NATURAL RADIONUCLIDES IN SOIL PROFILES SURROUNDING THE LARGEST COAL-FIRED POWER PLANT IN SERBIA

by

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This study evaluates the influence of the largest Serbian coal-fired power plant on radionuclide concentrations in soil profiles up to 50 cm in depth. Thirty soil profiles were sampled from the plant surroundings (up to 10 km distance) and analyzed using standard methods for soil physicochemical properties and gamma ray spectrometry for specific activities of natural radionuclides (^{40}K, ^{226}Ra and ^{232}Th). Spatial and vertical distribution of radionuclides was determined and analyzed to show the relations between the specific activities in the soil and soil properties and the most influential factors of natural radionuclide variability were identified. The radiological indices for surface soil were calculated and radiological risk assessment was performed. The measured specific activities were similar to values of background levels for Serbia. The sampling depth did not show any significant influence on specific activities of natural radionuclides. The strongest predictor of specific activities of the investigated radionuclides was soil granulometry. All parameters of radiological risk assessment were below the recommended values and adopted limits. It appears that the coal-fired power plant does not have a significant impact on the spatial and vertical distribution of natural radionuclides in the area of interest, but technologically enhanced natural radioactivity as a consequence of the plant operations was identified within the first 1.5 km from the power plant.

Key words: natural radioactivity, gamma ray spectrometry, soil property, principal component analysis, radiological risk, dose assessment

INTRODUCTION

The mineralogy of the parent material mainly determines the content of naturally occurring radionuclides in soil, and soil physicochemical properties further influence their mobility and bio accessibility in terrestrial ecosystems [1]. The content of natural radionuclides in soil can be altered by human activities through the release of effluents containing different amounts of radioactive elements and their later deposition. In a situation where the human factor is present, the influence of particular local factors governing radionuclide distribution cannot be easily distinguished, since the level of natural background radiation varies substantially even within the same region [2].

Coal is an abundant natural resource that plays an important role in the world's energy production. It belongs to naturally occurring radioactive materials (NORM) and coal mining and coal combustion are classified as a source of technologically enhanced naturally occurring radioactive materials (TENORM) [3]. Coal combustion releases natural radionuclides from the coal matrix and leads to their enhanced concentration in combustion by-products and radionuclides are redistributed between ash, fine particles and gases that are released into the atmosphere [4, 5]. In general, the radionuclide enrichment factor in ash is about 10 [6]. Such emitted radionuclides may reach soil through dry or wet deposition or by runoff from ash deposits modifying the natural radioactivity background level. In recent years, the number of investigations related to the potential influences of the high level of natural radionuclides over areas affected by the coal-fired power plant (CFPP) operation has greatly increased [7-11]. The researchers have been mainly focused on the distribution of natural radionuclides in soil profiles under the influence of CFPP [12-17].

The CFPP represent 64.1 % of the total installed capacities in the electric power industry of Serbia [18]. Serbian energy reserves consist of various types of coal, predominantly low-quality lignite, with a share of over

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95 % in the total balance sheet reserves. Although some studies has been conducted on the impact of CFPP in Serbia on the surrounding soils and water in terms of natural radioactivity enhancement and radiological hazard evaluation [19-21], and distribution of natural radionuclides in soil from areas affected by coal production [22, 23], there is a scarcity of data describing the distribution of natural radionuclides in soil profiles selected from areas under the influence of CFPP [24].

The objectives of this study were to: (1) determine the specific activities of ^{40}K, ^{226}Ra, and ^{232}Th in the soil profiles surrounding the largest CFPP in Serbia; (2) investigate the spatial distribution of those radionuclides and their variation at different depths; (3) asses the relationships between specific activities of natural radionuclides and measured soil properties; (4) estimate the radiological hazard associated with gamma radiation of natural origin.

MATERIALS AND METHODS

Study area

The CFPP "Nikola Tesla A", with a total installed capacity of 1650 MW in its six units, is the most important thermal power plant in of the electric power industry of Serbia. It is located on the right bank of the Sava River in the vicinity of Obrenovac, 35 km upstream from Belgrade. Its first unit was commissioned in 1970, and since then the CFPP has been in permanent operation [25].

The feed coal for the CFPP is lignite from the open pit mines of the "Kolubara basin" with an average consumption of 2510 t per hour [25]. The mean content of ^{40}K, ^{226}Ra, and ^{232}Th in the lignite is 123 Bq/kg, 28 Bq/kg, and 22 Bq/kg, respectively [26]. Annual ash generation from the CFPP is approximately 2.4 million tons of which 80-85 % is fly ash, 15-20 % bottom ash and 0.2-2.0 % unburned coal. The reaction of ash is alkaline (7.7-7.9) [27]. There are two ash disposal sites. The first is located in the vicinity of the CFPP "Nikola Tesla A" within a 1 km distance in the north and covers 400 ha. The second is the ash repository settled 10 km away from CFPP in the southwestern part of the study area with a surface area of 600 ha, and it belongs to the CFPP "Nikola Tesla B".

Calcaric fluvisols (alluvial soil) with a loamy-clay texture is the most common soil type in the study area. The other soils are represented by vertisols, gleyosols, phaeozems, stanic gleyosols, cutanic cambisols, and humic gleyosols. As a direct result of CFPP activity, technosols and spolic regosols are also present [28].

The study area is characterized by a moderate continental climate with mean monthly temperature ranges from 0.1 °C in January to 22.1 °C in July. Annual precipitation is 670 dm^3/m^2. The mean relative humidity is 70 %, and monthly local pressure is 1001 mb [29]. Wind is a very important element of climate for the study area. The prevailing winds are from the south-southeast, west-northwest, southeast, and west directions with a relative frequency of 121 %, 106 %, 105 %, and 99 %, and annual speed of 2.9 m/s, 2.3 m/s, 3.0 m/s, and 2.2 m/s, respectively. Ash deposits under the influence of the western and north-western wind directly threaten the study area. Nonetheless, the most frequent south-eastern wind brings pollutants from the lignite open pit mine to the entire area of interest.

The study area is densely populated with more than 64000 inhabitants in 18 settlements (Obrenovac and 17 villages) according to the 2011 census.

Soil sampling

Sampling was conducted from the summer of 2011 to the spring of 2013 at 30 locations and was performed based on established procedures [30, 31]. The distances between the CFPP and sampling locations were approximately 1, 2, 4, 6, 8, and 10 km in the west (W), southwest (SW), south (S), southeast (SE), and east (E) direction. Each sampling site was georeferenced by a hand-held GPS device (locations of the sampling point are marked in fig. 2). The surface layer covered by vegetation and debris was removed prior to s oil sampling. The vertical soil profile (50 cm 50 cm 50 cm) was then opened using a spade, after which samples were taken carefully from every 10 cm from the upper most layer down to a 50 cm depth. The layers were labeled as follows: *a* (0-10 cm), *b* (10-20 cm), *c* (20-30 cm), *d* (30-40 cm), and *e* (40-50 cm). To ensure the representativeness of the specimen, four subsamples for each layer (vertices of 2 m 2 m rectangle) were taken from each location and thoroughly mixed to obtain a bulk composite sample. Approximately, 2 kg of soil from each layer at a single location were enclosed in a plastic bag and transported to the laboratory.

