

INVESTIGATION OF ACCESSORY ELEMENTS OF REPRESENTATIVE PETROLOGIC RADIOACTIVITY CARRIERS AT STARA PLANINA, SERBIA

by

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The present study deals with the abundance of the most common accessory elements of radioactivity carriers on the Stara Planina mountain at four defined locations. All investigated locations have increased radioactivity, greater than 200 cps, up to 1250 cps. In all examined samples the following elements were detected and their concentrations were determined: Zr, Rb, W, Mo, Sn, Zn, Cu, As, Sb, Ba, Ni, Cr, V, and Ti. The analysis showed that depending on the sample, elevated concentrations of all detected elements except titanium were found. As all localities containing the listed detected elements are located near watercourses, all the present elements, and in particular, those with elevated concentrations can be relatively easily transferred to the environment by water action. Likewise, there is the possibility of eolic erosion from the investigated deposits and tailing dams whereby these chemical elements and/or radionuclides would be distributed to areas away from the primary sources of natural radioactivity. These have a high risk of spreading and therefore have harmful or radioecological effects on the environment. The study indicated a need for adequate monitoring, and risk assessment of the examined locations, which could prevent the distribution of these elements further into the environment.

Key words: accessory element, radioactivity, toxicity, biohazard, environment

INTRODUCTION

Humans are exposed to natural radioactivity, which represents continuous radioactive radiation from extra-terrestrial and terrestrial sources [1]. Almost 90 % of radiation exposure arises from natural sources [2]. The extra-terrestrial source represents the cosmic ray-produced radioisotopes continuously generated by the interactions of cosmic rays with the atmosphere and crust constituents [1, 3, 4]. Radionuclides, which were incorporated into the Earth at the time of its formation, are still present because of their long half-lives and represent the second source. The main radioactivity carriers are uranium and thorium isotopes (²³⁸U, ²³⁵U, and ²³²Th) as the primordial radionuclides and their radioactive decay products as the secondary radionuclides [1, 3, 4]. The main non-chain primordial radionuclides

in the Earth's crust of particular interest are ⁴⁰K, ⁸⁷Rb, and ⁹⁶Zr [4, 5]. Natural radioactivity mainly comes from the ²³⁸U, ²³⁵U, and ²³²Th decay series, and natural ⁴⁰K [6-8]. The presence of these isotopes in nature is related to their long half-lives, for example, more than 10⁸ years for ²³⁵U [4]. Consequently, all naturally occurring radionuclides are contained in rocks, soils, water and groundwater resources, living organisms, building materials, etc. [4]. Radionuclides present in soil significantly affect terrestrial radiation levels [9]. Since they are not uniformly distributed, the knowledge of its concentration and radiation levels in the environment, especially in particular localities, is important for assessing the effects of human exposure [10]. Depending on the rock or soil, within the uranium deposits often there are trace elements, radionuclides or stable isotopes. The known carriers of radioactivity are granites, graphitic schists, sandstones and siltstones [11-13]. The primary minerals are quartz, oligoclase, K-feldspar (microcline,

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rarely orthoclase), and biotite. The accessory minerals that have been formed during the solidification of the rocks from the magma represent sphene (titanite), apatite, zircon, and magnetite. Likewise, the secondary minerals such as sericite, chlorite, epidote, calcite, limonite, tourmaline, zircon, plagioclase (albite-oligoclase), microcline, biotite, allanite, muscovite, and hematite represent the minerals formed by the alteration of a pre-existing primary mineral in igneous rock [14]. The composition and quantity of minerals depend on the rock type [14, 15]. The primary, accessory, and secondary minerals might contain stable or radioactive isotopes of some elements, also the main radioactive carriers and/or toxic elements that could be hazardous to the environment and human health, as well as the other living beings [16]. Under atmospheric conditions and transfer to the environment by water action, they might have influence even beyond their place of origin. The potential health impacts of uranium ores or deposits could include the effects of the accessory elements (trace elements), many of which are commonly regarded as toxic to human health when concentrated, *e. g.* arsenic, lead, nickel, selenium, etc. Sulfide minerals associated with breccia pipe deposits have the potential to produce acidic waters [16].

