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Distribution of natural radionuclides in surface soils in the vicinity of abandoned uranium mines in Serbia

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Abstract The activity concentrations of natural radionuclides in soils from the area affected by uranium mining at Stara Planina Mountain in Serbia were studied and compared with the results obtained from an area with no mining activities (background area). In the affected area, the activity concentrations ranged from 1.75 to 19.2 mg kg^{-1} for uranium and from 1.57 to 26.9 mg kg^{-1} for thorium which is several-fold higher than those in the background area. The Th/U, K/U, and K/Th activity ratios were also determined and compared with data from similar studies worldwide. External gamma dose rate in the air due to uranium, thorium, and potassium at 1 m above ground level in the area affected by uranium mining was found to be 91.3 nGy h^{-1}, i.e., about two-fold higher than that in background area. The results of this preliminary study indicate the importance of radiological evaluation of the area and implementation of remedial measures in order to prevent further dispersion of radionuclides in the environment.

Keywords Uranium · Thorium · Potassium · Uranium mining · Soil

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Introduction

Estimates of the total radiation dose to the world population have shown that a fraction of about 96 % is from natural sources, while 4 % is from artificial ones (Chougaonkar et al. 2003). External gamma dose contributions are predominantly from the presence of ^{40}K and of ^{232}Th and ^{238}U and their progeny in various rocks and soils. Only minute concentrations of the so-called cosmogenic radioisotopes are present (^{7}Be, ^{14}C, and ^{3}H). Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions. Namely, the specific levels

of terrestrial environmental radiations are related to the type of rocks from which the soils originate (Florou and Kritidis 1992; UNSCEAR 2000; Chiozzi et al. 2002).

The concentration of uranium is often higher in soils in and around uranium mining sites. The radiological impact generally is the main impact of uranium mines. Even when mining activities have ceased, installations at uranium mines can have a significant impact on the environment. Investigations on terrestrial natural radiation near abandoned uranium mines have received particular attention worldwide and led to extensive surveys in many countries (Fernandes et al. 1996; Vandenhove et al. 2006; Carvalho et al. 2007). These results are of importance in making estimations of population exposures and serve as reference information to assess any changes in radioactivity level.

Geologically, the territory of Serbia includes a great number of different magmatic, sedimentary, and metamorphic rock complexes (Dimitrijević 1995). Outstanding differences in natural radioactivity of soils were observed (Dragović et al. 2006). In the present study, the specific activities of primordial radionuclides ^{238}U, ^{232}Th, and ^{40}K were determined in soil samples taken from the mountain of Stara Planina, the largest uranium mineralized area in Serbia. An area affected by uranium mining was characterized radiologically and compared with unaffected areas nearby.

Materials and methods

Study area

Stara Planina, the largest mountain in Serbia, is located in the east of the country and forms a part of the Carpathian–Balkan mountain range, i.e., the western portion of the wider Stara Planina (Mt. Balkan) massif. It covers the area between Zaječar in the northwest and Senokos in the southwest. The western and southeastern border of Stara planina Mt. is spread through the valleys of the rivers Beli Timok (from Zaječar to Knjaževac), Trgoviški Timok, Temska, and Nišava (from Pirot to Dimitrovgrad). The northern border towards the Negotin plain is represented by a stream flowing from Vrška Čuka to the confluence with Timok near Veliki Izvor (Kovačević 2006). Major towns in the region are Zaječar in the north, Knjaževac in the center, and Pirot in the southeast. There are many small villages in the mountain area.

From the geological point of view, Stara Planina Mt. presents an area with rock complexes which are markedly different with respect to age, genesis, mineral content, and petrochemical and geochemical characteristics. The variety of geological characteristics in this area caused the different levels of natural radioactivity of soils.

There were two centrally situated uranium mines on Stara Planina Mt. The mines were working from 1960 until 1966. The maximum production of uranium ore was 200 tonnes per day in Gabrovnica plant and 60 tonnes per day in Mezdreja plant. After the mining activities had been ceased, local population intensified using land around the mines for agricultural production.

Sampling

Samples of undisturbed soils were taken from 30 locations (A1–A30) in the central part of Stara Planina Mt. where the abandoned uranium mines are placed (affected area) and from 25 locations (B1–B25) in the northern part where mining activities have never been performed (background area; Fig. 1). The geographic coordinates of the sampling locations obtained via the global positioning system (Garmin eTrex Vista) are presented in Table 1. From each location three subsamples (0–10 cm) were collected with the distance of 3 m between each other. All soil samples were weighed and air-dried until constant weight was reached. The samples were then pulverized, homogenized, and sieved to pass through a 2-mm mesh. They were kept hermeticaly sealed for 1 month prior to radioactivity measurements.

Radioactivity measurements

Samples were measured in Marinelli beakers of total volume 500 cm^3. The specific activities of primordial radionuclides were measured using HPGe gamma-ray spectrometer (ORTEC-AMETEK, 34 % relative efficiency and 1.65 keV FWHM for ^{60}Co at 1.33 MeV). Sample weight was about 0.5 kg and the counting time for each sample was 60 ks. A mixed calibration source (MBSS 2) from the Czech Metrological Institute was used for efficiency calibration in the same geometry

