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ASSESSMENT OF ENVIRONMENTAL RADIOACTIVITY AND HEALTH HAZARD AT STARA PLANINA REGION

by

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The aim of the study was to evaluate the potential risks of radiation near abandoned uranium mines, tailing dumps, or uranium deposits on Mountain Stara Planina, Serbia. For risk assessment several parameters were determined: radium equivalent activity, Ra_{eq}, external hazard index, H_{ex}, gamma radiation absorbed dose rate, D, annual effective dose on background outdoor gamma exposure, $AED_{outdoor}$, and excess lifetime cancer risk, ELCR. Obtained results showed that all the samples, except one, have the Ra_{eq} value up to two times higher than the reference limit. The Ra_{eq} of the sample from the Mezdreja mine tailings was increased by almost eight times. The value of H_{ex} followed the same pattern as Ra_{eq}. All the investigated localities have increased D values, while all the samples have shown the moderately low $AED_{outdoor}$, except Mezdreja mine tailings that have 5.5-2.8 times higher dose relative to the world's average. ELCR at the Mezdreja mine tailings is 4.58 times higher than the world's average of $1.45 \cdot 10^{-3}$. In the context of human activity in the area of Stara Planina such as different kinds of tourism, livestock breeding, dairy products, and herbal manufacturing, *etc*. there is a need for detailed analysis in order to evaluate potential human exposure and health impacts.

Key words: natural radioactivity, environment, health, hazard, risk assessment

INTRODUCTION

Radionuclides are naturally present in the air, soil, and water, and living organisms as a result of natural radiation from cosmic and terrestrial sources. Human activities also generate radionuclides by preparation of nuclear weapons, their testing, uranium mining, nuclear fuel reprocessing, and nuclear accidents, *etc*. [1, 2]. The main external source of irradiation to the human body is represented by gamma radiation emitted by naturally occurring radioisotopes. Most of the terrestrial background radiation is due to non-chain primordial radionuclide potassium ^{40}K and chain primordial radioisotopes of the uranium (^{238}U to ^{206}Pb), thorium (^{232}Th to ^{209}Pb), and actinium series (^{235}U to ^{207}Pb) [1, 2]. These radionuclides were incorporated into the Earth at the time of its formation, and still are present because of their long half-lives and significantly affect terrestrial radiation levels [1]. The natural environmental radioactivity and the associated external exposure due to gamma radiation are in correlation with the geological and geographical characteristic and varies depending on the geological region. In addition, the existence of mining and quarrying increases the dose rate of background radiation in some regions that are known as high-level background radiation regions [1]. Since radionuclides are not uniformly distributed, the knowledge of their concentration and radiation levels in the environment, especially in particular localities, is important for assessing the effects of human exposure and potential health impacts.

The present study is the continuation of the systematic geological and radiological investigation of mountain Stara Planina in the Republic of Serbia [3, 4]. This mountain was known for its natural beauty, intact nature but also by natural radioactivity mainly due to uranium mineralization and exploitation [3, 5, 6]. Stara Planina was constituted a protected area and Nature Park in 1997 due to its high biodiversity of flora and fauna, and its special geological, hydrological, and ethno-cultural characteristics [7, 8]. In the late

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2000 s, this mountain became a popular tourist destination [5]. Although some of these destinations are localized away from the deposits and former mines, the hiking trails and picnic areas are positioned all over this mountain. Moreover, the mines and tailing dumps are abandoned and neglected, and it is easy to get to this area, even to highly contaminated places. Also, tourists often visit this area out of curiosity, ignoring that this is a restricted area due to inadequate security measures in past decades. Likewise, activities typical for this region, especially in the tourist season, *e. g.* dairy products or herbal manufacturing and sales, might transfer the contamination [9, 10].

The study deals with the effects of gamma radiation on Stara Planina and is not concerned with alpha and beta particles that affect only specific parts of the body. The aim of this study was to evaluate the potential risks of radiation near former uranium mines and tailing dumps, as well as of spreading the radioactive contamination. For this purpose following parameters were calculated: radium equivalent activity, Ra_{eq}, external hazard index, H_{ex}, gamma radiation absorbed dose rate, D, annual effective dose on background outdoor gamma exposure, $AED_{outdoor}$, and excess lifetime cancer risk, ELCR.