The present study is the extension of geological and radioactivity investigation of the Stara Planina mountain in the Republic of Serbia in order to systematize all collected data. In December 2008, Stara Planina was designated a nature park, *i. e.* protected area. These areas include places where people live or spend holidays [17]. The impact of natural radioactivity on the environment has not been systematically processed in the Stara Planina area [18, 19]. Regarding this, accessory elements of representative petrologic radioactivity carriers (primarily of the ^{235}U isotope) were investigated. In addition to uranium, the various elements such as Mg, Mn, V, Cr, Cd, Ni, Cu, Zn, As, Mo, W, Fe, Cd, might be found, due to available data on their presence, in soils and/or aquatic ecosystems located on former uranium mines, tailing dumps or deposits as they are commonly associated with uranium enrichment in sediments near uranium layers. The lack of defined distribution limits of the accessory elements of the natural radioactivity carriers in this ecosystem indicates the necessity for a comprehensive study in order to assess its impact on living beings and the environment, taking into account activities typical for this region, *e. g.* livestock breeding, dairy farming, herb production, *etc.*, and possibly transferred contamination [20-23].

EXPERIMENTAL PART

Main characteristics and location of the investigated area

The Stara Planina mountain is known for its deposits and numerous occurrences of uranium mineralization. Natural radioactivity on Stara Planina, whose

smaller western part is located in the Republic of Serbia, has been examined since the middle of the last century. At the end of the 20th century, the waters of the so-called Colorful Series and the ecological impact of the former Kalna mines were examined. Since 2000, non-systematic tests of radioactivity and distribution of natural radioactivity carriers (mostly ^{238}U , ^{232}Th , and ^{40}K isotopes) and their environmental impact have been conducted [24-26]. Previous investigation as part of the study of natural radioactivity on Stara Planina conducted in 2016 and 2017 was related to the determination of characteristic geological members of radioactivity carriers in the area of Stara Planina, generally granites and related geological units, graphitic schists and sedimentary material represented by reddish sandstones and gray siltstones of the Colorful Series [27].

The study reported in this paper particularly focused on the presence of non-ferrous and other metals at the previously investigated [27] locations with radioactivity greater than 200 cps, based on an investigation over several decades, from geochemical and environmental viewpoints [19, 27]. In the earlier reported study [27] the radioactivity at the observation points was measured in counts per second (cps) units since the hand-held radiation survey instrument was used.

The investigation included four locations at Stara Planina, fig. 1. The first two locations are natural

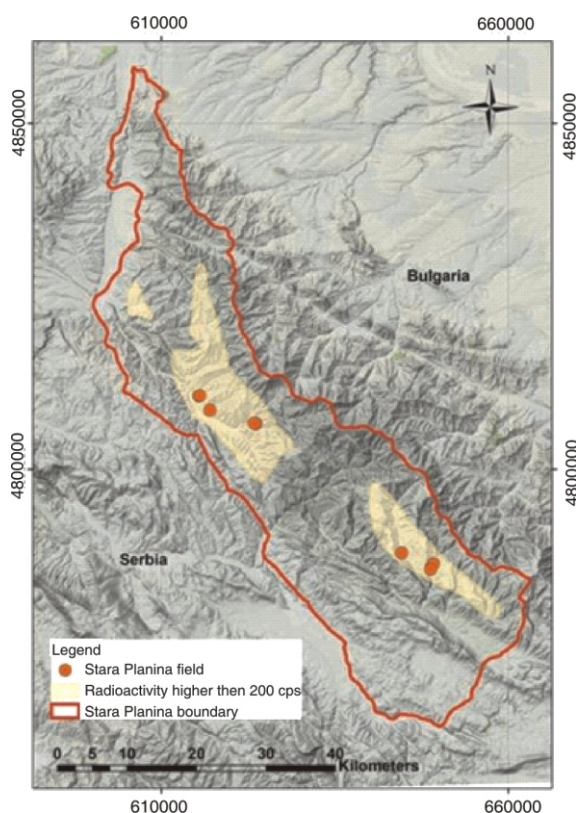


Figure 1. The boundary of Stara Planina in Serbia (full line), zones of radioactivity greater than 200 cps (clear), and points of observation and testing in 2016 and 2017 (dots) [27]. Raster basis Google Landscape w/n, SASPlanet

uranium ore deposits, where ore exploration was carried out. These locations are granites in the vicinity of the village of Janje, and the Mezdreja abandoned mines. In addition to natural radioactivity, they also represent an anthropogenic source of radioactivity in the sense that mining activities have established tailing dumps, *i. e.* brought the radioactive carriers to the surface. Uranium mining creates so-called TENORM (Technically Enhanced Naturally Occurring Radioactive Material) [28], which in this way, has become exposed to direct atmospheric influence and might be transferred, and spread to the environment, especially by water action. The other two sampling points are graphitic schists, *i. e.* the metamorphic Inovo Series which transgresses the southwestern part of the Janja granite-metamorphic system, located in the vicinity of the Gabrovnica deposit, and sedimentary materials of the Early Triassic, represented by red sandstone and gray alevrolites of the Colorful Series, both usually charged with radioactive and trace elements [29] in the Jelovica-Dojkinci river region.