Sample preparation and determination of soil properties

The stones, visible roots and other debris were removed from the soil samples in the laboratory, upon which they were air dried at room temperature for two weeks. Air-dried soil samples were oven dried to a constant mass at 60 °C for determination of soil properties and at 105 °C for radionuclide measurement. Finally, soil samples were grained and sieved to pass through a 2 mm mesh-size stainless steel sieve.

Soil physicochemical properties – potential (pH_{H_2O}) and total (pH_{KCl}) soil acidity, specific electrical conductivity (EC_{25}), total organic carbon (*TOC*) content, carbonate content ($CaCO_3$), dry bulk (ρ_b) and particle density (ρ_p) – were analyzed following standard procedures [32]. Granulometric analysis was done by the traditional pipette method [33] and fractions of coarse

(0.20 mm to 2 mm) and fine sand (0.05 mm to 0.20 mm), silt (0.002 mm to 0.05 mm) and clay (<0.002 mm) were determined.

Radioactivity measurement

For radionuclide analysis, a total of 150 soil samples were placed into 500 cm^3 Marinelli beakers, and hermetically sealed for 40 days prior to analysis in order to attain a secular equilibrium between ^{226}Ra and its daughters within the ^{238}U decay series. The mass of each sample was approximately 0.5 kg. The specific activities of ^{40}K, ^{226}Ra, and ^{232}Th were determined using a coaxial HPGe detector (ORTEC-AMETEC, GEM 50195-P, 8192 channels) with an energy resolution of 1.95 keV at the 1.33 MeV of ^{60}Co and relative efficiency of 46 %. 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). Energy and efficiency calibrations were done using a MBSS2 standards source certified by the Czech Metrological Institute (silicone resin with homogeneously dispersed radionuclides ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{113}Sn, ^{85}Sr, ^{88}Y, and ^{203}Hg; total activity 38.34 kBq on August 16, 2010) in the same geometry as the measured samples. The counting time for each sample was 60000 s. Prior to the sample measurement, the environmental background at the laboratory site was determined with an empty Marinelli container under identical measurement conditions. Background spectral intensities were later subtracted from corresponding sample intensities. It is possible to conduct the measurement of ^{226}Ra directly through the 186 keV line, but its intensity is relatively low and the contribution from the ^{235}U line at 186 keV must be subtracted. A more precise and sensitive method of measurement of ^{226}Ra specific activity utilizes the gamma lines of ^{214}Pb and ^{214}Bi following a waiting period for achievement of a secular equilibrium between ^{226}Ra and ^{222}Rn. Consequently, the specific activity of ^{226}Ra was determined from the peak areas at 609.3, 1120.3, and 1764.5 keV of ^{214}Bi and 295.2 and 351.9 keV of ^{214}Pb. Moreover, the most intense post-radium lines originating from these two radionuclides (609.3 and 351.9 keV line) were selected, as there are no lines of any other radionuclide belonging to a natural family that might interfere. Since in each of the analyzed samples the specific activity of ^{226}Ra, as deduced from all the lines, gave consistent values within the uncertainties of their intensities, what justified the assumption of undisturbed equilibrium in the series, the final results of the analysis were based on the most intense post-radium lines, which yielded the highest statistical accuracy. Gamma ray peaks with energies of 911.2 and 969.0 keV (^{228}Ac) and 238.6 keV (^{212}Pb) were used for calculation of ^{232}Th. Specific activity of ^{40}K was determined using its own 1460.8 keV peak. The total uncertainty of the specific activity measurements, composed of the systematic errors (the uncertainty of the calibration source activity, the uncertainty of the activity, the uncertainty of the efficiency calibration and uncertainties in the nuclide master library used) and counting statistical errors was typically in the range 3-10 %. The minimum detectable activity for radionuclides of interest was calculated from the background measurement and found to be 0.4 Bq/kg for ^{226}Ra, 0.8 Bq/kg for ^{232}Th, and 2.0 Bq/kg for ^{40}K.

Statistical analysis

Shapiro Wilk's test of normality showed that only datasets of $A_s(^{232}\text{Th})$ and clay content were normally distributed. To improve normality of the data, all other variables that showed non-normal distribution were subjected to the Box-Cox transformation and transformed data were used for further statistical treatment. After transformation, all variables were tested for outliers according to Grubbs' criteria and no outliers were detected. All statistics were done using the IBM SPSS Statistics 20.0 software.

Pearson correlation coefficients were used to assess the relationships among soil properties and specific activity of radionuclides. An analysis of variance (ANOVA) was performed to determine if the distance and direction of sampling points from the CFPP affect the variation of $A_s(^{40}\text{K})$, $A_s(^{226}\text{Ra})$, and $A_s(^{232}\text{Th})$ in soil profiles. Principal component analysis (PCA) was used to obtain more reliable data about the relationship among specific activities of natural radionuclides, physicochemical soil characteristics, and some sampling properties.

To represent the spatial distribution and to create isolevel maps of the radionuclides investigated in the surface soil, as well as the absorbed gamma dose rate, the kriging gridding method with a linear variogram with no nugget effect was used. All of the maps and interpolation were performed using the Surfer 12 Golden Software.

Radiological risk assessment

Possible health effects due to the external exposure to natural gamma radiation were estimated based on the obtained results for specific activities of ^{40}K, ^{226}Ra, and ^{232}Th in the 0-10 cm soil layer, since it is considered that this layer predominantly contributes to the external gamma irradiation.

The absorbed gamma dose rate, $\dot{D}$ [nGyh^{-1}], in the air at a height of 1 m above the ground surface was calculated according to the UNSCEAR recommendation [6]

$$\dot{D} \quad 0.462A_s^a(^{226}\text{Ra}) \quad 0.604A_s^a(^{232}\text{Th}) \quad 0.0417A_s^a(^{40}\text{K}) \qquad (1)$$

where A_s^a [Bqkg^{-1}] is the specific activity of the corresponding radionuclide in the *a* sampling layer, and 0.462, 0.604, and 0.0417 are dose conversion factors for radionuclides in soil from the ^{238}U and ^{232}Th series and ^{40}K, respectively.