EXPERIMENTAL PART

Investigated area locations

The present research was particularly focused on background radiation risks at the locations of Stara Planina with increased radioactivity, greater than 200 cps (counts per second), from health and environmental viewpoints) [3].

A geographic information system (GIS) database has been established with a raster background that consists mainly of collected non-referenced and referenced maps, and vector materials including shape dots and polygon files of interest, *e. g.* sample points and areas, radioactivity in cps and mSv units. The environmental and health risk assessment was conducted at four locations, fig. 1 [3, 4].

The two locations represent uranium ore natural deposits and former mines, Gabrovnica and Mezdreja close by Janje village. Further, these areas, in the context of mining activities, which brought the radioactive carriers to the surface, could be considered as an anthropogenic source of radioactivity. Uranium mining generates so-called TENORM (technically enhanced naturally occurring radioactive material) [2], which is an undesirable process due to its direct influence on the atmosphere and potential of spreading through the ecosystem, especially by water action. The other two sampling locations are graphitic schists (so-called Inovo Series), located in the vicinity of the Gabrovnica deposit, and sedimentary materials represented by red sandstone and gray alevrolites of the

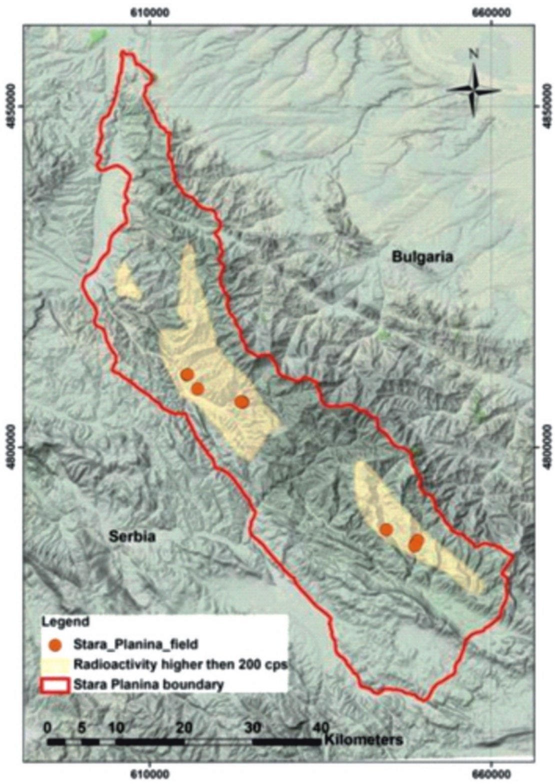

Figure 1. The boundary of Stara Planina in Serbia (full line), zones of radioactivity greater than 200 cps (clear), and points of observation and testing in 2016 and 2017 (dots); raster basis Google Landscape w/n, SASPlanet [3, 4]

so-called Colorful Series, both usually charged with radioactive and trace elements in the Jelovica-Dojkinci rivers region [11].

Listed localities have already been examined geologically and radiologically in previous studies [3]. The radiological investigation was limited to representative petrologic radioactivity carriers and their accessory elements without assessing the direct radiological hazard in the environment or impact on humans and other living beings.

The radioactivity of the terrain and observation points on the ground was measured by hand-held Gamma-Scout radiation detector (or Geiger counter), Gamma-Scout GmbH & Co.KG, Germany. The results were obtained in cps and μSvh^{-1}. In all locations, radioactivity was greatly elevated, *i. e.*, higher than the natural radiation level in this area. The counting rate of the gamma-rays was 70 cps [3]. Radioactivity in Gabrovnica was 240 cps at the mine portal, and 360 cps at the mine dump. Radioactivity in Mezdreja was 420 cps at the mine portal, and up to 1250 cps at the mine dump. Radioactivity at the sampling site in the Inovo Series area was 650 cps. Radioactivity in the Colorful Series area at the redox zone was 280 cps [3].