In a previous study, the radioactivity of these locations was measured [27]. Radioactivity in Gabrovnica was 240 cps and the radiation dose rate was 0.210 Sv h^{-1} at the mine portal, and 254 and 360 cps and 0.248 Sv h^{-1} at the mine dump. Radioactivity in Mezdreja was 420 cps and 0.322 Sv h^{-1} at the mine portal, and up to 1250 cps and 0.421 Sv h^{-1} at the mine dump. Radioactivity at the sampling site in the Inovo Series area was 650 cps and $0.279 \mu\text{Sv h}^{-1}$. Radioactivity in the Colorful Series area at the redox zone was 280 cps and $0.429 \mu\text{Sv h}^{-1}$ [27].

Qualitative and quantitative analysis

Clearly defined petrologic units were sampled in situ from outcrops and mine dumps, at pre-determined locations that were 100 m relative to the map and registered by GPS with an error of 5 m.

In total, 16 samples were collected and assays performed by X-ray fluorescence spectroscopy (XRF) on a Niton XL3t Gold+ analyzer (Thermo Fisher Scientific, USA) in two modes – “Soil” and “Test All Geo” (*i. e.* “soil and mining” mode), to check and compare the results [27]. The “Test All Geo” mode automatically applies the correct measurement algorithm for elements both at low, as well as elements at high concentrations, *i. e.* automatically determines the correct analytical test mode for rapid analysis of major, minor and trace elements in geological samples. The excitation time was about three minutes per sample. The samples were ground to 70 μm .

All the samples were collected in areas of elevated radioactivity, where the radiation detector Exploranium GR-110, Exploranium Radiation Detection Systems, Canada, measured 200 cps or more and the hand-held Gamma-Scout radiation detector,

Gamma-Scout GmbH & Co.KG, Germany, measured $0.230 \mu\text{Sv h}^{-1}$ or more [27], near the ground surface (on the ground or maximum 5-10 cm above). The background radiation level in the study area was 70 cps and $0.135 \mu\text{Sv h}^{-1}$ [27].

Seven samples were collected from the Janja granite and the Gabrovnica and Mezdreja mines (from the ground surface). Four samples of partially altered granite were collected near Mezdreja and Gabrovnica, one from the Mezdreja tailing dump that contained clayey and cataclazed granitic material, one from a highly silicified, carbonized and limonitized vein near Mezdreja, and one from monomineral grains of pink K-feldspar. Three samples were collected from graphitic metamorphic rocks of the so-called Inovo Series. It should be noted that the composition varied over relatively small distances (decimeter scale); the concentrations of silicate and graphitic materials differed, as did occurrences of limonitization on sheared surfaces. Six samples were collected from the Early Triassic batch of sediments, *i. e.* from the Colorful Series in the vicinity of the Jelovica-Dojkinci rivers: three of gray siltstone and three of reddish-pink sandstone.

In this phase of the study, the following elements were measured: Mo, Zr, Sr, Rb, Pb, Au, Se, As, Hg, W, Cu, Ni, Co, Fe, Mn, Ba, Sb, Sn, Cd, Pd, Ag, Nb, Bi, Re, Ta, Hf, Cr, V, Ti, Sc, S, Cs, and Te. The ppm units ($1 \text{ ppm} = 1 \text{ mg kg}^{-1}$) were used as a measure of concentration for ease of comparison with the literature data related to the trace element concentration in the Earth's crust.

RESULTS AND DISCUSSION

In all four locations, the following elements were detected: Zr, Rb, W, Mo, Sn, Zn, Cu, As, Sb, Ba, Ni, Cr, V, and Ti.

Table 1 shows the results from granitic samples in the vicinity of the Mezdreja and Gabrovnica mines for Zr, Rb, W, Sn, Zn, Ba, Sb and Ni in ppm. Explanations of the sample abbreviations in the tables are: Mzd Granite Mine – sample of granite taken from the outcrop at the entrance of the Mezdreja mine; Mzd Granite – sample of granite taken from the outcrop at the Mezdreja area; Mzdr Kfeld Gabrr – assay done on large (3 cm) pink K-feldspar from gabbroid at the Mezdreja area; Mzd Sil Lim – a silicified and highly limonitized sample with carbonates, collected in situ from altered granite with veinlets at Mezdreja; Mzd Tail Dump – crushed altered granite fragments with clay from the Mezdreja tailing dump; Gabr Granite Mine – granite taken from an outcrop at the entrance of the Gabrovnica mine; Gabr Tail Dump – a granite sample taken from the Gabrovnica mine tailing dump.