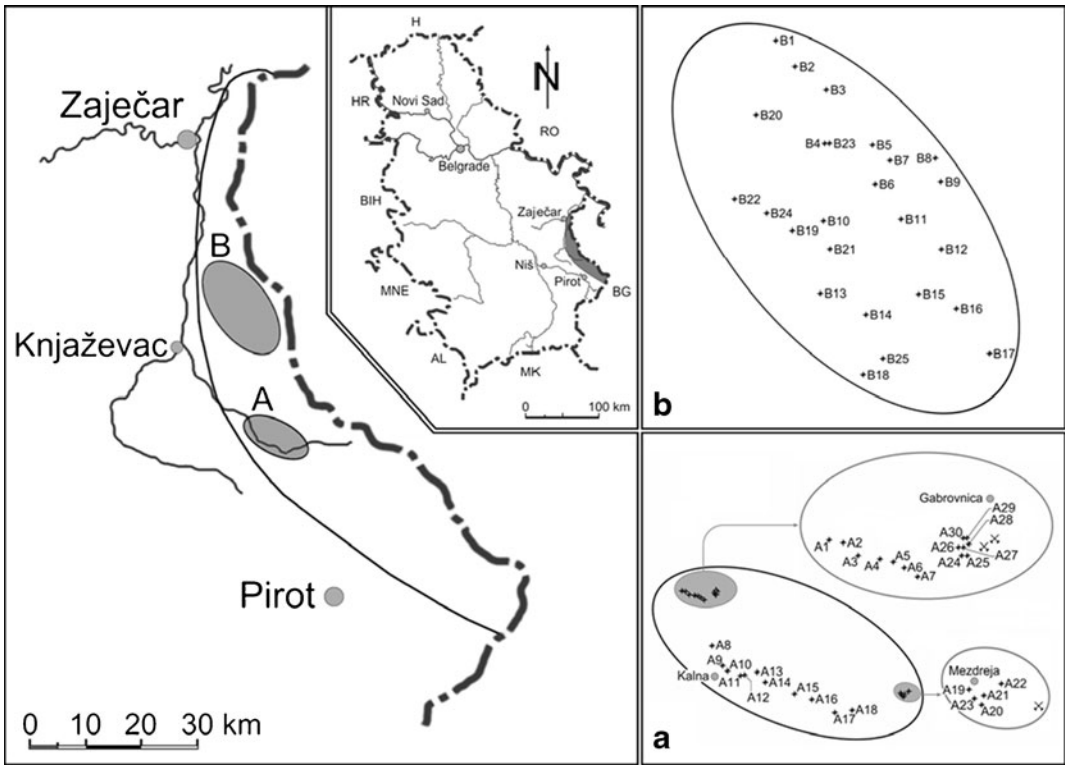

Fig. 1 Simplified map of Stara Planina Mt. showing sampling locations in the affected (**a**) and background (**b**) area

as the soil samples. Quality assurance checks on calibration were performed through the International Atomic Energy Agency (IAEA) proficiency test for the determination of gamma emitting radionuclides (IAEA 2007). Due to the possible disequilibrium between ^{238}U and ^{226}Ra, the activity of ^{238}U was evaluated from all the lines in the spectrum which originate from the isotopes in this series. Since in each of our samples the activity of ^{238}U, as deduced from all the lines, gave consistent values within the uncertainties of their intensities, which justifies the assumption of unperturbed equilibrium in the series, the final results were based on gamma-ray lines of ^{214}Bi at 609.3, 1,120.3, and 1,764.5 keV, which yield the highest statistical accuracy (Hamby and Tynybekov 2002; Al-Jundi 2002; Tzortzis and Tsertos 2004; Dragović et al. 2005). The specific activity of ^{232}Th was evaluated from gamma-ray lines of ^{228}Ac at 338.4, 911.1, and 968.9 keV. The specific activity of ^{40}K was determined from its 1,460.8 keV gamma-ray line. The minimum detectable activity for each radionuclide was determined from the background radiation spectrum for the same counting time as for soil samples and was estimated to be 0.3 Bq kg^{-1} for ^{238}U, 0.7 Bq kg^{-1} for ^{232}Th, and 2.1 Bq kg^{-1} for ^{40}K. Gamma Vision 32 MCA emulation software was used to analyze gamma-ray spectra (ORTEC 2001). The obtained results were evaluated statistically using Statistical Package for the Social Sciences—SPSS 10.0 for Windows (SPSS Inc. 1999). The external gamma dose rates were geographically mapped using ArcGIS from ESRI and Terrain Sculptor (ESRI 2009; Terrain Sculptor 2011).

Calculation of elemental concentrations

Specific activities of ^{238}U, ^{232}Th, and ^{40}K were converted into concentrations of uranium, thorium, and potassium, respectively, according to the following equation:

$$F_E = \frac{M_E \cdot C}{\lambda_{E,i} \cdot N_A \cdot f_{E,i}} \cdot A_{E,i} \quad (1)$$

where F_E is the fraction of element E in the sample, M_E—the atomic mass (kilograms per mole), $\lambda_{E,i}$—the decay constant of the measured isotope i of the

Table 1 Geographic coordinates of sampling locations

Background area			Affected area		
Location	Latitude [N]	Longitude [E]	Location	Latitude [N]	Longitude [E]
B1	43°41′54″	22°22′11″	A1	43°26′23″	22°24′47″
B2	43°41′19″	22°22′46″	A2	43°26′20″	22°24′54″
B3	43°40′48″	22°23′44″	A3	43°26′18″	22°25′00″
B4	43°39′37″	22°23′41″	A4	43°26′17″	22°25′10″
B5	43°39′35″	22°25′09″	A5	43°26′16″	22°25′16″
B6	43°38′43″	22°25′14″	A6	43°26′14″	22°25′21″
B7	43°39′14″	22°25′41″	A7	43°26′11″	22°25′27″
B8	43°39′17″	22°27′06″	A8	43°25′11″	22°25′42″
B9	43°38′46″	22°27′14″	A9	43°24′45″	22°26′01″
B10	43°37′54″	22°23′39″	A10	43°24′38″	22°26′10″
B11	43°37′56″	22°26′01″	A11	43°24′32″	22°26′34″
B12	43°37′16″	22°27′15″	A12	43°24′33″	22°26′41″
B13	43°36′16″	22°23′33″	A13	43°24′36″	22°27′04″
B14	43°35′48″	22°24′57″	A14	43°24′23″	22°27′19″
B15	43°36′15″	22°26′33″	A15	43°24′07″	22°28′12″
B16	43°35′56″	22°27′43″	A16	43°24′00″	22°28′44″
B17	43°34′57″	22°28′46″	A17	43°23′43″	22°29′26″
B18	43°34′28″	22°24′52″	A18	43°23′46″	22°29′58″
B19	43°37′42″	22°22′42″	A19	43°24′09″	22°31′26″
B20	43°40′15″	22°21′35″	A20	43°24′04″	22°31′32″
B21	43°37′16″	22°23′51″	A21	43°24′07″	22°31′33″
B22	43°38′23″	22°20′56″	A22	43°24′11″	22°31′41″
B23	43°39′37″	22°23′50″	A23	43°24′06″	22°31′29″
B24	43°38′04″	22°21′55″	A24	43°26′18″	22°25′50″
B25	43°34′50″	22°25′28″	A25	43°26′18″	22°25′48″
			A26	43°26′21″	22°25′46″
			A27	43°26′21″	22°25′48″
			A28	43°26′22″	22°25′51″
			A29	43°26′24″	22°25′50″
			A30	43°26′24″	22°25′49″