Radiological analyses

The specific activities, $Bqkg^{-1}$, of gamma emitters (^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs), in investigated characteristic petrologic samples from Stara Planina, were previously determined [3]. The specific activities of the samples were measured using a high-resolution coaxial semiconductor 63 detector with high-purity germanium crystal HPGe ORTEC GEM 50, with 50 % relative efficiency at 64 1332 keV [3]. These specific activities were used for further investigation in this study, and ten samples were investigated. Considering the investigation of natural radioactivity impact in this research, the specific activity of radioisotope ^{137}Cs was not considered in calculation due to its anthropogenic origin.

Radium equivalent activity

The Ra_{eq} is the most used radiation hazard index [12]. Radium equivalent activity has been widely used for the past 40 years for the assessment of gamma radiological hazards in the building materials, but it is also applied to identify the consistency of radiation exposure from soils. The term *radium equivalent activity* was introduced for the activity of a radionuclide, which would have the same therapeutic effect as 1 mg of ^{226}Ra. This index is convenient for the comparison of specific activities of samples/materials with different radionuclide content (above all ^{226}Ra, ^{232}Th, and ^{40}K), [12]. The unit of measure by SI is Bqkg^{-1}. Thus, radium equivalent activity represents an external exposure risk, and is based on [12]

$$Ra_{eq} \quad C_{Ra} \quad 1.43\ C_{Th} \quad 0.077\ C_{K} \tag{1}$$

where C_{Ra}, C_{Th}, and C_K represent particular isotopes specific activities ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in Bqkg^{-1}.

The equation is based on the value of 1 mSv per year as an annual exposure limit, assumed that based on the supposition that 370 Bqkg^{-1} of ^{226}Ra, 259 Bqkg^{-1} of ^{232}Th, and 4810 Bqkg^{-1} of ^{40}K all produce the same gamma-ray dose rate. Hence, factors in equation correspond to ratios: 370/259 = 1.43 and 370/4810 = 0.077. Therefore, Ra_{eq} is the weighted sum of activities of these three radionuclides. The radium equivalent is related to both the external gamma dose and the internal alpha dose from radon and its progeny from the decay of the radium resulting in respiratory tract doses to the residents of the building [12]. In order to keep the annual radiation dose below 1.5 mGy per year, the maximum Ra_{eq} value has to be less than 370 Bqkg^{-1}.

External hazard index

The H_{ex} is a broadly used hazard index that reflects external exposure to radiation. Index H_{ex} was defined by Beretka and Mathew [12] to evaluate the indoor radiation dose rate due to the external exposure to gamma radiation from the natural radionuclides in building materials of dwellings.

External hazard index can be calculated by [12]

$$H_{ex} \quad \frac{C_{Ra}}{370} \quad \frac{C_{Th}}{259} \quad \frac{C_K}{4810} \tag{2}$$

where C_{Ra}, C_{Th}, and C_K represent the certain isotope specific activities ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in Bqkg^{-1}.

The maximum value of H_{ex} equal to one corresponds to the upper limit of Ra_{eq} (370 Bqkg^{-1}) [12].

Gamma radiation absorbed dose rate

The *D* represents the amount of radiation delivered to some medium over a time period. It is a measure of ionizing radiation energy absorbed by some medium per time unit. It is equal to the energy deposited per unit mass of a medium per time unit, and has the unit joule [J] per kilogram [kg] per time unit, *i. e.* gray per second [Gys^{-1}], according to the SI system [13].

In the air from cosmic radiation at sea level, the value of *D* is approximately 30 nGyh^{-1} [13]. External exposures to this radiation arise from trace amounts of terrestrial radionuclides in all ground formations. Therefore, natural environmental radiation mainly depends on the geological and geographical characteristics of the area. For example, higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks [14].

The specific activities ^{226}Ra, ^{232}Th, and ^{40}K [Bqkg^{-1}] were used to calculate absorbed dose rate in nanograys per hour [nGyh^{-1}] at a height of 1 m above the ground using the following equation recommended by the UNSCEAR [13]

$$D \quad 0.462\ C_{Ra} \quad 0.604\ C_{Th} \quad 0.042\ C_{K} \tag{3}$$

where conversion factors from the equation are the dose coefficients in nGyh^{-1} per Bqkg^{-1} also recommended by UNSCEAR [13] and C_{Ra}, C_{Th}, and C_K represent the certain isotope specific activities ^{226}Ra, ^{232}Th, and ^{40}K, respectively, in Bqkg^{-1}. There is considered that the ^{226}Ra and ^{232}Th radionuclide decay products are in equilibrium, and the contribution of other natural and artificial radionuclides is negligible.