Tables 2 and 3 show results from granitic samples for Zr, Rb, W, Sn and graphitic schist for Zn, Ba, Sb, Ni, Mo, Cu, and As in Inovo, in ppm. Explanations of the sample abbreviations in the tables are: Inov

Table 1. Results from granitic samples in the vicinity of Mezdreja and Gabrovica mines (ppm)

Sample	Type	Zr	Rb	W	Sn	Zn	Ba	Sb	Ni
Mzd granite mine	Soil	212	80	102	2214	59	22	70	63
Mzd granite mine	Test all geo	213	39	181	2125	57	84	98	42
Mzd granite	Soil	226	69	133	1577	46	25	<LOD*	44
Mzd granite	Test all geo	228	32	156	1678	42	102	65	44
MzdrKfeldGabrr	Soil	108	49	<LOD	16	46	3654	28	114
MzdrKfeldGabrr	Soil	35	71	<LOD	<LOD	21	5634	13	92
MzdSil lim	Soil	126	40	91	<LOD	55	36	<LOD	166
MzdSil lim	Test all geo	100	19	119	448	56	128	132	153
Mzd tail dump	Soil	141	99	39	953	35	27	29	35
Mzd tail dump	Test all geo	109	54	59	1017	40	81	58	<LOD
Gabr granite mine	Soil	131	127	139	1009	40	19	<LOD	29
Gabr granite mine	Test all geo	122	59	118	1169	38	113	39	26
Gabr tail dump	Soil	119	110	166	1066	40	144	<LOD	<LOD
Gabr tail dump	Test all geo	138	56	1567	1208	53	1501	47	44

*LOD – Limit of detection

Graph – Inovo Series, a highly graphitic sample (almost only graphite), friable material from schists; Inov Graph SiSchist – Inovo Series, graphitic schists with more silica (SiO₂); Inov Graph Schist – Inovo Series, graphitic schist with more graphitic material.

Tables 4 and 5 show results from red sandstone and gray siltstone in the Jelovica – Dojkinci area. Explanations of the sample abbreviations in the tables are: JelDojsandstred – Jelovica – Dojkinci area red sandstone; JelDojsandstLredt – Jelovica – Dojkinci area light red sandstone; JelDojsnastorange – Jelovica – Dojkinci area orange sandstone; JelGsilstRedox – Jelovica area gray siltstone from the redox zone;

JelGsilst – Jelovica area gray siltstone; JelGsilstmica – Jelovica area gray siltstone with mica.

A comparison with the average content in the Earth's crust was made according to available data [30-33]. Considering previous average values and repercussions for a particular element, the following values in ppm are taken: Zr – 130, Rb – 90, W – 0.7 and mafic, 1.2 and intermediates and 1.9 in acidic rocks, for Mo – 1.5, Sn – 2.3, Zn – 75, Cu – 55, As – 1.5, Sb – 0.5, Ba – 500, Ni – 80, Cr – 102, V – 80, and Ti – 5000.

It is important to note that Zr, Rb, W, Mo, and Sn originate and are mainly related to acid magmatic rocks (granites, riolites, etc.), as well as postmagmatic processes related to acid rocks. The Zn and Cu are

Table 2. Results from granitic samples Inovo for Zr, Rb, W, Sn (ppm)

Sample	Type	Zr	Rb	W	Sn
Inov graph	Soil	271	168	<LOD*	2242
Inov graph sischist	Soil	237	138	50	4759
Inov graph sischist	TestAll geo	235	69	<LOD	4990
Inov graph schist	Soil	174	95	107	4104
Inov graph schist	TestAll geo	159	47	76	4166

*LOD – Limit of detection

Table 3. Results from graphitic schist Inovo Zn, Ba, Sb, Ni, Mo, Cu, and As (ppm)

Sample	Type	Zn	Ba	Sb	Ni	Mo	Cu	As
Inov graph	Soil	207	467	38	154	94	108	54
Inov graph sischist	Soil	136	59	174	46	<LOD*	74	38
Inov graph sischist	Testall geo	142	178	274	59	4	64	47
Inov graph schist	Soil	62	52	122	29	11	21	53
Inov graph schist	Testall geo	55	123	150	44	6	19	46