The annual outdoor effective dose, E [μSv], was calculated using the factor of 0.7 Sv/Gy to convert the absorbed dose rate in air $\dot{D}$ [nGyh^{-1}] to the effective dose rate for adults for environmental exposures to gamma rays and the outdoor occupancy factor (the fraction of time spent outdoors) of 0.2, both proposed by UNSCEAR [34]

$$E \quad \dot{D} \; t \;\; 0.7 \;\; 0.2 \;\; 10^{\;3} \tag{2}$$

where t represents the time of one year expressed in hours (8760 h).

External hazard index, H_{ex}, and radium equivalent activity, A_s^{eq} [Bqkg^{-1}],were calculated according to formulae [35]

$$H_{ex} \quad \frac{A_s^a(^{226}\mathrm{Ra})}{370} \quad \frac{A_s^a(^{232}\mathrm{Th})}{259} \quad \frac{A_s^a(^{40}\mathrm{K})}{4810} \tag{3}$$

$$A_s^{eq}(^{226}\mathrm{Ra}) \quad A_s^a(^{226}\mathrm{Ra})$$
$$1.43A_s^a(^{232}\mathrm{Th}) \quad 0.077A_s^a(^{40}\mathrm{K}) \tag{4}$$

These two indices have been introduced to represent the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K by a single quantity which takes into account the radiation hazards associated with them. The annual effective dose for radium equivalent activity of 370 Bq/kg corresponds to the dose limit of 1 mSv for the general population. H_{ex} has to be lower than unity for the radiation risk to be negligible.

RESULTS AND DISCUSSION

Specific activity and distribution of radionuclides in soil profiles

Descriptive statistics of specific activities of analyzed natural radionuclides are presented in tab. 1. Here we will mostly analyze mean values, while depth distribution will be considered later. The specific activities ranged from 12.7 1.5 to 52.4 5.6 Bq/kg with an average 31.3 Bq/kg for ^{226}Ra, from 13.5 2.3 to 55.0 5.6 Bq/kg with an average 32.8 Bq/kg for ^{232}Th and from 319 29 to 862 64 Bq/kg with an average 577 Bq/kg for ^{40}K. The values of measured specific activities of natural radionuclides in this study were not significantly different from those reported in two recent investigations carried out in the same study area [19, 24]. Specific activity of ^{226}Ra was higher than the specific activity of ^{232}Th in only 50 samples from 19 sampling sites where the mean values of pH_{H_2O}, $c(CaCO_3)$ and sand content (especially coarse sand) were remarkably higher than the mean values in other samples, indicating that higher alkaline conditions due to higher carbonate content and coarser soil fraction favor accumulation of ^{226}Ra in comparison with ^{232}Th. ^{40}K showed the lowest variation with the value of coefficient of variation (CV) of 18.1 %, while A_s (^{226}Ra) and A_s (^{232}Th) were almost equally variable with coefficients 24.8 and 26.0 %, respectively. The specific activities of ^{226}Ra and ^{232}Th in the studied soil samples were lower than their values in fly ash samples from the CFPP "Nikola Tesla" (120 and 72 Bq/kg) as reported by Janković *et al*. [20], while the mean A_s (^{40}K) found in this study was considerably higher than the specific activity of ^{40}K in fly ash (360 Bq/kg) as reported by the same authors. The mean values of A_s in

Table 1. Activity concentrations A_s [Bqkg^{-1}] of primordial terrestrial radionuclides in soils surrounding the CFPP "Nikola Tesla A"

Radionuclide	Sampling depth [cm]	Mean	Median	SD	CV [%]	Skewness	Kurtosis	Min	Max
A_s (^{40}K)	*a* (0-10)	598	590	98	16.4	0.38	0.47	372	833
	b (10-20)	579	578	103	17.7	0.07	0.60	325	804
	c (20-30)	568	547	112	19.7	0.65	1.42	319	861
	d (30-40)	566	560	102	18.0	0.79	2.23	331	862
	e (40-50)	573	549	109	19.1	0.40	0.19	335	814
	Mean (0-50)	577	563	104	18.1	0.42	0.63	319	862
A_s (^{226}Ra)	*a* (0-10)	32.0	30.1	7.8	24.3	0.45	–0.07	19.0	49.0
	b (10-20)	31.0	31.2	7.6	24.4	0.44	0.67	18.3	51.0
	c (20-30)	31.5	31.5	7.7	24.4	0.24	0.68	16.0	51.7
	d (30-40)	30.7	31.1	8.0	26.2	0.30	1.13	12.7	52.4
	e (40-50)	31.1	30.0	8.2	26.4	0.33	1.05	12.9	52.3
	Mean (0-50)	31.3	31.0	7.8	24.8	0.33	0.48	12.7	52.4
A_s (^{232}Th)	*a* (0-10)	33.7	33.1	8.6	25.6	0.02	0.55	14.3	55.0
	b (10-20)	32.6	32.3	8.2	25.2	–0.19	0.21	13.5	48.6
	c (20-30)	32.5	32.6	8.6	26.6	–0.44	–0.13	13.5	47.9
	d (30-40)	32.5	33.4	8.9	27.5	–0.39	–0.15	13.7	48.2
	e (40-50)	32.9	32.5	8.8	26.7	–0.22	–0.45	14.9	48.0
	Mean (0-50)	32.8	32.6	8.5	26.0	–0.24	–0.13	13.5	55.0

the soil surrounding the plant were quite close to the mean values of 32.8 Bq/kg, 37.8 Bq/kg, and 550 Bq/kg reported for the specific activities of ^{238}U, ^{232}Th, and ^{40}K, respectively, for the surface soil for the whole territory of Serbia [36]. Moreover, the A_s values around the plant were comparable to those obtained for regions in Serbia where no CFPP exist. Agricultural soils in western Serbia [37] showed noticeably higher specific activities for ^{232}Th and lower for ^{40}K, which is probably the consequence of the underlying parent material [36].

Comparative data found in the literature for soils around CFPP worldwide showed higher values of A_s (^{226}Ra) and A_s (^{232}Th) than ours for CFPP in China [14, 38, 39], India [40], Malaysia [7], and Brazil [13]. Similar specific activities of ^{226}Ra and ^{232}Th were measured for soils surrounding CFPP in European countries – Poland [16], Turkey [10, 11], Spain [4, 8], Greece [9, 17], with the exception of Hungary [15] where substantially higher concentrations of ^{226}Ra were determined.

Soil properties

Table 2 summarizes the mean values for physicochemical properties of soils by sampling layers and for whole soil profiles.According to the soil pH most samples (85 %) were neutral to moderately alkaline with a net negative charge ($pH_{KCl} < pH_{H_2O}$). Based on EC_{25} values, all soil samples were characterized as non-saline. The average *TOC* content of layers showed a steadily decreasing trend with the soil depth and it varied from 2.12 % in the *a* layer to 1.13 % in thee layer. The carbonate content varied notably among the sampling sites. Densities of the soil samples were quite similar and tend to increase with depth.