element E (per second), $f_{E,i}$ the fractional atomic abundance in nature, $A_{E,i}$—the measured specific activity (becquerels per kilorgram) of the radionuclide considered (^{238}U, ^{232}Th, or ^{40}K), N_A—Avogadro's number (6.023×10^{23} atoms mol^{-1}), and C—a constant with a value of 1,000,000 for U and Th (concentration in milligrams per kilogram) or 100 for K (concentration in percent of mass fraction) (IAEA 1989).

Calculation of external gamma dose rates

The external gamma dose rate in the air at 1 m above ground level was calculated from the measured activities of ^{238}U, ^{232}Th, and ^{40}K in soil. The calculations of external gamma dose rate, D (nanograys per hour), were performed according to the following equation:

$$D = 0.462A_U + 0.604A_{Th} + 0.042A_K \quad (2)$$

where A_U, A_{Th}, and A_K are specific activities (becquerels per kilogram) of ^{238}U, ^{232}Th, and ^{40}K in soil, respectively, and 0.462, 0.604, and 0.042 conversion factors obtained by calculations based on the polynomial expansion matrix equation, the point-kernel algorithm, Monte Carlo algorithm, or photon transport equations (UNSCEAR 2010).

Results and discussion

The concentrations of uranium, thorium, and potassium in analyzed soils (mean for three sub-samples and corresponding standard deviation) are presented in Table 2. Their descriptive statistics and the type of theoretical frequency distribution that best fits each empirical distribution revealed by Shapiro–Wilk's test (significance level α was 0.05) (Shapiro and Wilk 1965) are summarized in Table 3. In the background area the mean concentrations were found to be 2.08± 0.71 mg kg^{-1} (1.05–3.94 mg kg^{-1}) for uranium, 7.65± 1.98 mg kg^{-1} (4.62–12.6 mg kg^{-1}) for thorium, and 1.87±0.50 % (1.10–2.82 %) for potassium. Much higher concentrations were found in the affected area, i.e., 6.23±4.61 mg kg^{-1} (1.75–19.2 mg kg^{-1}) for uranium, 12.8±5.6 mg kg^{-1} (1.57–26.9 mg kg^{-1}) for thorium, and 1.85±0.54 % (0.21–3.22 %) for potassium. The high standard deviation of uranium concentrations in the affected area indicates their high variability among sampling locations. The frequency distributions of uranium, thorium, and potassium concentrations in soils are shown in Fig. 2. The concentrations of analyzed elements in background area varied by a factor up to four for uranium and up to three for thorium and potassium, and in the affected area by a factor of up to 11, 17, and 15 for uranium, thorium, and potassium, respectively.

Comparing the distribution of uranium and thorium concentrations in affected area with background sites, it is clear that samples from the uranium mine surroundings have overall greater deposition of these radionuclides in the surface soils. Most of the contamination probably came directly from former mining or milling activities via wind and/or hydrological processes or from transport activities around facilities. The uranium and thorium concentrations in the affected area are higher than worldwide average values for these radionuclides as reported by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 2010). The uranium concentration in soils of affected area is also higher than uranium concentration in soils collected from uranium processing and tailings management facility in India, where various safety measures are taken to reduce radiation exposures to levels that are as low as reasonably achievable (Tripathi et al. 2008). The uranium concentrations obtained in this study are similar to those reported for uranium mining and milling sites in Portugal (Carvalho et al. 2007, 2009), Namibia

Table 2 Concentrations of U, Th, and K in soils from different locations in background and affected area of Stara Planina Mt.

Location	Concentration		
	U (mg kg^{-1})	Th (mg kg^{-1})	K (%)
Background area			
B1	1.31±0.10	4.84±0.40	2.19±0.08
B2	3.94±1.20	6.55±0.94	1.93±0.21
B3	1.40±0.19	6.57±3.14	1.96±0.05
B4	1.28±0.06	7.01±0.27	1.59±0.05
B5	1.86±0.04	7.81±0.57	1.36±0.09
B6	2.83±0.84	12.6±3.6	2.62±0.87
B7	2.26±0.20	7.71±0.55	1.56±0.09
B8	1.68±0.16	5.73±0.55	1.81±0.09
B9	1.42±0.14	6.15±0.47	1.27±0.10
B10	3.65±0.20	8.79±0.18	2.15±0.04
B11	1.87±0.06	6.64±0.25	1.17±0.08
B12	1.79±0.12	7.66±0.33	2.42±0.07
B13	1.53±0.19	4.69±0.62	1.10±0.07
B14	3.02±0.16	10.7±1.0	1.55±0.08
B15	2.03±0.22	7.34±0.25	1.80±0.14
B16	2.19±0.05	9.95±0.25	1.91±0.10
B17	2.65±0.09	6.84±0.28	1.17±0.04
B18	1.05±0.12	8.90±0.25	1.60±0.06
B19	2.11±0.13	8.00±0.60	2.82±0.16
B20	1.91±0.07	8.97±0.05	1.75±0.10
B21	1.94±0.41	4.62±0.30	2.27±0.14
B22	2.68±0.16	10.0±1.12	2.55±0.20
B23	2.04±0.06	10.1±0.23	2.07±0.14
B24	1.75±0.28	6.35±0.40	2.68±0.24
B25	1.92±0.05	6.77±0.87	1.46±0.03
Affected area			
A1	3.97±0.16	12.6±0.7	1.76±0.32
A2	3.84±0.14	11.2±0.8	1.73±0.23
A3	15.6±0.8	18.6±0.7	2.32±0.06
A4	8.80±0.43	11.9±1.0	1.94±0.05
A5	6.53±0.29	13.6±0.7	2.24±0.07
A6	6.10±0.61	13.7±0.9	1.83±0.10
A7	5.28±0.55	16.0±0.8	1.85±0.09
A8	1.75±0.05	7.73±0.40	1.33±0.05
A9	6.11±0.48	17.5±0.7	1.92±0.07
A10	3.46±0.17	13.4±0.3	1.96±0.40
A11	2.03±0.21	14.8±0.6	2.12±0.07
A12	2.18±0.24	12.0±0.4	2.05±0.09
A13	5.11±0.23	24.5±0.4	2.41±0.11
A14	8.38±0.59	10.1±1.1	1.54±0.05
A15	7.55±0.40	7.36±0.33	1.30±0.02