*LOD – Limit of detection

Table 4. Results from red sandstones Jelovica-Dojkinci area (ppm)

Sample	Type	Zr	Rb	Sb	Sn	Zn	Ba	Cr	V	Ti	Ni
JelDojsand stred	Soil	131	89	11	6	20	686	22	20	1632	20
JelDojsand stred	TestAll geo	62	94	16	9	25	786	56	41	2204	48
JelDojsand stLredt	Soil	54	93	18	11	14	743	18	12	636	47
JelDojsnast orange	Soil	156	90	49	2482	34	30	<LOD*	11	<LOD	35
JelDojsnast orange	TestAll geo	150	45	49	2491	30	68	<LOD	<LOD	<LOD	<LOD

*LOD – Limit of detection

Table 5. Results from gray siltstones Jelovica-Dojkinci area (ppm)

Sample	Type	Zr	Rb	Mo	Zn	Cu	As	Ba	Cr	V	Ti	Ni
JelGsilst redox	TestAll geo	215	88	5	86	21	10	603	<LOD*	<LOD	4678	77
JelGsilst redox	Soil	217	179	4	86	17	9	749	76	174	4741	86
JelGsilst	TestAll geo	217	87	5	87	22	11	590	179	305	4662	97
JelGsilst	Soil	216	178	6	84	20	5	747	69	199	4713	90
JelGsilst mica	TestAll geo	251	85	3	80	23	11	164	<LOD	<LOD	<LOD	4
JelGsilst mica	Soil	275	174	5	91	28	10	69	<LOD	<LOD	<LOD	52

*LOD – Limit of detection

mainly bound to polymetallic medium-temperature hydrothermal deposits, in connection with medium-sized rocks. The Sb and Ba originate from low temperature hydrothermal processes associated with medium-sized magmatism. The Ni, Cr, V, and Ti groups originate from ultramafic and mafic rocks. The Ni is also concentrated in liquid segregates that arise in the first phases of differentiation of magma, as well as sulphide deposits, and Cr in crystallization differentiations – the first oxides that also occur in the initial phases of primary magma crystallization [34]. Therefore, the presence of these elements is quite expected in the tested samples. Elevated concentrations of zircon were measured in the samples of the Mezdreja granites, the graphitic schists of the Inovo Series and the gray siltstones of the Colorful Series. Additionally, they are present in red Early Triassic sandstones, but to a lesser extent. Since zircon is a rather resistant mineral, it is possible that it was transported to the siltstones during the course of formation of the Colorful Series and reached and retained its form in the metamorphic rocks of the Inovo Series.

The rubidium concentrations were generally lower than the abundance in the Earth's crust. However, elevated concentrations were recorded in the Gabrovnica granites, graphitic schists (batches rich in graphitic material), and gray siltstones (in the Soil mode). Tungsten concentrations were distinctly elevated (up to 100x the abundance in the Earth's crust), in all samples associated with the granites, except in a monomineral sample of pink K-feldspar where there was none. Elevated tungsten concentrations (ranging from 50 to 107 ppm) were also detected in the schists, but not in all of the samples. Molybdenum concentrations were low and nearly non-existent in the granite samples, except in clayey material from the Mezdreja tailing dump. The molybdenum was present in all the gray siltstone samples, from 3 to 6 ppm, which is two to four times greater than the abundance in the Earth's crust. Tin concentrations in the granites were up to 1000 higher than average (2214 ppm) and up to 4990 ppm in the graphitic schists.

The Zn was present in almost all samples, but generally below average abundance. Somewhat elevated concentrations were measured in the gray siltstones and graphitic schists, up to 2 times. The Cu in the granite samples was generally absent. It is found

in the gray siltstones and graphitic schists were slightly higher than average, and the highest concentration (108 ppm) was measured in the sample of schist rich in graphitic material.

In nature, arsenic and antimony are often associated in such cases. Significantly increased arsenic content was found in graphitic schists, up to 54 ppm and slightly less in gray siltstones, up to 11 ppm. All units exhibited elevated Sb concentrations; the highest was measured in the graphitic schists (up to 274.09 ppm). The highest barium concentration was detected in a monomineral fraction of the pink K-feldspar (5634 ppm), and elevated concentrations were found in Early Triassic sediments, both the red sandstones (up to 786 ppm) and the gray siltstones (up to 749 ppm).