Granulometric analysis revealed that the most dominant soil fraction was silt, with a mean content of 55.1 %. The average fraction of clay was 24.7 %. The percentage abundance of sand exhibited the greatest variability with values varying from 2.3 % to 95.1 %. Fine sand content was higher than the content of coarse sand in superficial layers (*a*, *b*), while the coarse sand was more abundant in the *c*, *d*, and *e* layer.

Correlation between soil properties and specific activities of radionuclides

The correlations between specific activities of radionuclides and soil properties as well as mutual correlations between radionuclides are presented in tab. 3. Correlation analysis confirmed that the natural radioactivity increases with the particle size decrease and increase of the surface-to-volume ratio, which has been well documented in other investigations [1, 41]. The relation between A_s (^{40}K) and the clay content suggests that potassium activity tends to increase with the increasing clay fraction in soil [17]. Taking into account positive and significant relationships of all investigated radionuclides with the silt and negative ones with the sand, it is obvious that radionuclides were most abundant in finer soil fractions [19, 24, 41, 42]. That is additionally supported by the significant negative interrelations between all radionuclides with bulk density and significant negative correlation of ^{226}Ra and ^{232}Th with the particle density.

The lack of significant correlation between *TOC* and specific activities of ^{40}K, ^{226}Ra, and ^{232}Th indicates their association with the mineral soil fraction [1, 24, 43].

The abundance of ^{226}Ra and ^{232}Th was inversely and significantly related to the soil carbonate content, pH_{H_2O}, pH_{KCl}, and EC_{25} which is in agreement with data available in literature [1, 19, 43]. It appears that ^{226}Ra and ^{232}Th tend to accumulate in soil layers where carbonates have been leached [42]. Specific activities of radionuclides were not significantly correlated with *TOC*, but organic matter could influence their distribution indirectly through carbonate leaching. Decomposition of organic matter in the upper soil horizons re-

Table 2. Mean values of physicochemical properties of soil at different depths

Soil properties	Soil depth [cm]							
	a (0-10)	*b* (10-20)	*c* (20-30)	*d* (30-40)	*e* (40-50)	Mean (0-50)	Min (0-50)	Max (0-50)
pH_{H_2O}	7.23	7.28	7.29	7.32	7.31	7.29	5.50	8.75
pH_{KCl}	6.15	6.15	6.13	6.15	6.16	6.15	4.09	7.27
EC_{25} [Scm^{-1}]	220.9	186.1	166.7	163.3	184.3	184.3	32.8	489.0
TOC [%]	2.1	1.7	1.4	1.2	1.1	1.5	0.0	6.0
$c(CaCO_3)$ [%]	4.8	4.5	5.2	5.2	5.0	4.9	0.3	18.2
ρ_b [gcm^{-3}]	1.26	1.32	1.34	1.36	1.37	1.33	1.06	1.67
ρ_p [gcm^{-3}]	2.27	2.36	2.40	2.45	2.46	2.39	0.83	2.79
Sand coarse [%]	8.8	9.8	11.4	10.6	10.7	10.3	0.6	83.7
Sand fine [%]	10.6	9.9	9.7	9.5	9.9	9.9	1.1	42.8
Sand total [%]	19.4	19.7	21.1	20.1	20.6	20.2	2.3	95.1
Slit [%]	57.0	55.6	54.5	54.9	53.5	55.1	3.6	81.1
Clay [%]	23.6	24.7	24.5	24.9	25.9	24.7	1.3	43.8

Table 3. Pearson correlation coefficients between the specific activities of natural radionuclides and physicochemical properties in soils surrounding the CFPP "Nikola Tesla A"

Parameter	A_s (^{40}K)	A_s (^{226}Ra)	A_s (^{232}Th)
Depth	–0.088	–0.044	–0.030
pH_{H_2O}	0.044	–0.408**	–0.502**
pH_{KCl}	0.008	–0.410**	–0.518**
EC_{25}	0.103	–0.298**	–0.384**
TOC	0.040	0.152	0.130
$c(CaCO_3)$	–0.040	–0.264**	–0.339**
ρ_b[gcm^{-3}]	–0.209*	–0.388**	–0.306**
ρ_p[gcm^{-3}]	–0.146	–0.312**	–0.292**
Sand fine	–0.025	–0.540**	–0.656**
Sand coarse	–0.246**	–0.405**	–0.540**
Sand (total)	–0.172*	–0.560**	–0.696**
Silt	0.172*	0.540**	0.629**
Clay	0.257**	0.004	0.119
A_s (^{40}K)	1	0.035	0.104
A_s (^{226}Ra)	0.035	1	0.890**
A_s (^{232}Th)	0.104	0.890**	1

* Correlation is significant at the 0.05 level (2-tailed), ** Correlation is significant at the 0.01 level (2-tailed)

leases CO_2 which promotes lower pH and creates a suitable environment for carbonate dissolution. Carbonate leaching further induces an increase of soil alkalinity and specific conductivity in deeper layers due to soil solution transport. This hypothesis is supported by positive significant correlation between *TOC* and $c(CaCO_3)$ and *TOC* and EC_{25}.

The specific activities of ^{226}Ra and ^{232}Th were positively and strongly correlated, which is an indication that they have mineral components of the soil as a common source as pointed out by Navas *et al*. [1]. The lack of correlation between these two radionuclides and ^{40}K may reflect their origin from a different parent rock composition and their different chemical behavior in the soil [41].

Vertical distribution of radionuclides

The specific activities of natural radionuclides exhibited very little variation with the soil depth. As it is indicated in tab. 1 and shown in tab. 3, in case of the radionuclides investigated ^{40}K, ^{226}Ra, and ^{232}Th, the correlation analysis did not identify any significant distribution trend correlated with the depth. This is in consonance with the findings of Dragović *et al*. [43] who determined specific activities of radionuclides in six profiles of different soil types corresponding to those analyzed in this work and revealed their homogenous distribution with depth.

Despite no significant correlation between soil depth and specific activities of radionuclides, when analyzing the mean A_s (^{40}K) in different soil layers some trends can be noticed. The highest mean specific activity of ^{40}K was determined for the surface soil layer *a* (tab. 1), and a generally decreasing trend with depth was observed until the 40-50 cm layer where A_s (^{40}K) was greater than in the *c* and *d* layers. Potassium can be weathered from silicate minerals and transferred to the soil solution, adsorbed in clays, and as a result of colloidal transport be accumulated in deeper layers [23]. Additionally, depletion of ^{40}K in subsoil layers can be explained due to nutrient uptake by the root system of plants [44].