Annual effective dose

The absorbed dose rate is not good enough indicator of biological damaging. The risk of stochastic effects, due to radiation exposure for the population, can be quantified using the effective dose. The $AED_{outdoor}$ is based on the risks of radiation-induced health effects [15].

To estimate the annual effective doses conversion coefficient from absorbed dose rate in the air to effective dose and the outdoor occupancy factor must be taken into account. In the UNSCEAR report in 2000 [13] 0.7 SvGy^{-1} was used for the conversion coefficient from absorbed dose in the air to the effective dose received by adults. Coefficient 0.8 was used for the indoor occupancy factor, implying that an average of 20 % (0.2) of the time is spent outdoors around the world. The effective dose equivalent rate is calculated from the following formula [15]

$$AED_{\text{outdoor}} \quad D \; N_{\text{h}} \; 0.7 \; 0.2 \tag{4}$$

where N_h is the number of hours in one year (8760), and 0.7 and 0.2 are the aforementioned conversion coefficients.

The worldwide annual effective dose from natural gamma terrestrial radiation is estimated to be 0.5 (or range 0.3-0.6) mSv [13].

Excess lifetime cancer risk

Possible human activities in some areas may determine the need for the assessment of some specific risk factors for exposure to gamma radiation due to the presence of uranium mineralization. The most illustrative among radiological risk indices is ELCR, *i. e.* risk of cancer occurring during the expected lifetime. Excess lifetime cancer risk for the local population and visitors, mostly tourists, can be calculated according to annual effective doses by the following equation [16, 17]

$$ELCR \quad AED_{\text{outdoor}} \; LE \; RF \tag{5}$$

where $AED_{outdoor}$ is expressed in mSv unit, LE is average lifespan expectancy (70.1 years) and RF [Sv^{-1}] is a fatal risk factor per Sievert, which is 0.057 as per International Commission on Radiological Protection [15]. It is common to obtain the ELCR value displayed as ELCR 10^{-3}.

RESULTS AND DISCUSSION

The results, obtained by measuring the radioactivity of the samples, were used to calculate the following parameters of radiation risk: Ra_{eq} [$Bqkg^{-1}$] (tab. 1), H_{ex} (tab. 2), D [$nGyh^{-1}$] (tab. 3), $AED_{outdoor}$ [mSv per year] (tab. 3), and ELCR (tab. 4).

Explanations of the sample abbreviations in the tables are: Mezdr granite mine1 – sample of granite taken from outcrop at the entrance of Mezdreja mine; Mezdreja silicified lim1 – silificified and highly limonitized sample with carbonates, collected in situ from altered granite with veinlets at Mezdreja; Mezdr granite2 – sample of granite taken from outcrop at the Mezdreja area; Mzdreja clay tailings1 – crushed altered granite fragments with clay from Mezdreja tailing dump; Schist graphitic silicified1 – Inovo Series, graphitic schists with more silica (SiO_2); Schist graphitic2 – Inovo Series, graphitic schist with more graphitic material; Gabrovnica mine1 – granite taken from outcrop at the entrance of Gabrovnica mine; Gabrovnica tailings1 – granite sample taken form Gabrovnica mine tailing dump; Siltstone gray1 – Jelovica area gray siltstone; and Sandstone red2 – Jelovica – Dojkinci area red sandstone.

As previously mentioned, for keeping the annual radiation dose below 1.5 mGy per year, the maximum value of Ra_{eq} has to be less than 370 $Bqkg^{-1}$ [13]. Sandstones of the Colorful Series have a value less than the reference limit for Ra_{eq}, while other samples have an increased value up to two times, except in the case of clay materials with fragments of granite from the Mezdreja mine tailings, which value is increased by almost eight times. These values indicate a very high risk of gamma radiation at the investigated locations, especially in the area of Mezdreja mine tailings. (tab. 1) However, this value was expected due to formerly uranium exploitation and the formation of TENORM on the surface [2].

