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NATURAL RADIONUCLIDES IN SOILS IN SERBIA: DOSE CALCULATIONS AND ENVIRONMENTAL RISK ASSESSMENT

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Abstract. *The paper presents the results of environmental risk assessment from natural radionuclides in soils in Serbia during 1983–2009. Soils were sampled in urban and mountainous areas, around several coal power plants, at an experimental farm, and in the region exposed to depleted uranium ammunition in 1999. External dose rates, radium equivalent activities, external hazard indices, and annual effective doses are calculated and the environmental risk from natural radionuclides in soils is estimated.*

Key words: *natural radionuclides, soils, dose, risk assessment*

1. INTRODUCTION

Since the level of outdoor exposure mainly depends on radionuclides in the first (20–30) cm of soil, natural radionuclides in soil (e.g., ^{40}K and the radionuclides of the ^{232}Th and ^{238}U series) significantly contribute to the total dose of irradiation from natural sources of radiation to population and environment (Fisenne, 1993; Kathreen, 1994). Content of natural radionuclides in soils is primarily determined by their geological origin. Higher contents are found in igneous rocks (granite) and some shale and phosphate rocks, and lower contents in sedimentary rocks.

Complexes of magmatic, sedimentary and metamorphic rocks, different in age and petrogeochemical characteristics, can be found in Serbia. The northern part of the country (Pannonian Basin) mainly consists of alluvial deposits, while the eastern (Carpatho-Balkan) and western (Dinaric) parts are composed of karstified limestone (ODD, 1992).

Various soil treatments (agricultural versus undisturbed soil) and other anthropogenic activities (urban soils exposed to different sources of pollution), as well as local climate and position, also contribute to differences in the content of natural radionuclides and thus to the level of annual effective dose and external hazard index (ODD, 1992; Popovic et al., 2008).

Radionuclide ^{40}K (half-life $1.248 \cdot 10^9$ years) is an isotope of potassium (0.012 %) that is found in nature as an ionic salt in sea water and minerals. It makes up to 1.5 % of the Earth's crust and is crucial for functioning of all living cells. As a fertiliser, potassium is used in the form of potassium chloride, sulphate or nitrate, thus enhancing the level of natural radioactivity (Kathreen, 1994).

Natural uranium is a mixture of three radioisotopes: ^{238}U (99 %, half-life $4.5 \cdot 10^9$ years), ^{235}U (0.71 %), and ^{234}U (0.006 %). Its most important daughter is ^{226}Ra (half-life 1602 years), found in majority of uranium ores, which decays to another important radioactive isotope – radon. Average global concentrations of uranium in soils are (50–125) Bq/kg, and up to 5000 Bq/kg in some shale rocks (Eisenbud, 1989). In some areas of central-east and south-east Serbia, the activities of uranium can reach 200 Bq/kg, with the mean (50–100) Bq/kg (Todorovic et al., 2001). Different mining, industrial and agrotechnical activities present pathways through which uranium enters the environment (Djuric and Popovic, 2000).

Thorium-232 (half-life $1.405 \cdot 10^{10}$ years) is less radioactive than uranium, but more abundant in nature. Its world average concentrations in soils are around 12 ppm, thus presenting a significant cancer risk in the area of enhanced natural radioactivity (Eisenbud, 1989; Kathreen, 1994).

Since the 1960's, radioactivity monitoring programme in Serbia has included studies on the contents of natural and anthropogenic radionuclides in soils, plants, air, food and feed, in urban as well as in rural areas. A study of Radovanovic and Vukotic (1985) showed 15 different radioecological areas in Serbia, while Bikit et al. (1995) found soils with an enhanced natural radioactivity near the mine of Kalna.

In 1999, four sites, Borovac, Bratoselce, Reljan and Plačkovica, near the town of Vranje were contaminated by depleted uranium (DU) with the total contaminated area of about 16000 m^2 (RA Report, 2002; Sarata et al., 2004). The targeted sites were subsequently isolated and decontaminated, and missile fragments stored as radioactive waste (Popovic et al., 2002). The United Nation Mission in Serbia and Montenegro in 2002 reported levels of uranium in soils in the range (1.0–9.5) mg/kg, with traces of depleted uranium found in less than 15 % of the samples (UNEP, 2002).

2. Materials and Methods

Natural radionuclides were determined in soils of the mountains Tara, Šara and Stara Planina (1983–2000) (Popovic et al., 1996; Todorovic et al., 1996, 2001), Rudnik, Goč, Crni vrh and Besna Kobila (2002–2008).

