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Original scientific paper

RISK ASSESSMENT RESULTING FROM RADIONUCLIDES IN SOILS OF THE REPUBLIC OF MACEDONIA[#]

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In the last decade, many studies have been made for measuring natural radioactivity in regions of the Republic of Macedonia. However, the information on terrestrial radiation exposure, and, consequently, risk assessment were not available. In this study, a risk assessment was done considering a specific activity concentration of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the topsoil. The results indicate that the external gamma doses due to natural radionuclides in soils are higher than those of ^{137}Cs. The absorbed dose rate in air varies in intervals: from 4.3 to 57 nGy/h (due to ^{226}Ra); from 3.9 to 88 nGy/h (due to ^{232}Th); from 3.3 to 58 nGy/h (due to ^{40}K); and from 0.01 to 5.3 nGy/h (due to ^{137}Cs). In addition, the mean annual effective doses due to natural radionuclides and ^{137}Cs in the soils of the Republic of Macedonia are estimated to be 78 μSv and 1.01 μSv, respectively. As well as, the values of external hazard index H_{ex} indicate in general low gamma radiation risk for populations living in the Republic of Macedonia.

Key words: outdoor; gamma dose; external hazard index

INTRODUCTION

The study of different radioactive sources in the environment has led to extensive surveys in many countries. Cosmic and terrestrial radiation as well as the associated public exposure due to it, depend primarily on the geological and geographical conditions and seem at completely different levels of every region within the world [1, 2].

Public exposure due to cosmic radiation varies with altitude and solar activity while exposure from the terrestrial materials depends on its geological origin. Terrestrial radioactivity arises mainly from natural radionuclides, like ^{40}K, also the radionuclides from ^{232}Th- and ^{238}U-chains [2]. These radionuclides, which are formed as a result of the primary processes during the formation of the Earth, are named "primordial radionuclides". Because of their very long half-lives, they still exist within the Earth crust up to nowadays. In addition to primordial radionuclides in the soil, the artificial ^{137}Cs occurs mostly as a result of the Nuclear weapon tests and Chernobyl reactor accident [3] within the last century. The radionuclides presence in the Earth surface leads to external exposure of the people, proportionally to their concentration in soil. The natural radionuclides concentrations in soil depend on the radioactivity of the rock from which the soil is formed. Furthermore, many processes, such as sorption, deposition, and washout of radionuclides upon the influence of natural waters in the soil, caused changes in the radionuclides concentrations.

[#]Dedicated to academician Gjorgji Filipovski on the occasion of his 100th birthday

In specific, radon decay products and gamma radiation emitted by terrestrial natural radionuclides arise in the soil and in building materials are the main sources of the internal and external exposures of the peoples. Studying of the radionuclides content in the soils provides essential radiological information about the background level and the possibility for population risk assessment considering its spatial variation.

The quantities commonly used for population risk assessment due to external exposure on terrestrial radioactivity are external absorbed dose rate in air at 1 m above the ground level and the annual effective dose. In many studies, for the population radiological risk evaluation due to natural radionuclides in the soil the external hazard index is engaged as well [4–6]. Even, it has been developed for building materials [7], its application for soils was widely used because of the proportionality between the outdoor, and the indoor gamma doses.

Concerning the great benefits from the studies of radioactivity in the environment, extensive surveys in the Republic of Macedonia were conducted within the last decade. Beside several investigations of radon and thoron [8–18] and radioactivity in some types of cement [19] investigations of the radionuclides in the surface soils across the country were conducted as well [20–23]. In this work, estimation of the external radiation risk was done using the results of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs specific activities measured in the 213 soil samples, sampled over the entire territory of the country.

MATERIAL AND METHODS

The Republic of Macedonia is located within the central part of the Balkan Peninsula, occupying an area of 25 713 km^2. The area is characterized by diverse relief and complex geology. Over 50 % of the total area is mountain massive. The territory is organized into 8 statistical regions: Polog (POL); Southwest (SW); Pelagonija (PEL); Skopje (SKO); Vardar (VAR); Southeast (SE); East (EAST); Northeast (NE).

Soil samples and measurements

The topsoil (0–20cm) was sampled throughout the period of 2007–2010 from 213 locations. The sampling locations were on the uncultivated fields in/or close to inhabited areas. Their geographical positions across the eight statistical regions of the country are shown in Figure 1. In the laboratory, the samples were crushed, dried at 105 °C, sieved and then transferred in 500 ml Marinelli beakers. Furthermore, each sealed Marinelli was kept aside for about one month to ensure secular equilibrium between ^{226}Ra and its decay products prior to gamma spectrometry measurements.

