure of the biota originating from radionuclides from soil was 1.31 10^{-4} Gyd^{-1} for animals and 1.28 10^{-4} Gyd^{-1} for plants, which is within the limits of the recommended exposure of biota, less than 1 mGyd^{-1} for animals, that is, less than 10 mGyd^{-1} for plants [22].

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AUTHORS' CONTRIBUTIONS

Theoretical analysis was carried out by S. B. Ilić, M. M. Djurašević, A. B. Kandić, and T. D. Golubović. Samples were collected by S. B. Ilić and N. D. Pajić.

All authors analyzed and discussed the results. The manuscript was written by S. B. Ilić and translation of the paper made by M. M. Djurašević and N. D. Pajić.

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ИСПИТИВАЊЕ САДРЖАЈА РАДИОНУКЛИДА У УЗОРЦИМА ЗЕМЉИШТА И РАДИОЛОШКИ РИЗИЦИ У ОКОЛИНИ ЛЕЖИШТА ГЛИНОВИТОГ МАТЕРИЈАЛА "ЗБЕГОВИ" ДОЊЕ ЦРНИЉЕВО, СРБИЈА

У раду су приказани резултати испитивања садржаја радионуклида у узорцима околног земљишта и глиновитог материјала површинског копа "Збегови" Доње Црниљево, Србија. Прикупљени су и припремљени узорци са 78 локација и одређене су концентрације активности радионуклида: ^{238}U, ^{232}Th, ^{40}K, ^{226}Ra, ^{137}Cs. Добијене средње вредности су: 23 Bqkg^{-1}, 89 Bqkg^{-1}, 372 Bqkg^{-1}, 56 Bqkg^{-1}, и 11 Bqkg^{-1}, редом. Концентрације ^{238}U, ^{40}K и ^{226}Ra на истраживаном простору не одступају од вредности које су добијене за земљишта у Србији. Концентрација ^{232}Th на истраживаном простору има незнатно више вредности у односу на просечне вредности за земљишта, а нешто мање у односу на слична лежишта глиновитог материјала у свету. Извршена мерења су показала да је површински коп глиновитог материјала потпуно неконтаминирана површина што се тиче ^{137}Cs, док постоје локације на којима су измерене концентрације ^{137}Cs значајно веће, а што је последица топографских разлика и нехомогене површинске контаминације земљишта после акцидента у Чернобиљу.

Да би се проценили радиолошки ризици на посматраном простору одређени су: јачина апсобоване дозе, годишња ефективна доза, апсорбована доза за биоту, фактор ризика појаве канцера за становништво као и индекс радијационог ризика. Средња вредност процењене јачине апсорбоване дозе на датом простору износи 80.1 nGy h^{-1}, а годишња ефективна доза варира у опсегу 46.9 до 134 Sv. Јачина апсорбоване дозе за биоту на истраживаном простору износи 1.31 10^{-4} Gyd^{-1}. Средња вредност фактора ризика појаве канцера за становништво износи 3.8 10^{-4}, а средња вредност индекса радијационог ризика добијена овим истраживањем износи 0.48, што је у складу са светским просеком. Ниска доза зрачења неће представљати ризик за становништво и биоту на истраживаном простору.

Кључне речи: глина, радиоактивност, гама спектрометрија, ^{238}U, ^{232}Th, ^{40}K, ^{226}Ra, ^{137}Cs, радиолошки ризик