The highest concentration of Ni was measured in granitic samples in the vicinity of Mezdreja and Gabrovnica mines (166 ppm) and somewhat lower in the graphite schist Inovo (154 ppm), which represents almost twice the higher concentration than the average for the Earth's crust. Titanium was found in the gray siltstones of the Jelovica-Dojkinci area (4741 ppm). Vanadium and chromium concentrations were highest in the Jelovica area gray siltstone (305, 179 ppm, respectively). The amount of vanadium in this sample is almost four times higher than expected.

Since all these localities from which the samples are examined in this study are located near watercourses, all the present elements, and in particular those with elevated concentrations, can be relatively easily transferred to the environment by the water action. Some of the investigated metals, such as copper, chromium, molybdenum, nickel, and zinc, are essential nutrients (so-called microelements) needed for various biochemical and physiological functions of organisms [35]. However, some of the detected metals or microelements in higher concentrations are toxic and pose a threat to human health and the environment [36, 37]. In addition, some of these elements such as ^{96}Zr , ^{87}Rb , ^{50}V , which represent the naturally occurring radioactive isotopes have radioactive isotopes [5, 38, 39]. Although, their radioactivity is not of significance in comparison with the main radioactivity carriers in this area, in case of their penetration into watercourses, groundwaters, and soil they might represent serious environmental pollutants. Likewise, there is the possibility of eolic erosion from the investigated

deposits and tailing dams whereby these chemical elements and/or radionuclides would be distributed to areas away from the primary sources of natural radioactivity. Regarding this, they could enter the food chain, which would also result in harmful effects on human health. Since this is a natural phenomenon, without the influence of anthropogenic factors (except for TENORM which was exposed during uranium ore exploitation), the concentration of these elements is difficult to reduce. Getting into the environment, all detected elements might contaminate food and/or drinking water, and accumulate in the body over time provoking harmful health effects [40]. This study indicated a need for adequate monitoring, and risk assessment of the examined locations, which could prevent the distribution of these metals in the environment.

CONCLUSION

The Zr, Rb, W, Mo, Sn, Zn, Cu, As, Sb, Ba, Ni, Cr, V, and Ti were detected as the most common accessory elements of the radioactivity carriers at the examined locations. Depending on the location, elevated concentrations of all detected elements except titanium were found. Increased concentrations of zirconium and rubidium were found in samples of granite, graphitic schists, and gray alevrolites. Tungsten was found in up to 100 times higher concentrations in all other granite samples. Molybdenum is present in all samples of gray alevrolite, 2 to 4 times. Tin was detected in granites up to 1000 times higher concentrations than the average. Although zinc is present in almost all samples, the measured values were generally below average except for gray alevrolites and graphitic schists (up to 2 times higher). Copper was found in gray alevrolites and graphitic schists slightly more than average values. The significantly increased content of arsenic was found in graphitic schists and somewhat less in gray alevrolites. Antimony had an elevated concentration at all locations, mostly in graphitic schists. Increased barium content was found in K-feldspar and sedimentary material. The highest content of Ni was found in granite and graphite schist samples, in almost double concentrations relative to the average values. Chromium and vanadium have the highest concentrations in the gray siltstones. Since the observed accessory elements of the representative petrologic radioactivity carriers are present in the soil mostly due to natural processes their existence is inevitable. However, as all localities contained increased concentrations of some of the investigated elements (except titanium), a high risk of their harmful effects on the environment exists and continuous monitoring is required.

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AUTHORS' CONTRIBUTIONS

The experiments were carried out by B. B. Vakanjac. All authors analyzed the results and participated in the preparation of the final version of the manuscript.