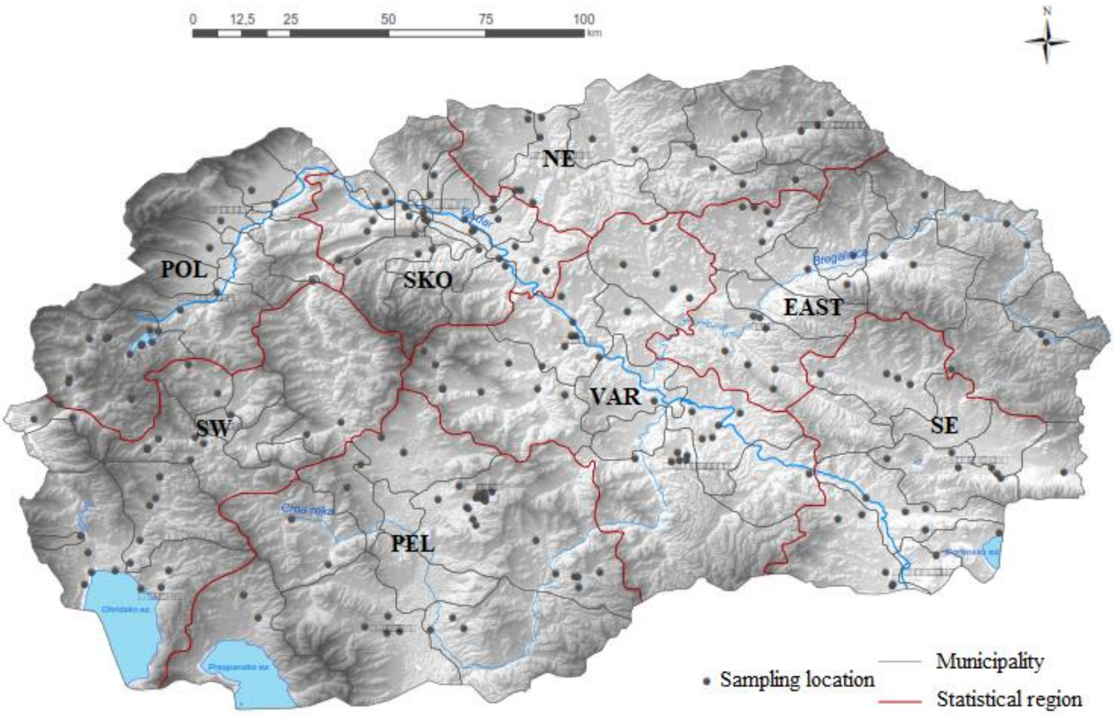

Figure 1. Soil sampling locations across the territory of the Republic of Macedonia

The measurements were carried out with a p-type HPGe detector (Canberra Inc.; 25 % relative efficiency, resolution of 1.79 keV at 1.33 MeV, 8192 ch. digital analyzer), and with software GENIE 2000 system for spectrum evaluation. The methodology of measurements is already explained in our previous published paper [20].

External effective dose due to natural and artificial radioactivity in soil

The external gamma dose rate in the air at 1 m above ground level was determined from measured specific activities considering factors of 0.462, 0.604, 0.042 and 0.030 for converting the activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs to absorbed dose [1–3], as given below:

$$D = 0.462A_{Ra} + 0.604A_{Th} + 0.042A_{K} + 0.030A_{Cs} \quad (1)$$

Where D is the dose rate in nGy/h and A_{Ra}, A_{Th}, A_K and A_{Cs} are the specific activities in Bq/kg of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs, respectively.

Furthermore using the estimated absorbed dose rate D, the annual effective dose is determined by [1, 2]:

$$D_E = D \cdot 0.7 \cdot 8760 \cdot 0.2 \quad (2)$$

where to determine the biological hazard to which a person is exposed, Gy is converted to Sv considering the conversion factor of 0.7, and the occupancy factor of 0.2 specifies the proportion of the total time (8760 h/y) spent outdoors.

External hazard index due to natural radioactivity in soil

The external hazard index is a useful coefficient for comparing the activities of materials that contain ^{226}Ra, ^{232}Th, ^{40}K considering the radiation effect associated with them. The external hazard index [7] is defined as follows:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (3)$$

where, A_{Ra}, A_{Th}, A_K and A_{Cs} are the specific activities (Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. If $H_{ex}<1$, the radiation hazard is insignificant that corresponds to the dose limit of 1 mSv for the population [24].