The external hazard index follows the same pattern as Ra_{eq} (tab. 2). The value of H_{ex} is lower than the reference value only in the case of the Colorful Series sandstone sample and slightly higher in sample gray siltstones of the Colorful Series and Gabrovnica mine with values of 1.08 and 1.14, respectively. Other samples have an increased H_{ex} value up to two and eight times in the case of Mezdreja mine tailings, which is in line with obtained Ra_{eq} values.

Determined Ra_{eq} and H_{ex} were found to be much higher than common values for the soil in the Republic of Serbia or neighboring countries [18, 19]. In these cases, some measures should be taken to ensure gamma radiation below the allowed levels [13].

All the investigated localities have increased gamma radiation absorbed dose rate (tab. 3). Sandstone of the Colorful Series sample has the lower absorbed dose, while the Mezdreja mine tailings, *i. e.* Mezdreja TENORM has the highest, even 23 times more than the world's average dose. The situation is particularly concerned since the majority of specimens come from in situ soil samples and represent the

Table 1. Specific activities of ^{226}Ra, ^{232}Th, and ^{40}K radionuclides in characteristic petrologic samples from Stara Planina [3]

Locality/sample	Specific activity, C [$Bqkg^{-1}$]		
	C_{Ra}	C_{Th}	C_K
Mezdreja granite mine1	142 7	250 10	1420 60
Mezdreja silicified lim1	400 20	188 9	600 30
Mezdreja granite2	116 5	230 10	1020 50
Mzdreja clay tailings1	2600 100	169 8	1240 60
Schist graphitic silicified1	220 10	141 7	1420 60
Schist graphitic2	380 20	169 8	900 40
Gabrovnica mine1	58 3	163 8	1700 80
Gabrovnica tailings1	206 9	250 10	1690 80
Siltstone gray1	102 5	97 5	2080 90
Sandstone red2	28 1	52 3	1270 60

Table 2. Radium equivalent activity and external hazard index

Locality/sample	Ra_{eq}, [Bqkg^{-1}]	H_{ex}
Mezdreja granite mine1	609 26	1.64 0.07
Mezdreja silicified lim1	715 35	1.93 0.09
Mezdreja granite2	523 23	1.41 0.06
Mzdreja clay tailings1	2937 116	7.93 0.31
Schist graphitic silicified1	531 25	1.43 0.07
Schist graphitic2	691 35	1.87 0.09
Gabrovnica mine1	422 21	1.14 0.05
Gabrovnica tailings1	694 29	1.87 0.08
Siltstone gray1	401 19	1.08 0.05
Sandstone red2	200 10	0.54 0.03
Reference value	370 [12]	1 [12]

Table 3. Gamma radiation absorbed dose rate and annual effective dose

Locality/sample	D [nGyh^{-1}]	$AED_{outdoor}$, [µSv]
Mezdreja granite mine1	276.24 11.79	339 14
Mezdreja silicified lim1	323.55 15.93	397 19
Mezdreja granite2	235.35 10.45	289 13
Mzdreja clay tailings1	1355.36 53.55	1662 66
Schist graphitic silicified1	246.44 11.37	302 14
Schist graphitic2	315.44 15.75	387 19
Gabrovnica mine1	196.65 9.58	241 12
Gabrovnica tailings1	317.15 13.56	389 17
Siltstone gray1	193.07 9.11	237 11
Sandstone red2	97.68 4.79	120 6
World's average	57 [13]	300-600 [13]

Naturally Occurring Radioactive Material (NORM) with high radioactivity [20].

Contrary to previous parameters all the samples have shown the moderately low annual effective dose of gamma radiation, excluding the Mezdreja mine tailings. This location has shown 5.5-2.8 times higher dose relative to the world's average, *i. e.* world range values. The primary radiological contaminant of concern is ^{226}Ra, which would contribute to the greatest risk from external exposure to the occasional recreationalist and tourists at the Stara Planina. Uranium may also be a contaminant of concern, especially if it can migrate to a drinking-water source where its chemical toxicity becomes a health hazard [21].

The calculated ELCR values in samples are shown in tab. 4. Excess lifetime cancer risk index at the Mezdreja mine tailings is 4.58 times higher than the world's average of $1.45\cdot10^{-3}$ [17]. However, other indices are not alarming since they are lower or slightly higher than the world's average value.