REFERENCES

- [1] Kumari, R., et al., Natural Radioactivity in Rock Samples of Aravali Hills in India, *International Journal of Radiation Research*, 15 (2017) 4, pp. 391-398
- [2] El-Taher, A., Makhul, S., Natural Radioactivity Levels in Phosphate Fertilizer and Its Environmental Implications in Assuit Governorate, Upper Egypt, *Indian J. Pure and Appl. Phys.*, 48 (2010), 10, pp. 697-702
- [3] Cochran, K., et al., *Encyclopedia of Ocean Sciences*, 3rd Ed., Academic Press, Elsevier, Amsterdam, The Netherlands, 2019, pp. 4306
- [4] ***, Evaluation of Guidelines for Exposures to Technologically Enhanced Naturally Occurring Radioactive Materials, National Research Council (US) Committee on Evaluation of EPA Guidelines for Exposure to Naturally Occurring Radioactive Materials, National Academies Press, Washington, 1999
- [5] Janković, M., Todorović, D. Concentrations of Natural Radionuclides in Imported Zirconium Minerals, *Nucl Technol Radiat*, 26 (2011) 2, pp. 110-114
- [6] Joshua, E. O., et al., Natural Radionuclides and Hazard of Rock Samples Collected from Southeastern Nigeria, *Radiation Measurements*, 44 (2009), 4, pp. 401-404
- [7] Sadiq, A. A., Agba, E. H. Background radia on in Akwanga, Nigeria, *Working and Living Environmental Protection*, 8 (2011), Dec., pp. 7-11
- [8] Kant, K., et al., Natural Radioactivity in Indian Vegeta on Samples, *International Journal of Radiation Research*, 13 (2015) 2, pp. 143-150
- [9] Kaur, M., et al., Seasonal Variation of Indoor and Outdoor Gamma Dose Rates of Reasi District of Jammu and Kashmir, *Nucl Technol Radiat*, 33 (2018) 1, pp. 106-111
- [10] Mehta, V., et al., Measurement of Radon, Thoron And Their Progeny In Indoor Environment Of Mohali, Punjab, Northern India, Using Pinhole Dosimeters, *Nucl Technol Radiat*, 31 (2016), 3, pp. 299-305
- [11] Myers, J. S., Geology of Granite, *Journal of the Royal Society of Western Australia*, 80 (1997), 3, pp. 87-100
- [12] Rogers, J. J. W., et al., Varieties of Granitic Uranium Deposits and Favorable Exploration Areas in the Eastern United States, *Economic Geology*, 73 (1978), 8, pp. 1539-1555
- [13] Rene, M., Dolniček, Z. Uraninite, Coffinite, and Brannerite from Shear-Zone Hosted Uranium Deposits of the Bohemian Massif (Central European Variscan Belt), *Minerals*, 7 (2017) 4, pp. 1-17
- [14] Grotzinger, J. P., Jordan, T. H., *Understanding Earth*, 7th Ed., W. H. Freeman, New York, USA, 2014, p. 752