Soils in the Vranje area were sampled in 2001, at the locations exposed to DU ammunition. The results did not include samples from the craters of the targeted sites, with high concentrations of depleted uranium (Miljevic et al., 2001).

Samples of soils around coal power plants: Obrenovac, Lazarevac, Kostolac, and Svilajnac, were collected over the 2003–2009 period (RC, 2009).

Over 2002–2008, soils were sampled in towns of Belgrade, Paraćin, Ćuprija, Jagodina, Šabac, Aranđelovac, and at the Radmilovac farm, which is in the vicinity of Belgrade (Vukasinovic et al., 2009).

2.1. Sampling and Instrumentation

Soils, taken from a depth of 5 cm, were dried up to 105 °C, grounded, sieved, and left in Marinelli beakers for four weeks to reach the radioactive equilibrium. Radionuclides were determined on two High Purity Germanium detectors (Canberra, relative efficiencies 20 % and 23 %) by standard gamma spectrometry.

Geometric calibration was performed using: reference source CBSS 2 (soil, Czech Metrological Institute, Inspectorate For Ionizing Radiation, Praha); reference soil material (National Office of Measures OMH, Budapest, MIX-OMH SZ) spiked with a series of radionuclides ^{22}Na, $^{57/60}$Co, ^{89}Y, ^{133}Ba and ^{137} Cs (total activity 1.5 kBq/kg on 1.7.1991, uncertainty 5 %); IAEA-Soil 6 standard spiked with ^{137}Cs (58 Bq/kg) and ^{226} Ra (93 Bq/kg) on 30.1.1993, significance level 0.005; and a secondary reference material made from the reference soil material Czech Metrological Institute, Praha 9031-OL-116/8, type ERX (spiked with ^{241}Am, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{88}Y, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{210}Pb; total activity 114.9 kBq on 03.03.2008) in plastic 100 g boxes.

Counting times were (60.000–300.000) s.

Shauvenet criterion was applied for statistical analysis of the data. The spectra were analysed by GENIE 2000 program. The total standard error of the method was estimated to 20 %.

2.2. Calculations

Radium equivalent activity Ra_{eq} and external hazard index H_{ex} are reliable parameters for estimating radiation hazard from radionuclides in soils. Under the assumption that 370 Bq/kg of ^{226}Ra, or 259 Bq/kg of ^{232}Th, or 4810 Bq/kg of ^{40}K, produce the same gamma dose rate level, the radium equivalent activity Ra_{eq} (in Bq/kg) was calculated according to the following equation (Beretka and Mathew, 1985):

$$Ra_{eq} = C_{\mathrm{Ra}} + 1.43C_{\mathrm{Th}} + 0.077C_{\mathrm{K}} \qquad (1)$$

where C_{Ra}, C_{Th} and C_{K} are the activities of ^{226}Ra, ^{232}Th and ^{40}K (in Bq/kg), respectively.

The external hazard index H_{ex}, defined in Beretka and Mathew (1985), was calculated as:

$$H_{ex} = \frac{C_{\mathrm{Ra}}}{370} + \frac{C_{\mathrm{Th}}}{259} + \frac{C_{\mathrm{K}}}{4810} \qquad (2)$$

The maximum value of H_{ex} (equal to 1) corresponds to the upper limit of Ra_{eq} (370 Bq/kg). On the other hand, the radiation hazard is insignificant for the H_{ex} values less than 1.

The external gamma absorbed dose rate in air 1 m above ground D (in nGy/h) was calculated taking into account only the activities of ^{226}Ra, ^{232}Th and ^{40}K, thus assuming that other radionuclides from the ^{238}U series and the anthropogenic ^{137}Cs contribute insignificantly to the total dose from the environmental background (Kocher and Sjoreen, 1985; Leung et al., 1990). The dose was calculated according to UNSCEAR (2000) as:

$$D = 0.462C_{\mathrm{Ra}} + 0.604C_{\mathrm{Th}} + 0.0417C_{\mathrm{K}} \quad (3)$$

To estimate the annual effective dose D_{ann} (in Sv), the conversion coefficient from absorbed dose in air to effective dose and the indoor occupancy factor were taken into account. The conversion factor 0.7 mSv/Gy and the outdoor occupancy factor 0.2 (assuming that, on average, an adult spends 20 % of daily active time outdoors) were used in our calculations (UNSCEAR, 2000). The annual effective dose (for 24 hours and 365 days) was, therefore, calculated as:

$$D_{ann}[\mathrm{mSv}] = 0.7\mathrm{Sv/Gy} \cdot 0.2 \cdot 365 \cdot 24 \cdot D \quad (4)$$

3. Results and Discussion

The measured activities of natural radionuclides in soils in Serbia were mostly in the range of values reported for the region (Popovic et al., 1996, 2008). The differences are due to the type of soil, its geological origin and agricultural treatment. Higher values of uranium were found in soils treated with phosphate fertilisers, as was the case at the experimental university farm Radmilovac (Vukasinovic et al., 2009). Similarly, high values found in Aranđelovac were most likely caused by chemical treatment of soil.