Specific activity of ^{226}Ra remains fairly constant with depth. The average vertical mobility of radium in undisturbed soil profiles is about 0.005 cm per year [45]. Numerous studies of radium mobility in soil applying sequential extraction analysis showed that more than 50 % of Ra is associated with the residual soil fraction, 15-30 % with Fe and Mn oxides, 15 % of Ra is organically bound, 5 % is associated with carbonates and approximately 5 % was in easily exchangeable form [45]. Since correlation analysis showed a significant negative relation between A_s (^{226}Ra) and carbonate content and an absence of significant correlation with organic matter, uniformity of ^{226}Ra distribution can be explained by the strong adsorption of divalent Ra cation by Fe and Mn oxides and formation of stable radium complexes [1].

Thorium specific activity was quite uniform down the profiles. Homogenous distribution of ^{232}Th is in agreement with the fact that thorium has low geochemical mobility, an insoluble nature in water, high sorption tendency and it is firmly bound to the soil fractions and increased mobility can be expected only in the extreme acidic environments [46].

Spatial distribution of radionuclides

ANOVA was carried out to determine if there were significant differences between average specific

activities of natural radionuclides in different subgroups, based on the distance and direction from the CFPP. The average values of A_s (^{40}K), A_s (^{226}Ra), and A_s(^{232}Th) for each soil profile 0-50 cm were input variables for ANOVA.

To assess the effects of distance from CFPP on specific activities of ^{40}K, ^{226}Ra, and ^{232}Th, profiles were divided into six groups – profiles located at the distance: up to 1.5 km (group A), 1.5 km to 3 km (B), 3 km to 5 km (C), 5 km to 7 km (D), 7 km to 9 km (E), and 9 km to 11 km (F). Mean values of A_s(^{40}K), A_s(^{226}Ra), and A_s(^{232}Th) in the whole profile together with their mean values at different sampling depth depending on distance are presented in fig. 1(a). For all radionuclides investigated, notably higher activity concentrations were found in distance group A than in soil samples classified in group B where the lowest specific activities were measured. This enrichment of radionuclides in soil profiles positioned closest to the CFPP is attributed to CFPP operations due to the high fly ash concentration within the CFPP and its closest proximity, as well as flying coal dust originating from coal stockpiles. This is in good agreement with the findings that high radionuclide concentrations in soil are generally confined within the 1 km distance from the power plant [40]. Similar results have been reported in studies of environmental impact of the CFPP in Turkey [11], China [12], and Brazil [13] where significantly higher specific activities were found in samples collected at distances within 1 km from the CFPP than 3-4 km away. The highest specific activities of ^{40}K were determined in soil sampled from approximately 4 km and 6 km away from the CFPP (C and D group). Since this part of the study area is in agricultural use, the higher concentration of ^{40}K is probably due to potassium fertilizer application [47]. The distributions of ^{226}Ra and ^{232}Th showed very similar patterns of spatial distribution, which implies a common geological media from which they were derived and an analogous response to soil and environmental processes. The specific activities of ^{226}Ra and ^{232}Th gradually increased up to 10 km, and the highest values in all sampling layers were determined in the most distant groups (E and F). Results of ANOVA presented in tab. 4 showed a significant difference among groups for means of ^{226}Ra and ^{232}Th specific activities, with a large effect size quantified by eta squared value (η^2) 0.43 and 0.48 respectively. From these results one can conclude that gaseous combustion products and particles containing both radionuclides are carried, dispersed and then accumulated far from the CFPP. Similar behavior of A_s (^{226}Ra) and A_s (^{232}Th) with

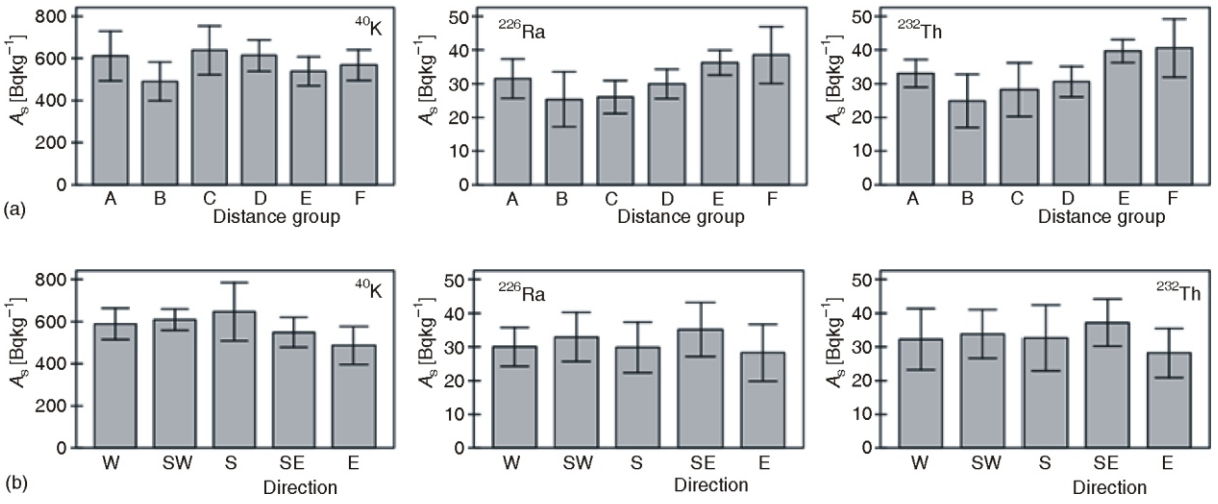

Figure 1. Average values of specific activities of radionuclides in soil profiles in (a) different distance groups, (b) different directions (subregions)

Table 4. The analysis of variance for specific activities of natural radionuclides in soil samples collected at different directions and distance from the CFPP

Source of variation	Homogeneity of variances		ANOVA		
A_s	Levene statistic	Significance	F-statistic	Significance	Effects size (η^2)
Distance					
^{40}K	0.256	0.933	1.700	0.173	0.26
^{226}Ra	1.536	0.216	3.665	0.013	0.43
^{232}Th	1.579	0.204	4.513	0.005	0.48
Direction					
^{40}K	2.476	0.070	2.746	0.051	0.31
^{226}Ra	0.189	0.942	0.789	0.543	0.11
^{232}Th	0.114	0.976	0.881	0.490	0.12

distance was found in the study of Charro *et al*. [8], but according to the same authors [4] the evidence of deposition processes of radionuclides from airborne fly ash discharges can be perceived through the *K*-parameter, *i. e.* ratio between A_s of a particular radionuclide in the top and deeper soil horizons. The calculated mean *K*-ratio (A_s in *a*-layer divided by A_s in *b*-layer) for the whole study area was 1.04 for all the radionuclides investigated. In addition, the depth distribution of ^{226}Ra and ^{232}Th was fairly uniform in the E and F distant group, with *K*-parameter of 1.04 (E) and 0.98 (F) for ^{226}Ra and 1.04 (E) and 1.03 (F) for ^{232}Th. It is worth mentioning that some of the highest A_s (^{226}Ra) and A_s (^{232}Th) values were measured for sampling sites W08, SW08 (E group) and W10 and SW10 (F group) that are located very closely to the ash repository of the CFPP "Nikola Tesla B", as well as for samples taken from S08 and E08 belonging to the F group and SE10 sampling point belonging to the E group that are positioned nearest to the Kolubara basin and can experience the direct effect from the coal open pit mines. This is in good agreement with the research of Nenadović *et al*. [22, 23] who studied the vertical distribution of natural radionuclides in soil profiles taken at Rudovci and located very close to the Kolubara open pit coal mine, and found markedly higher specific activities of ^{226}Ra and ^{232}Th than values measured in this study, ranging from 57-84 Bq/kg and 59-71 Bq/kg, respectively. Furthermore, radioactivity measurement at the CFPP "Nikola Tesla B" ash disposal site conducted in the period 1990-2011 showed substantially increased specific activities of ^{226}Ra (111 Bq/kg) and ^{232}Th (75 Bq/kg) [48].