Table 2 (continued)

Location	Concentration		
	U (mg kg^{-1})	Th (mg kg^{-1})	K (%)
A16	3.22±0.14	7.97±0.49	1.38±0.02
A17	6.94±0.43	10.8±1.0	2.23±0.06
A18	9.56±0.24	10.8±0.8	2.22±0.09
A19	3.86±0.29	12.5±0.4	1.88±0.10
A20	18.9±0.6	26.9±1.4	3.22±0.21
A21	9.78±0.55	14.0±0.5	1.73±0.07
A22	4.38±0.24	16.0±0.4	1.93±0.11
A23	2.78±0.09	6.09±0.24	1.67±0.06
A24	19.2±2.2	23.8±0.8	2.99±0.09
A25	4.16±0.13	15.0±0.6	1.60±0.05
A26	2.50±0.09	6.60±0.25	1.32±0.04
A27	7.27±0.21	11.5±0.4	1.54±0.05
A28	2.51±0.08	1.57±0.10	0.21±0.01
A29	2.62±0.09	7.45±0.27	1.40±0.05
A30	2.66±0.09	7.90±0.30	1.90±0.06

(Oyedele et al. 2010), and Brazil (Fernandes et al. 2006). About ten-fold higher values have been reported for uranium mining areas in Kyrgyzstan (Vandenhove et al. 2006) and Colorado, United States (Rood et al. 2008). Uranium concentrations in soils collected in the vicinity of abandoned uranium mines in Spain (Blanco et al. 2005) and Australia (Hancock et al. 2006) were found to be about 30-fold higher than uranium concentrations obtained in this study. Thorium concentrations obtained in this study fall into the range of concentrations reported for uranium mining area in Brazil (Fernandes et al. 2006) and were about three-fold lower than those reported for uranium mining sites in Portugal (Carvalho et al. 2009) and Namibia (Oyedele et al. 2010).

The original uranium, thorium, and potassium concentrations in rocks may vary because of alteration or metamorphic processes (Verdoya et al. 2001). Figure 3 shows the Th/U, K/U, and K/Th ratios, which may indicate whether relative depletion or enrichment of radionuclides had occurred. The best-fitting relations between Th and U, K and U, and K and Th are of the linear type with correlation coefficients of 0.412, 0.198, and 0.285 for the background area, and 0.652, 0.660, and 0.845 for the affected area, respectively. In the background area the Th/U ratio was 4.11±0.21 which is close to the value expected for a normal continental crust 3.7–4.0 (Van Schmus 1995). The Th/U ratio for the background area obtained in this study indicates the lack of metasomatic activity of analyzed radionuclides (Chiozzi et al. 2002). The K/U ratio of 0.72±0.06 and that of K/Th of 0.23±0.01 in soils of background area were consistent to the typical values obtained in a large variety of unaltered lithologies, e.g., to those calculated from data reported by Galbraith and Saunders (1983) (K/U=0.63, K/Th=0.21), Chiozzi et al. (2002) (K/U=0.94, K/Th=0.25), and Dragović et al. (2006) (K/U=0.76, K/Th=0.50). In soils collected from the affected area the Th/U ratio was found to be 1.72±0.20. The Th/U ratio obtained

Table 3 Descriptive statistics of concentrations of U, Th, and K in soils from background and affected area

Parameter	Concentration					
	Background area			Affected area		
	U (mg kg^{-1})	Th (mg kg^{-1})	K (%)	U (mg kg^{-1})	Th (mg kg^{-1})	K (%)
Range	2.89	7.98	1.72	17.4	25.3	3.01
Mean	2.08	7.65	1.87	6.23	12.8	1.85
Standard deviation	0.71	1.98	0.50	4.61	5.59	0.54
Minimum	1.05	4.62	1.10	1.75	1.57	0.21
Maximum	3.94	12.6	2.82	19.2	26.9	3.22
Median	1.92	7.34	1.81	4.74	12.2	1.86
Mode	1.05	4.62	1.17	1.13	10.8	1.54
Skewness	1.113	0.589	0.269	1.719	0.731	−0.146
Kurtosis	1.131	0.193	−0.859	2.665	0.905	2.968
Distribution	Normal	Log-normal	Log-normal	Log-normal	Normal	Normal