- [15] Pellant, C., *Rocks and Minerals*, DK Publishing, London, 2010, pp. 256
- [16] Bills, D., et al., Geologic and Hydrologic Issues Related to Uranium Mining in the Grand Canyon region, *Proceedings*, northern Arizona, Annual Water Symposium "Managing Hydrologic Extremes", Arizona Hydrological Society, American Institute of Hydrology, Ariz., USA, 2009
- [17] Džoljić, J., et al., Natural And Artificial Radioactivity In Some Protected Areas of South East Europe, *Nucl Technol Radiat*, 32 (2017), 4, pp. 334-341
- [18] Gertik, S., *Uranium Deposits and Occurrences at Stara Planina Mountain* (in Serbian), Dedraplast, Belgrade, 2003, pp. 63-141
- [19] Kovačević, J., Metallogeny of Stara Planina Mt. Region (in Serbian), Ph. D. thesis, 2006, Faculty of Mine and Geology, Belgrade
- [20] Carlsson, E., Buchel, G., Screening of Residual Contamination at a Former Uranium Heap Leaching Site, Thuringia, Germany, *Geochemistry*, 65 (2005), Sept., pp. 75-95
- [21] Kipp, G., et al., Arsenic and Uranium Transport in Sediments Near Abandoned Uranium Mines in Harding County, South Dakota, *Applied Geochemistry*, 24 (2009) 12, pp. 2246-2255
- [22] Cuvier, A., et al., Trace Elements and Pb Isotopes in Soils and Sediments Impacted by Uranium Mining, *Science of The Total Environment*, 566-567 (2016), Oct., pp. 238-249
- [23] Rodriguez, J. H., et al., Biomonitoring of Atmospheric Trace Elements in Agricultural Areas and a Former Uranium Mine, *Biomonitoring*, 1 (2014), 1, pp. 63-74
- [24] Momčilović, M., et al., Population Doses from Terrestrial Exposure in the Vicinity of Abandoned Uranium Mines in Serbia, *Radiation Measurements*, 45 (2010) 2, pp. 225-230
- [25] Kovačević, J., et al., Natural Radioactive Elements in the Region of Closed Uranium Mines on Stara Planina, Eastern Serbia, in: *The New Uranium Mining Boom*, (Merkel B., Schipek M. eds), Springer Geology, Springer, Berlin, Heidelberg, 2011
- [26] Tanić, M., et al., Assessment of Radiation Exposure Around Abandoned Uranium Mining Area of Stara Planina Mt., Serbia, *Nucl Technol Radiat*, 29 (2014), 1, pp. 58-66
- [27] Abdulqader, S. M., et al., Characterization of the Main Naturally-Radioactive Geologic Units of Stara Planina, *Minerals*, 8 (2017), 6, pp. 1-17
- [28] ***, EPA, Technically Enhanced Naturally Occurring Radioactive Materials from Uranium Mining, Radiation Protection Division, Washington DC, USA, 2008
- [29] Arbuzov, S. I., et al., Geochemistry of Radioactive Elements (U, Th) in Coal and Peat of Northern Asia (Siberia, Russian Far East, Kazakhstan, and Mongolia), *International Journal of Coal Geology*, 86 (2011), 4, pp. 318-328
- [30] Mellor, J. W., Mellor's Modern Inorganic Chemistry Journal of Chemical Education, Longmans Green & Co LTD, USA, 1963
- [31] Wiberg, N., et al., Lehrbuch der Anorganischen Chemie, Gebundenes Buch, Berlin, 2007
- [32] Wedepohl, K. H., The Composition of the Continental Crust, *Geochimica et Cosmochimica Acta*, 59 (1995), 7, pp. 1217-1232
- [33] Kabata-Pendias, A., Pendias, H., *Trace Elements in Soils and Plants*, CRC Press LLC, Boca Raton, Fla., USA, 2001
- [34] Tomanec, R., Vakanjac, B., Mineral Paragenesis with Test Methods and Atlas of Characteristic Examples, Faculty of Applied Ecology, Belgrade, 2015
- [35] ***, World Health Organization, Trace Elements in Human Nutrition and Health, World Health Organization, Geneva, 1996, pp. 361
- [36] Nies, D., Silver, S., *Molecular Microbiology of Heavy Metals*, Springer, Berlin, 2007, p. 451
- [37] Markert, B., *Plants as Biomonitors: Indicators for Heavy Metals in the Terrestrial Environment*, Wiley-Blackwell, New York, USA, 1993, p. 645
- [38] ***, National Research Council, Evaluation of Guidelines for Exposures to Technologically Enhanced Naturally Occurring Radioactive Materials, The National Academies Press, Washington, 1999, p. 294
- [39] Heiskanen, H., et al., Theoretical half-life for beta decay of ⁹⁶Zr, *Journal of Physics G: Nuclear and Particle Physics*, 5 (2007), 1, pp. 837-845
- [40] Karatasli, M., Radionuclide and Heavy Metal Content in the Table Olive (*Olea Europaea* L.) From the Mediterranean Region of Turkey, *Nucl Technol Radiat*, 33 (2018), 4, pp. 386-394

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**ИСТРАЖИВАЊЕ ПРАТЕЋИХ ЕЛЕМЕНАТА РЕПРЕЗЕНТАТИВНИХ
ПЕТРОЛОШКИХ НОСИЛАЦА РАДИОАКТИВНОСТИ НА
СТАРОЈ ПЛАНИНИ, СРБИЈА**

Ова студија бави се присуством најчешћих пратећих елемената носилаца радиоактивности на Старој планини на четири дефинисане локације. Све истражене локације имају повећану радиоактивност, већу од 200 cps, све до 1250 cps. У свим испитиваним узорцима детектовани су следећи елементи и утврђене су њихове концентрације: Zr, Rb, W, Mo, Sn, Zn, Cu, As, Sb, Ba, Ni, Cr, V и Ti. Анализа је показала да су у зависности од узорка нађене повишене концентрације свих детектованих елемената осим титанијума. Будући да се све локације које садрже наведене елементе налазе у близини водотокова, сви присутни елементи, а посебно они са повишеним концентрацијама могу се релативно лако пренети у животну средину оцедним водама. Такође, постоји могућност и еолске ерозије са испитиваних лежишта и јаловишта, при чему би се ови хемијски елементи и радионуклиди дистрибуирали и у подручја удаљена од примарних извора природне радиоактивности. Ово представља висок ризик од њиховог ширења, а стога и штетне ефекте по животну средину. Студија је указала на потребу адекватног мониторинга и процене ризика на испитаним локацијама, што би могло спречити дистрибуцију ових елемената даље у животну средину.

Кључне речи: праћећи елементи, радиоактивност, токсичност, биолошки ризик, животна средина