In the case when the direction from the CFPP was investigated as a source of variance, the soil profiles were divided into five subregions based on cardinal (W, S, E) and ordinal (SW, SE) directions. The mean values of specific activities of ^{40}K, ^{226}Ra, and ^{232}Th in these subregions are shown in fig. 1(b). The highest profile mean values for A_s (^{226}Ra) and A_s (^{232}Th) were observed in the SE subregions, which corresponds to the downwind area of NW winds. Those winds have the strongest influence on the spreading of airborne polluting agents originating from the CFPP and ash deposits, especially during the summer, when W and NW are the prevalent winds. Additionally, a consequence of winds from the SE quadrant (annual predominant downwind directions) is the pollution from the nearby lignite reserve. The highest specific activity for ^{40}K was recorded in profiles located on in the south from the CFPP. For all radionuclides the lowest specific activities were measured in profiles from the E subregion.

The combination of previous considerations suggests that the CFPP is an unlikely source of the radionuclides found in the surrounding area, except for the soil located in the 1.5 km zone around the CFPP. It appears that the soil properties can act as the main source of radionuclide variability and that only elevated specific activities of radionuclides found in the southwestern part of the study area could be a consequence of the fly ash from the repository site of the CFPP "Nikola Tesla B". Additionally, the pollution coming from open pit mines should be also considered as a source of elevated specific activities of radionuclides in the southeastern part of the study area.

Principal component analysis

Table 5 presents the results of the principal components' (PC) loadings with a Varimax rotation, as well as eigenvalues and communalities. Five PC were obtained through PCA with eigenvalues greater than 1, summing up 78.2 % of total variance.

The PC1, explaining the largest proportion of 26.8 % of the total variance is a bipolar factor. The positive pole is heavily loaded by specific activities of ^{226}Ra and ^{232}Th and silt content and moderately by distance from the CFPP, while the negative pole represents the variability of the sand amount and particle density. This PC makes clear the close relationship between ^{226}Ra and ^{232}Th and the finer soil granulometric fraction. The PC2 illustrates the joint effect of pH, soil salinity (EC_{25}), and carbonate content and may be designated as a "carbonate leaching" factor. High positive loadings of *TOC*, and low negative loadings of sampling depth are dominant in PC3, which accounts for 11.1 % of the total variance, and can be considered as an "organic material" factor. The PC4 was related to high positive values of loading associated with clay content and A_s (^{40}K), and low negative values for the sand fraction. This PC elucidates 10.8 % of the total variance and can be termed as a "mineral" factor. About 90-95 % of the potassium in soil is fixed potassium in the lattice of silicate minerals such as feldspar and micas. Further weathering and leaching releases potassium ions which are transferred to the soil solution and are adsorbed on the cation exchange sites of clays. There exists an equilibrium among fixed, exchangeable and potassium in the solution in which ^{40}K is contained only approximately 0.01 % [44].The "depth" factor is determined by the PC5 which is characterized by high positive loadings of sampling depth and soil particle density, and explains 10.4 % of the overall data variability.

Where as PC represent a set of linearly uncorrelated variables, allocation of A_s (^{40}K), A_s (^{226}Ra), and A_s (^{232}Th) together with soil grain size fractions apart from other soil physicochemical properties that loaded overwhelmingly other PC is a clear indication that the soil texture is the most important controlling factor of radionuclides in the soil investigated. In general, the fine-grained soil fraction has a higher tendency for radionuclide adsorption than the coarse-grained soil because it contains soil particles with large surface areas such as clay minerals, Fe and Mn oxides and hydroxides,

Table 5. Loadings of the selected variables (soil properties, sampling parameters and specific activities of natural radionuclides) for significant principal components for the soil samples

Variable	Communalities	Principal component[a]				
		1	2	3	4	5
Depth	0.831	0.083	0.086	–0.396	–0.007	0.813
Distance	0.522	0.585	–0.269	–0.306	–0.081	–0.089
pH_{KCl}	0.765	–0.322	0.808	–0.061	0.020	0.066
EC_{25}	0.825	–0.218	0.804	0.323	0.111	–0.118
TOC	0.909	0.049	0.192	0.929	0.017	–0.080
$c(CaCO_3)$	0.783	–0.143	0.841	0.087	–0.218	0.002
ρ_p	0.730	–0.366	–0.112	0.267	–0.054	0.714
Sand	0.837	–0.786	0.234	0.061	–0.393	–0.073
Silt	0.765	0.836	–0.002	–0.181	0.073	–0.167
Clay	0.792	–0.032	–0.381	0.132	0.750	0.255
$A_s(^{40}K)$	0.687	0.098	0.154	–0.053	0.776	–0.221
$A_s(^{226}Ra)$	0.819	0.847	–0.204	0.216	–0.102	–0.058
$A_s(^{322}Th)$	0.900	0.882	–0.291	0.187	0.025	–0.031
Variance explained [%]						
Total		26.767	19.131	11.065	10.816	10.420
Cumulative		26.767	45.898	56.963	67.779	78.199

[a] Rotation method: Varimax with Kaiser normalization

and humic acids. Radionuclides can be adsorbed onto clay surfaces that are mostly negatively charged, or become fixed within the lattice structure. Sand particles have a small specific surface area, and such particles are often chemically inert.

Radiological hazard

The calculated values of radiological risk parameters are presented in tab. 6. This study shows the range of the absorbed gamma dose rate from terrestrial natural radionuclides from 36.7 to 77.7 nGy/h with a mean value of 60.3 nGy/h, which is slightly higher than the world mean value (57 nGy/h) [34]. The mean contribution of ^{40}K, ^{226}Ra, and ^{232}Th to the dose rate is 42.2 %, 24.3 %, and 33.5 %, respectively. The average outdoor annual effective dose calculated for the study area (73.9 Sv) is not significantly greater than the worldwide average value of 70 Sv as given by UNSCEAR [6] and is well below the annual dose limit of 1 mSv recommended by the ICRP for the general public [49].