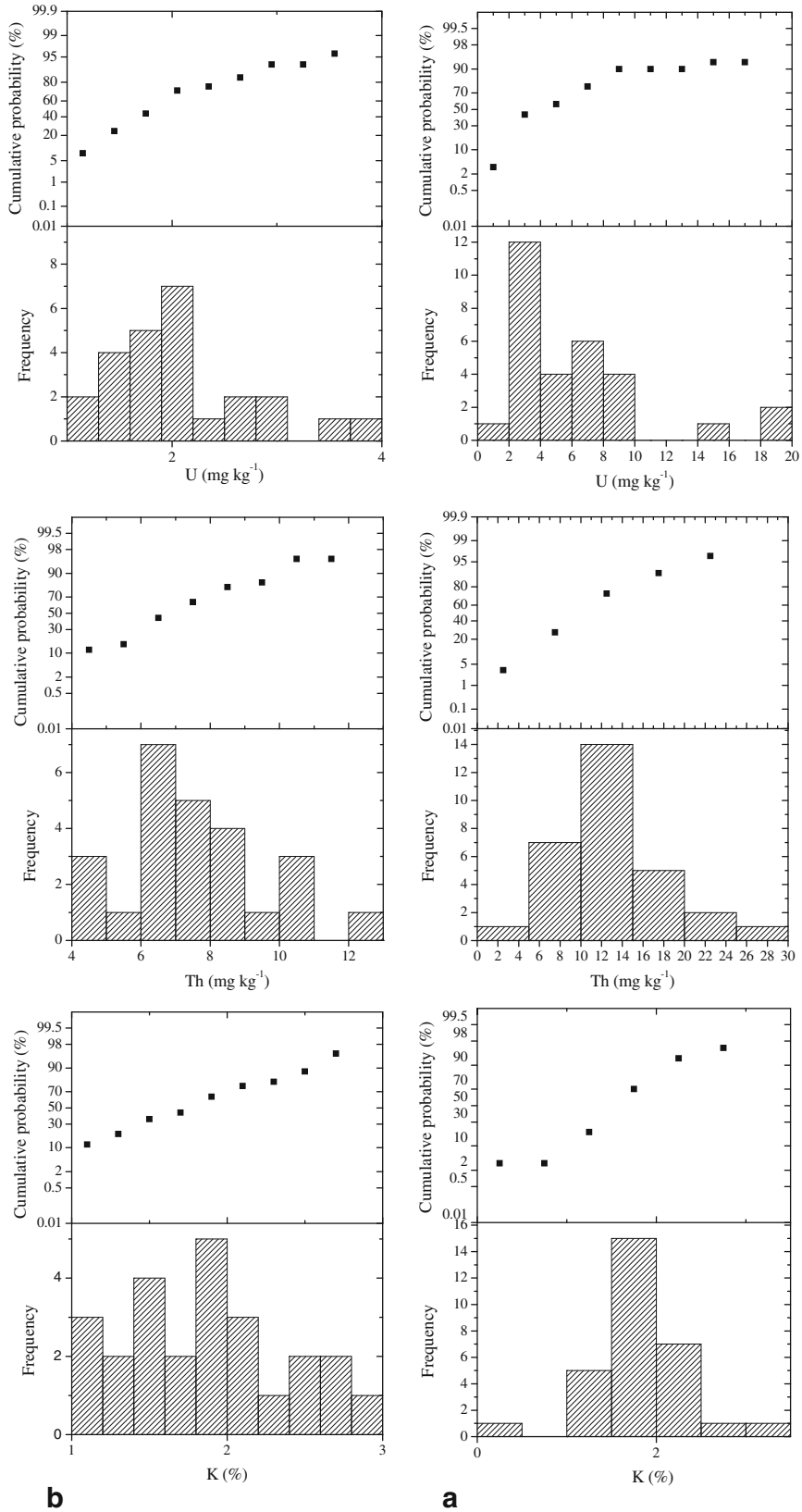

Fig. 2 Frequency distributions of uranium, thorium and potassium concentrations in soils collected from affected (**a**) and background (**b**) area

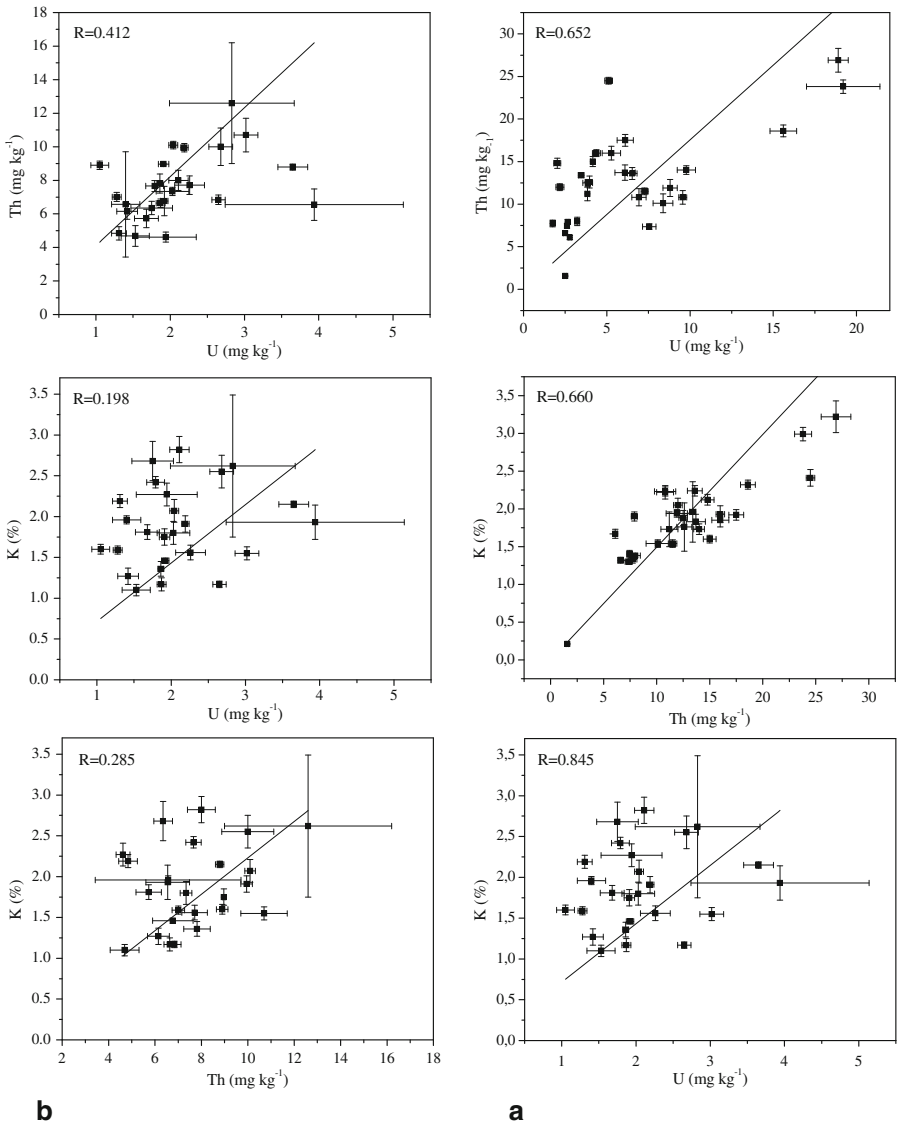

Fig. 3 Th versus U, K versus U, and K versus Th average concentrations in the affected (**a**) and background (**b**) area