Among the mountain soils, the highest content of natural radionuclides was found in the soils of Mt. Besna Kobila (in the Serbo-Macedonian massif), and the lowest in the soils of Rudnik and Goč (in the Vardar zone) and Crni Vrh (in the Carpatho-Balkanides).

The highest concentrations of uranium in soils were found in the Vranje region, where the sampling sites were relatively close to the target points in 1999. The differences between the sites were pronounced, with the Bratoselce location having the highest activities.

The external dose rates D, radium equivalent activities Ra_{eq}, external hazard indices H_{ex}, and annual effective doses D_{ann} were calculated taking into account the activities of natural radionuclides in soils. The results are given in Table 1. The radium equivalent activity in our study was significantly lower than its maximum of 370 Bq/kg, but some of the external dose rates were above the world average of 57 nGy/h (UNSCEAR, 2000). External hazard index is within the range (0.06–0.56) obtained for Serbia by other authors (Dragovic et al., 2006a,b). The total annual external

dose due to natural radionuclides in soils was below the world average (UNSCEAR, 2000), except for agriculturally treated soils and some mountainous soils with higher content of natural radionuclides.

A notable exception was the region of Vranje, which was exposed to DU ammunition in 1999. Significant differences in radiological risk assessment between the four sampling locations in this region were found. For example, the values of external dose rate and annual effective dose at Bratoselce were five times higher than at Borovac. Further, the annual external dose at Bratoselce was the highest for the country on the whole, and it exceeded the world average value by a factor of two (UNSCEAR, 2000).

Table 1 External dose rates D, radium equivalent activities Ra_{eq}, external hazard indices H_{ex}, and annual effective doses D_{ann} from natural radionuclides in soils in Serbia. Mountain regions, urban areas and the experimental university farm are marked with the M, U, and F superscripts, respectively

Site	D (nGy/h)	Ra_{eq} (Bq/kg)	H_{ex}	D_{ann} (mSv)
Tara^{M}	73±29	155±61	0.42±0.16	0.089±0.036
Šara^{M}	59±10	126±23	0.34±0.06	0.072±0.013
Stara Planina^{M}	79±27	168±59	0.45±0.16	0.097±0.034
Besna Kobila^{M}	98±22	209±47	0.56±0.13	0.120±0.027
Goč^{M}	20±4	43±8	0.12±0.02	0.025±0.004
Rudnik^{M}	40±8	86±18	0.23±0.04	0.049±0.008
Crni Vrh^{M}	31±6	64±13	0.17±0.03	0.038±0.008
Coal Power Plants	69±12	147±25	0.40±0.07	0.084±0.014
Vranje^{U}	69±14	122±30	0.40±0.07	0.085±0.017
Bratoselce	110±31	233±66	0.63±0.18	0.135±0.037
Borovac	22±3	48±6	0.13±0.02	0.027±0.004
Radmilovac^{F}	81±9	173±19	0.47±0.05	0.099±0.011
Belgrade^{U}	50±7	106±15	0.29±0.04	0.061±0.009
Paraćin^{U}	68±7	143±15	0.39±0.04	0.083±0.008
Ćuprija^{U}	60±12	126±27	0.34±0.07	0.073±0.008
Jagodina^{U}	81±16	169±34	0.45±0.09	0.099±0.020
Šabac^{U}	81±16	175±35	0.47±0.09	0.099±0.020
Aranđelovac^{U}	90±18	192±38	0.52±0.10	0.100±0.020

3. Conclusions

Generally, the activities of natural radionuclides in soils in Serbia were in the range of values in the region, with some exceptions, mainly due to anthropogenic activities.

The highest radiological risk was obtained for Mt. Besna Kobila, where the highest concentrations of the natural radionuclides were found. Relatively high radiological risks were found in some urban areas, most likely as a result of soil fertilization. Similar radiological risks were calculated for the Radmilovac farm, where soils are extensively treated. However, in Belgrade, the largest city, relatively low values of radiological risks were found. In the areas around the coal power plants radiological risks were not as high as in some of the urban areas, and were comparable to the average risk in the Vranje area. Differences between the sampling sites in the Vranje region were significant, with the risk parameters varying fivefold.

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