Figure 2 depicts kriging type estimated maps for $\dot{D}$ values over the study area, along with the maps for spatial distribution in top soil (*a*-layer) for radionuclides of interest. Although the greatest contribution to $\dot{D}$ comes from ^{40}K, it can be seen that spatial variability of $\dot{D}$ is mainly controlled by the content of ^{226}Ra and ^{232}Th (see eq. 1 for the contribution of particular isotopes to the total dose), hence the isodose lines follow to a great extent the distribution pattern of these two radionuclides.

The mean of A_s^{eq} (^{226}Ra) and H_{ex} was found to be 126.3 Bq/kg and 0.34, respectively. Moreover, in all soil samples, the values of A_s^{eq} (^{226}Ra) were below 370 Bq/kg and H_{ex} was less than the acceptable limit of unity, which demonstrates that concentrations of natural radionuclides in the area do not pose any significant health risk to the general public.

As it is presented in tab. 7, all calculated radiological risk parameters were within the ranges reported for the whole territory of Serbia and regions in Serbia where CFPP do not operate. In comparison with areas affected by CFPP operations in Serbia, slightly lower radiological risk was assessed in this study, which is in good agreement with Vukašinović *et al*. [19] who measured lower values of specific activities of natural radionuclides in soil around the CFPP "Nikola Tesla A" in comparison with other Serbian power plants. Furthermore, our results match well the radiological indices reported for soils surrounding CFPP in other European countries, except for Hungary where the mean artificial increment of 21.8 μSv from

Table 6. Estimated gamma dose rate $\dot{D}$ [nGyh^{-1}], annual effective dose E [μSv], external hazard index H_{ex} and radium equivalent activity A_s^{eq} (^{226}Ra)[Bqkg^{-1}] for the study area based on the results for the A_s (^{40}K), A_s (^{226}Ra), and A_s (^{232}Th) in 0-10 cm soil layer

Parameter	Mean	Median	SD	CV [%]	Skewness	Kurtosis	Min	Max
$\dot{D}$	60.3	60.4	9.0	15.0	–0.24	0.38	36.7	77.7
E	73.9	74.1	11.1	15.0	–0.24	0.38	45.0	95.3
H_{ex}	0.34	0.34	0.05	15.6	–0.16	0.17	0.21	0.45
A_s^{eq} (^{226}Ra)	126.3	125.4	20.1	15.9	–0.13	0.15	76.8	167.7

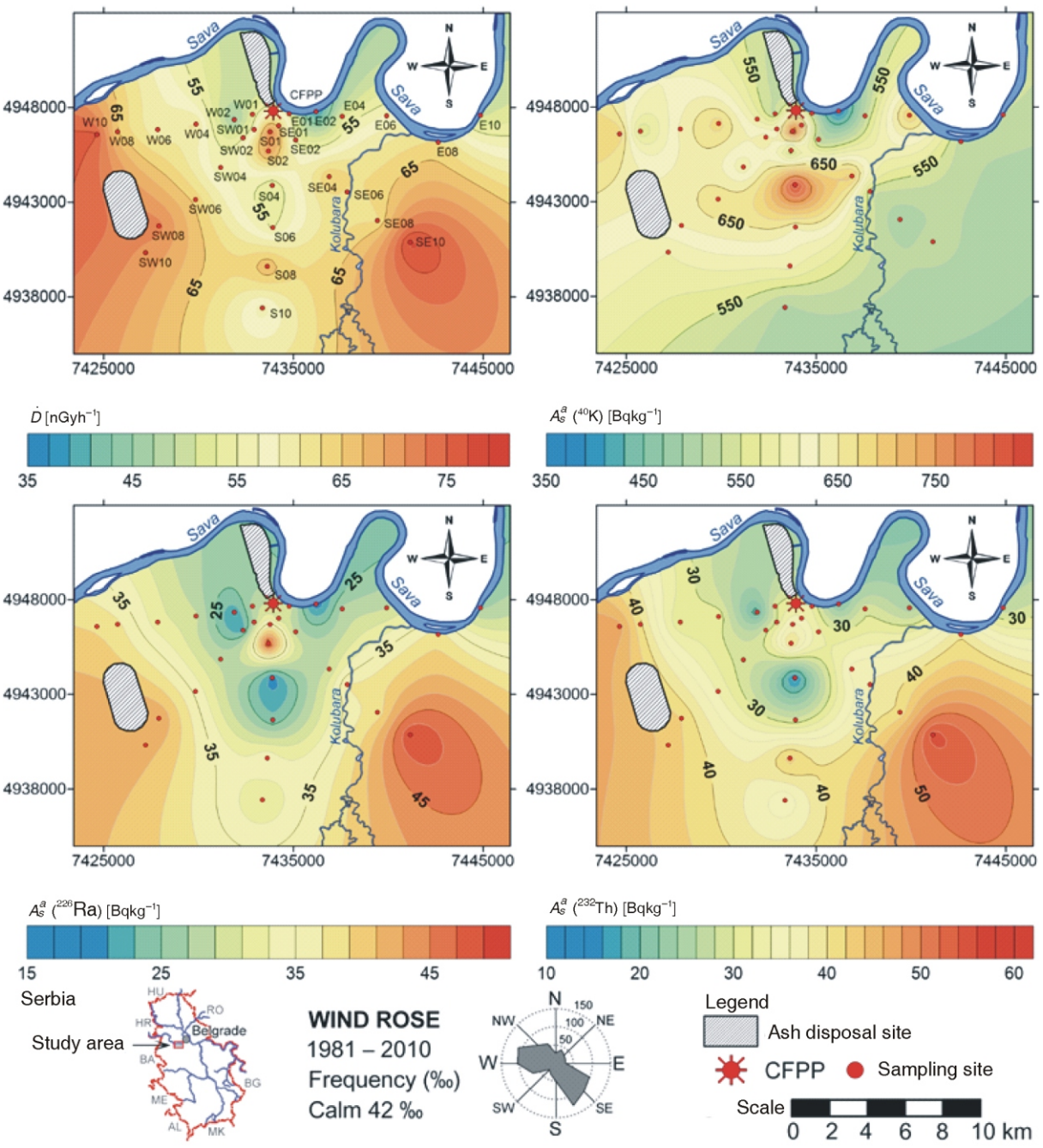

Figure 2. Spatial distribution of the calculated absorbed gamma dose rate due to natural radionuclides and measured specific activities of ^{40}K, ^{226}Ra, and ^{232}Th in the 0-10 cm soil layer in the study area (location of the study area and wind frequency wind rose for the period 1981-2010 is given at the figure bottom)

the CFPP "Ajka" in the annual effective dose from external exposure to terrestrial radionuclides outdoors has been found [50]. Literature data available for non-European countries like China, India and Malaysia showed a remarkably higher radiological hazard for the terrestrial environment around CFPP. It has been attributed to the high radionuclide background level or to the high radionuclide content of feed coal.