Obtained results indicate a need for further study which will evaluate potential risk for humans and possible transferred contamination [10]. Finally, it should be noted that the leachate from the Gabrovnica and Mezdreja mines, as well as the graphitic schists, flowing through the limestones, flush the material into the watercourses of the local rivers (Crnovrska, Inovo, and Gabrovnica) and further into Trgoviski Timok river. Therefore, the condition of the alluvion and watercourses should be also examined.

Table 4. Excess lifetime cancer risk

Locality/sample	ELCR (10^{-3})
Mezdreja granite mine1	1.35 0.05
Mezdreja silicified lim1	1.59 0.08
Mezdreja granite2	1.15 0.05
Mzdreja clay tailings1	6.64 0.26
Schist graphitic silicified1	1.21 0.06
Schist graphitic2	1.55 0.08
Gabrovnica mine1	0.96 0.05
Gabrovnica tailings1	1.55 0.07
Siltstone gray1	0.95 0.04
Sandstone red2	0.48 0.02
World's average	$1.45\cdot10^{-3}$ [17]

CONCLUSIONS

The present study evaluates the potential risks of radiation near former uranium mines, tailing dumps, or uranium deposits on Stara Planina. For this purpose following parameters were calculated: Ra_{eq}, H_{ex}, D, $AED_{outdoor}$, and ELCR. Obtained Ra_{eq} values indicate a very high risk of gamma-radiation at the investigated locations, especially in the area of Mezdreja mine tailings, while the external hazard index follows the same pattern as Ra_{eq}. Additionally, all the investigated localities have increased D. The situation is particularly concerned since the majority of specimens come from *in situ* soil samples, and the detected values represent the naturally occurring radioactivity. Contrary to previous parameters, all the samples have shown the moderately low $AED_{outdoor}$, excluding the Mezdreja mine tailings, which had 5.5-2.8 times higher dose relative to the world's average. The ELCR values in samples at the Mezdreja mine tailings are 4.58 times higher than the world's average of $1.45\cdot10^{-3}$. However, other indices are not alarming since they are lower or slightly higher than the worldwide average value. In the context of human activity in the area of Stara Planina, such as livestock breeding, dairy products, and herbal manufacturing, there is a need for detailed analysis in order to evaluate potential risk for the living beings and the environment.

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AUTHORS' CONTRIBUTIONS

The results processing and calculations were done by S. D. Dimović, I. V. Jelić, and M. G. Rikalović. The field experiments were carried out by B. B. Vakanjac and V. R. Ristić Vakanjac. All the authors analyzed the results and participated in the preparation of the final version of the manuscript.

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ПРОЦЕНА РАДИОАКТИВНОСТИ У ЖИВОТНОЈ СРЕДИНИ И ОПАСНОСТИ ПО ЗДРАВЉЕ НА ПОДРУЧЈУ СТАРЕ ПЛАНИНЕ

Процењени су потенцијални ризици од зрачења у близини напуштених рудника уранијума, јаловишта, или лежишта уранијума на Старој планини у Србији. У сврху процене ризика одређено је неколико параметара: радијум еквивалент, Ra_{eq}, екстерни хазардни индекс, H_{ex} апсорбована доза, D, годишња ефективна доза спољашњег зрачења, $AED_{outdoor}$ и ризик од појаве канцера током очекиваног животног века, *ELCR*. Добијени резултати показали су да сви узорци осим једног имају вредност Ra_{eq} и до два пута већу од референтне вредности. Вредност Ra_{eq} узорка из јаловишта рудника Мездреја повећан је за готово осам пута. Вредност H_{ex} следила је исти тренд као и Ra_{eq}. Сви истражени локалитети имају повећану вредност D, док су сви узорци показали умерено низак екстерни *AED*, осим јаловине рудника Мездреја која има 5.5-2.8 пута већу вредност у односу на светски просек. Вредност *ELCR* јаловишта рудника Мездреја 4.58 пута је већа од светског просека који износи 1.45 10^{-3}. Рад упућује на потребу за детаљном анализом како би се проценила потенцијална изложеност и утицаји на здравље у контексту људских активности на подручју Старе планине као што су, на пример, различите гране туризма, сточарство, производња млечних или биљних производа.

Кључне речи: природна радиоактивност, животна средина, здравље, хазард, процена ризика