in this study is comparable to that of 1.52 for soils of residential area impacted by mining and milling activites in Texas, United States (McConnel et al. 1998) and also to that of 2.10 for soils near the uranium mine in Portugal (Carvalho et al. 2009). About two-fold higher values of soil Th/U ratio have been found for uranium mining site located in a semi-arid region in Brazil (4.20; Fernandes et al. 2006), in surface soils around proposed uranium mining site at Lambapur, India (3.80; Sartandel et al. 2009), and in soils around high-uranium mineralization zone in Meghalaya, India (3.56; War et al. 2008). The K/U ratio was calculated to be equal to 0.22±0.02 for affected area which is several-fold higher than values obtained for Naberlek uranium mine in Australia (0.01) (Hancock et al. 2006) and high-uranium mineralization area in India (0.04) (War et al. 2008). The K/Th ratio was found to be 0.15±0.01 for the affected area in this study. This value is about ten-fold higher than that obtained by War et al. (2008). It should be mentioned that K/U and K/Th values are highly variable in soils worldwide (Roger and Adams 1969; Chiozzi et al. 2002; Tzortzis and Tsertos 2004).

The comparison of external gamma dose rates due to analyzed radionuclides in background and affected area is presented in Fig. 4. The mean gamma dose rate in the background area was very close to the world-wide average of 54 nGy h^{-1} (UNSCEAR 2010). The

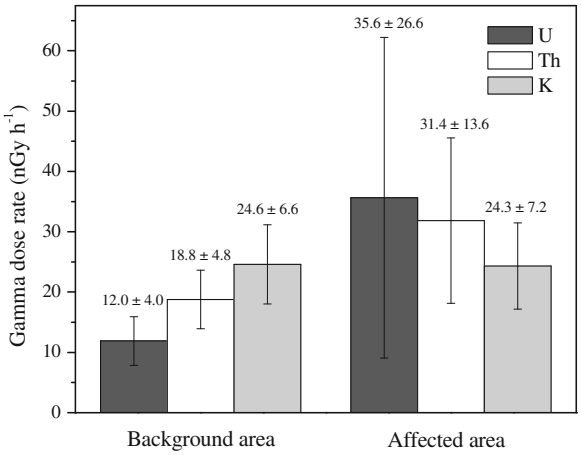

Fig. 4 The external gamma dose rates in the air at 1 m above ground level in background and affected area

mean gamma dose rates in the affected area were 3 and 1.7 times higher than those in the background area for ^{238}U and ^{232}Th, respectively. The mean total gamma dose rate in this area was almost two-fold higher than that in background area, with the highest values in the immediate vicinity of abandoned uranium mines (Fig. 5). The total external gamma dose rate in the area affected by uranium mining activities (91.3 nGy h^{-1}) was also higher than total gamma dose rate for Serbia of 62.8 nGy h^{-1} reported by Dragović et al. (2007).

Results presented in this preliminary study confirm that abandoned uranium mines are of particular concern with respect to radioactive contamination of the environment and require continuous monitoring. These areas are susceptible to cause serious disturbance in the trophic chains which could reflect at the ecosystem level. This study pointed out that more detailed radiological characterization of the area including a wider range of environmental samples (water, sediments, and plants) and contaminants is needed in order to evaluate an overall impact of mining activities on the environment. In addition to radionuclides, trace elements scattered in the area affected by uranium mining can also contribute to an extensive contamination of the soil compartment posing serious risks to humans that use surrounding areas for agruculture purposes as well as to edaphic communities which are extremely important for the future recovery of the area. It has been demonstrated that highly toxic heavy metals associated with uranium mining are a major source of surface and groundwater contamination (Neves and Matias 2008). The presence of complex mixtures of contaminants in sediments of mining areas and the potential for toxicological interactions among them are also documented by numerous studies (Bridges et al. 1996; Hancock et al. 2006; Antunes et al. 2007). According to increasing concern on protection of non-human biota, the risk assessment studies

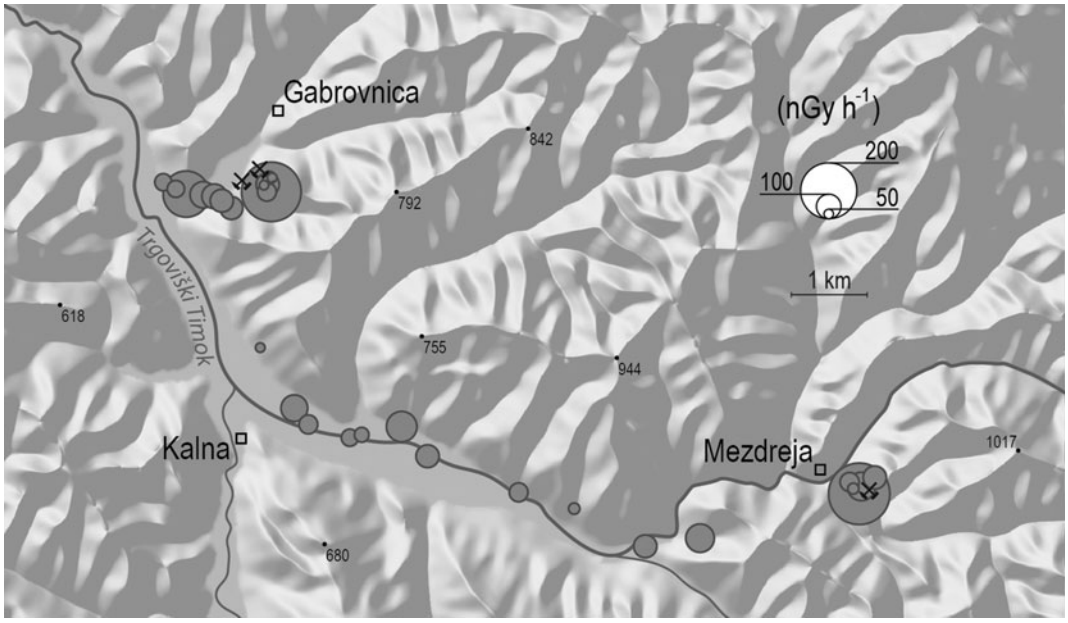

Fig. 5 The distribution map of the total external gamma dose rate in the air at 1 m above ground level in the affected area