Therefore, based on the facts that values of all calculated radiological risk parameters were within the permissible limits, it can be concluded that there is no health hazard from the soil surrounding the largest Serbian CFPP when natural gamma radioactivity is concerned.

CONCLUSIONS

The specific activities of ^{40}K, ^{226}Ra, and ^{232}Th in 30 soil profiles have been measured and used to evaluate distribution and possible impact of major Serbian CFPP on the surrounding area. Radionuclides were fairly

Table 7. Comparative data of radiological risk parameters for different parts of Serbia and soils around CFPP in Serbia and worldwide

Region, country (CFPP)	$\dot{D}$ [nGyh^{-1}]	E [μSv]	H_{ex}	A_S^{eq}(^{226}Ra) [Bqkg^{-1}]
This study	60.3 (36.7-77.7)	73.9 (45.0-95.3)	0.34 (0.21-0.45)	126.3 (76.8-167.7)
Serbia (without CFPP influence)				
Serbia, whole territory [51]	73.7 (32.3-87.1)	90.4 (39.6-106.8)	0.43 (0.19-0.51)	–
Belgrade [52]	60.5 (24.7-89.6)	72.3 (11.0-107)	0.35 (0.14-0.52)	129 (50.8-193)
Kragujevac [53]	63.7 (43.7-83.2)	78.1 (53.6-102.1)	0.4 (0.3-0.5)	138.1 (93.4-180.0)
Vojvodina [54]	73.7 (32.3-87.1)	90.4 (39.6-106.8)	0.43 (0.19-0.51)	158.7 (69.5-188.1)
Western Serbia [37]	73.4	90	–	–
Serbia (with CFPP influence)				
CFPP "Nikola Tesla" (A and B) [24]	76.3 (41.9-93.1)	93.6 (51.4-114.1)	0.4 (0.24-0.54)	–
CFPP "Nikola Tesla" (A, B, Kolubara, Morava) [19]	69.4 (35.0-97.9)	85 (43-120)	–	–
Other countries (with CFPP influence)				
China (Baoji II) [38]	86.6 (78.6-94.0)	106 (96-115)	0.49 (0.44-0.54)	182.9 (164.7-199.1)
China (Baqiao) [39]	78.9 (70.6-95.6)	97 (87-117)	–	165.7 (148.4-199.1)
China (Mawan) [14]	307 (163-406)	370 (200-500)	–	681 (346-878)
Greece (Amintaio, Ptolemas, Kardia, Agios) [9]	62 (44-75)	76 (54-92)	0.32 (0.24-0.37)	117 (88-138)
Greece (Megalopolis A and B) [17]	57 (2-101)	80 (10-150)	0.31 (0.22-0.58)	115 (83-214)
India (Kolaghat) [40]	178.4 (96.2-328.5)	220 (120-400)	1 (0.5-1.8)	375.2 (197.4-685.3)
Malaysia (Sultan SA Aziz) [7]	73.3 (31.5-124.8)	–	–	159.3 (66.7-270.6)
Spain (Velilla del Río Carrion) [8]	58 (18-85)	71 (22-108)	–	–
Turkey (Chairhan) [11]	58.90 (18.70-108.75)	17.99 (22.62-133.37)	0.34 (0.11-0.65)	123.90 (139.04-238.92)
Turkey (Yatagan) [10]	56 (30-100)	–	–	–
Turkey (Yenikoy, Kemerkoy) [10]	54 (15-126)	–	–	–

evenly distributed along the sampled soil profiles and the spatial distribution of natural radionuclides in the soil did not indicate any significantly increased specific activities in the area downwind from the plant. Despite findings of statistically significant dependence of the ^{226}Ra and ^{232}Th specific activities on the distance from the CFPP, we concluded that it was not a consequence of the CFPP operation. Correlation analysis and PCA confirmed that among the determined soil physicochemical properties, the soil texture was the most influential factor on spatial and vertical distribution of natural radionuclides. The study's results showed that the CFPP "Nikola Tesla A" did not increase the natural radioactivity level in the entire study area, but just in its closest vicinity at distances less than 1.5 km. All calculated radiation hazard indices were lower than the recommended values. Therefore it can be inferred that the soil around the CFPP does not pose any significant radiological threat to the population.

This study contributes to the better understanding of distribution of natural radionuclides and their behavior in soil profiles from the area influenced by operation of the CFPP. Since coal remains the main fossil fuel for power generation in Serbia, the monitoring of content of natural radionuclides in soils around the CFPP has to be continued in regular intervals, and in this particular case this study can serve as a reference point for future investigations.

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AUTHORS' CONTRIBUTION

Soil sampling, sample preparation, data processing, and graph and map plotting were carried out by M. N. Tanić under guidelines of S. D. Dragović, M. Z. Daković and G. G. Bačić. Measurement and experimental set up was carried out by Lj. J. Janković Mandić, B. A. Gajić and M. N. Tanić. All authors performed literature research, theoretical analysis, and discussion of the presented results. The manuscript was written by all the authors.

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ПРИРОДНИ РАДИОНУКЛИДИ У ПРОФИЛИМА ЗЕМЉИШТА У ОКОЛИНИ НАЈВЕЋЕ ТЕРМОЕЛЕКТРАНЕ У СРБИЈИ

Студија се бави проценом утицаја највеће термоелектране у Србији на концентрацију радионуклида у профилима земљишта дубине 50 cm. Тридесет земљишних профила је узорковано из околине термоелектране "Никола Тесла А" на растојању до 10 km од термоелектране и стандардним методама анализиране су физичко-хемијске карактеристике земљишта, а спектрометријом гама-зрачења измерене специфичне активности природних радионуклида (^{40}K, ^{226}Ra, и ^{232}Th). Анализирана је просторна и вертикална дистрибуција радионуклида у циљу одређивања односа између њихових специфичних активности у земљишту, карактеристика земљишта и одређивања фактора са највећим утицајем. Израчунати су радиолошки параметри и спроведена је процена радиолошког ризика. Измерене специфичне активности су биле сличне природним вредностима у Србији. Дистрибуција радионуклида се није значајно мењала са дубином. Гранулометријски састав је био најутицајнији фактор земљишта на дистрибуцију радионуклида. Сви параметри радиолошког ризика су били мањи од усвојених граница и препоручених вредности. Главни закључак истраживања је да термоелектрана значајно не утиче на просторну и вертикалну дистрибуцију природних радионуклида у земљишту испитиваног простора, али се технолошки повећани нивои природне радиоактивности могу уочити као последица рада термоелектране у пречнику од 1,5 km око термоелектране.

Кључне речи: природна радиоактивност, гама спектроскопија, карактеристика земљишта, анализа главних компонената, радијациони ризик, процена дозе