should be strengthened by data on transfer of contaminants to plants and animals. Recent studies have demonstrated selenium uptake by vegetation on uranium mine overburden and on land irrigated with groundwater from uranium mining areas (Baumgartner et al. 2000; Sharmasarkar and Vance 2002). The bioaccumulation of radionuclides and trace elements associated with uranium mining activities by vertebrates and invertebrates have also been documented by risk assessment studies (Pyle et al. 2001; Peterson et al. 2002).

Conclusions

Results obtained in this study confirmed the elevated radionuclide concentrations in the area affected by uranium mining activities. The uranium and thorium concentrations in soils from the affected area were found to be up to 19.2 and 26.9 mg kg^{-1}, respectively, i.e., several-fold higher than those in soils of background area. External gamma dose rate obtained in this study is about two-fold higher than worldwide average. Further investigation of this area is needed in order to assess the integrated risk from radionuclides and trace elements associated with mining activities in different compartments of the environment. The analysis should be strengthened with data from ecotoxicological screening. Since contaminants mobility vary strongly depending on soil type and physical and chemical properties, the risk assessment should include these site-specific information.

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References

Al-Jundi, J. (2002). Population doses from terrestrial gamma exposure in areas near to old phosphate mine, Russaifa, Jordan. *Radiation Measurements, 35*, 23–28.

Antunes, S. C., Pereira, R., & Gonçalves. (2007). Evaluation of the potential toxicity (acute and chronic) of sediments from abandoned uranium mine ponds. *Journal of Soils and Sediments, 7*, 368–376.

Baumgartner, D. J., Glenn, E. P., Moss, G., Thompson, T. L., Artiola, J. F., & Kuehl, R. O. (2000). Effect of irrigation water contaminated with uranium mill tailings on Sudan grass, *Sorghum vulgare* var. s*udanense*, and fourwing saltbush, *Artiplex canescens*. *Arid Soil Research and Rehabilitation, 14*, 43–57.

Blanco, P., Vera Tomé, F., & Lozano, J. C. (2005). Fractionation of natural radionuclides in soils from a uranium mineralized area in the south-west of Spain. *Journal of Environmental Radioactivity, 79*, 315–330.

Bridges, T. S., Soares, A. M. V. M., Girling, A., Bradley, M. C., & Callow, P. (1996). Chronic toxicity of Great Lakes sediments to *Daphnia magna*: elutriate effects on survival, reproduction and population growth. *Ecotoxicology, 5*, 83–102.

Carvalho, F. P., Madruga, M. J., Reis, M. C., Alves, J. G., Oliveira, J. M., Gouveia, J., et al. (2007). Radioactivity in the environment around past radium and uranium mining sites of Portugal. *Journal of Environmental Radioactivity, 96*, 39–46.

Carvalho, F. P., Oliveira, J. M., & Malta, M. (2009). Analyses of radionuclides in soil, water, and agriculture products near the Urgeiriça uranium mine in Portugal. *Journal of Radioanalytical and Nuclear Chemistry, 281*, 479–484.

Chiozzi, P., Pascale, V., & Verdoya, M. (2002). Naturally occurring radioactivity at the Alps–Apennines transition. *Radiation Measurements, 35*, 147–154.

Chougaonkar, M. P., Eppen, K. P., & Ramachandran, T. V. (2003). Profiles of doses to population living in the high background radiation areas in Kerala. *Journal of Environmental Radioactivity, 71*, 275–297.

Dimitrijević, M. (1995). *Geology of Yugoslavia*. Belgrade: Geoinstitute (in Serbian).

Dragović, S., Onjia, A., Stanković, S., Aničin, I., & Bačić, G. (2005). Artificial neural network modelling of uncertainty in gamma-ray spectrometry. *Nuclear Instruments and Methods in Physics Research A, 540*, 455–463.

Dragović, S., Janković, L., Onjia, A., & Bačić, G. (2006). Distribution of primordial radionuclides in surface soils from Serbia and Montenegro. *Radiation Measurements, 41*, 611–616.

Dragović, S., Janković-Mandić, L., Momčilović, M., & Onjia, A. (2007). Population doses from terrestrial gamma exposure in Serbia. *Archive of Oncology, 15*, 78–80.

ESRI (2009). Environmental Systems Research Institute Inc., Redlands, CA, <http://www.esri.com/>.

Fernandes, H. M., Franklin, M. R., Veiga, L. H. S., Freitas, P., & Gomiero, L. A. (1996). Management of uranium mill tailing: geochemical process and radiological risk assessment. *Journal of Environmental Radioactivity, 30*, 69–95.

Fernandes, H. M., Lamego Simoes Filho, F. F., Perez, V., Franklin, M. R., & Gomiero, L. A. (2006). Radioecological characterization of a uranium mining site located in a semi-arid region in Brazil. *Journal of Environmental Radioactivity, 88*, 140–157.

Florou, H., & Kritidis, P. (1992). Gamma radiation measurements and dose rate in the coastal areas of a volcanic island, Aegean Sea, Greece. *Radiation Protection Dosimetry, 45*, 277–279.

Galbraith, J. H., & Saunders, D. F. (1983). Rock classification by characteristics of aerial gamma-ray measurements. *Journal of Geochemical Exploration, 18*, 49–73.

Hamby, D. M., & Tynybekov, A. K. (2002). Uranium, thorium and potassium in soils along the shore of lake Issyk-Kyol in the Kyrghyz Republic. *Environmental Monitoring and Assessment, 73*, 101–108.

Hancock, G. R., Grabham, M. K., Martin, P., Evans, K. G., & Bollhöfer, A. (2006). A methodology for the assessment of

rehabilitation success of post mining landscapes—sediment and radionuclide transport at the former Nabarlek uranium mine, Northern Territory, Australia. *Science of the Total Environment, 354*, 103–119.

IAEA (1989). Construction and use of calibration facilities for radiometric field equipment, Technical Report Series No. 309, Vienna.

IAEA (2007). Report on the IAEA-CU-2006-03 World-Wide Open proficiency Test on the Determination of Gamma Emitting Radionuclides, Seibersdorf.

Kovačević, J. (2006). Metalogeny of Stara Planina Mt. region. Faculty of Mine and Geology, Belgrade, PhD Thesis (in Serbian).

McConnel, M. A., Ramanujam, V. M. S., Alcock, N. W., Gabehart, G. J., & Au, W. W. (1998). Distribution of uranium-238 in environmental samples from a residential area impacted by mining and milling activities. *Environmental Toxicology and Chemistry, 17*, 841–850.

Neves, O., & Matias, M. J. (2008). Assessment of groundwater quality and contamination problems ascribed to an abandoned uranium mine (Cunha Baixa region, Central Portugal). *Environmental Geology, 53*, 1799–1810.

ORTEC (2001). Gamma Vision 32, Gamma-Ray Spectrum Analysis and MCA Emulation, Version 5.3., Oak Ridge, USA.

Oyedele, J. A., Shimboyo, S., Sitoka, S., & Gaoseb, F. (2010). Assessment of natural radioactivity in the soils of Rössing Uranium Mine and its satellite town in western Namibia, southern Africa. *Nuclear Instruments and Methods in Physics Research A, 619*, 467–469.

Peterson, M. J., Smith, J. G., Southworth, G. R., Ryon, M. G., & Eddlemon, G. K. (2002). Trace element contamination in benthic macroinvertebrates from a small stream near a uranium mill tailings site. *Environmental Monitoring and Assessment, 74*, 193–208.

Pyle, G. G., Swanson, S. M., & Lehmkuhl, D. M. (2001). Toxicity of uranium mine-receiving waters to caged fathead minnows, *Pimephales promelas*. *Ecotoxicology and Environment Safety, 48*, 202–214.

Roger, J. J. W., & Adams, J. A. S. (1969). Uranium. In K. H. Wedephol (Ed.), *Handbook of geochemistry*. Berlin: Springer.

Rood, A., Voillequé, P. G., Rope, S. K., Grogan, H. A., & Till, J. E. (2008). Reconstruction of atmospheric concentrations and deposition of uranium and decay products released from the former uranium mill at Uravan, Colorado. *Journal of Environmental Radioactivity, 99*, 1258–1278.

Sartandel, S. J., Jha, S. K., Bara, S. V., Tripathi, R. M., & Puranik, V. D. (2009). Spatial distribution of uranium and thorium in the surface soils around proposed uranium mining site at Lambapur and its vertical profile in the Nagarjuna Sagar Dam. *Journal of Environmental Radioactivity, 100*, 831–834.

Shapiro, S. S., & Wilk, M. B. (1965). An analysis of variance test for normality (complete samples). *Biometrika, 52*, 591–611.

Sharmasarkar, S., & Vance, G. F. (2002). Ecological risk assessment: soil and plant selenium at a reclaimed uranium mine. *Journal of Environmental Quality, 31*, 1516–1521.

SPSS Inc. (1999). Statistical Package for the Social Sciences (SPSS). Chicago, Illinois, <http://www.spss.com>.

Terrain Sculptor (2011). Jenny, B., Institute of Cartography, ETH Zurich, Leonowitz, A., Department of Geography, University of Zurich, Zurich, <http://www.terraincartography.com/terrainsculptor>.

Tripathi, R. M., Sahoo, S. K., Jha, V. N., Khan, A. H., & Puranik, V. D. (2008). Assessment of environmental radioactivity at uranium mining, processing and tailings management facility at Jaduguda, India. *Applied Radiation and Isotopes, 66*, 1666–1670.

Tzortzis, M., & Tsertos, H. (2004). Determination of thorium, uranium and potassium elemental concentrations in surface soils in Cyprus. *Journal of Environmental Radioactivity, 77*, 325–338.

UNSCEAR (2000). Sources and Effects of Ionizing Radiation. Report to General Assembly, with Scientific Annexes, United Nations, New York.

UNSCEAR. (2010). *Sources and effects of ionizing radiation, UNSCEAR 2008 report*. New York: United Nations.

Van Schmus, W. R. (1995). Natural radioactivity of the crust and mantle. In T. J. Ahrens (Ed.), *Global earth physics: a handbook fo physical constants. American Geophysical Union*, AGU Reference Shelf 1, pp. 283–291.

Vandenhove, H., Sweeck, L., Mallants, D., Vanmarcke, H., Aitkulov, A., Sadyrov, O., et al. (2006). Assessment of radiation exposure in the uranium mining and milling area of Mailuu Suu, Kyrgyzstan. *Journal of Environmental Radioactivity, 88*, 118–139.

Verdoya, M., Chiozzi, P., & Pasquale, V. (2001). Heat-producing radionuclides in metamorphic rocks of the Brianconnais-Piedmont Zone (Maritime Alps). *Eclogae Geologicae Helvetiae, 94*, 1–7.

War, S. A., Nongkynrih, P., Khathing, D. T., Iongwai, P. S., & Jha, S. K. (2008). Spatial distribution of natural radioactivity levels in topsoil around the high-uranium mineralization zone of Kylleng-Pyndensohiong (Mawthabah) areas, Wst Khasi Hills District, Meghalaya, India. *Journal of Environmental Radioactivity, 99*, 1665–1670.