RESULTS

Descriptive statistics of the total gamma dose rates in the air at 1 m above ground level as well separately due to ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in soil collected at 213 different locations across the entire territory of the Republic of Macedonia is presented in Table 1.

Table 1. Descriptive statistic of the gamma dose rates in the air at 1 m above ground level from 213 different locations across the whole country territory

Statistic	^{40}K		^{226}Ra		^{232}Th		^{137}Cs		D(tot) nGy/h
	D nGy/h	%	D nGy/h	%	D nGy/h	%	D nGy/h	%	
Min.	3.34	14	4.34	17	3.93	18	0.01	0	13
Max.	57.96	58	57.03	76	87.73	84	5.34	7.61	176
AM	24.41	36	18.76	27	24.91	36	1.15	1.80	69
SD	7.99	7	8.26	6	10.88	7	0.93	1.46	24
GM	22.92	35	17.17	26	22.78	35	0.82	1.27	65
GSD	1.47	1.24	1.53	1.22	1.55	1.19	2.58	2.62	1.43

The values of the gamma doses rates due to both natural and artificial isotopes D(tot) varies between 13 nGy/h and 176 nGy/h with geometric mean (GM) of 65 nGy/h. In general, higher mean gamma doses rates that originate from the natural radionuclides were found, compared to doses from the artificial ^{137}Cs. On the other hand, the gamma doses rate due to exposure to ^{232}Th and ^{40}K are higher compared to doses from ^{226}Ra (Kruskal-Wallis test, error probability $p < 0.0001$). The dispersions of the results, expressed through the standard deviation and the geometric standard deviation, are smaller for natural radionuclides than for the ^{137}Cs. All data fitted well with log-normal distribution (Kolmogorov-Smirnov test, $p > 0.05$).

Descriptive statistics of the gamma dose rate due to natural radionuclides and ^{137}Cs in the soil, sampled in the 8 statistical regions of the Republic of Macedonia is given in Table 2.

Table 2. Descriptive statistic of external gamma dose rate due to Natural and artificial radionuclides in the soils of the 8 statistical regions of the Republic of Macedonia.

		Gamma dose rate (*D*) in the air at 1 m above ground							
	Statistical regions	SKO	VAR	SE	EAST	NE	PEL	SW	POL
Natural radionuclides	No. of observations	33	31	20	24	14	38	34	19
	Minimum (nGy/h)	32	39	13	43	36	56	51	38
	Maximum(nGy/h)	71	134	138	108	108	175	103	79
	AM (nGy/h)	50	70	57	69	63	86	75	61
	SD (nGy/h)	11	25	33	18	22	22	16	13
	GM (nGy/h)	48	66	48	67	60	84	73	59
	GSD	1.27	1.39	1.87	1.29	1.39	1.26	1.25	1.25
Artifitial radionuclides	Minimum (nGy/h)	0.15	0.20	0.07	0.15	0.01	0.01	0.36	0.03
	Maximum (nGy/h)	2.68	3.75	1.98	3.19	4.11	5.19	5.34	4.50
	AM (nGy/h)	1.08	1.08	0.73	1.15	0.85	1.15	1.46	1.56
	SD (nGy/h)	0.69	0.89	0.51	0.81	1.07	0.88	1.05	1.36
	GM (nGy/h)	0.83	0.81	0.57	0.87	0.43	0.86	1.19	0.87
	GSD	2.26	2.12	2.17	2.28	4.37	2.70	1.86	4.01

The geometric mean (GM) values of the gamma dose rate due to natural radionuclides and artificial (^{137}Cs) in soils of the statistical regions were in intervals from 48 nGy/h to 84 nGy/h and from 0.43 nGy/h to 1.19 nGy/h, respectively. Geometric standard deviations (GSD) of the doses were in intervals from 1.25 to 1.87 and from 1.86 to 4.37 for natural and artificial radionuclides, respectively. In all regions, the contribution of ^{226}Ra to the dose is lower compared to those of ^{232}Th and ^{40}K (Figure 2).

The descriptive statistic and interpolated maps of the annual effective doses estimated due to the natural radionuclides and ^{137}Cs activities in the soil are shown in Table 3 and Figure 3.

In the present work, the annual effective dose due to the natural radionuclides ranges between 15 μSv and 215 μSv with the geometric mean value of 78 μSv; the annual effective dose due to the ^{137}Cs ranges between 0.01 μSv and 6.55 μSv with the geometric mean value of 1.01 μSv (Table 3).

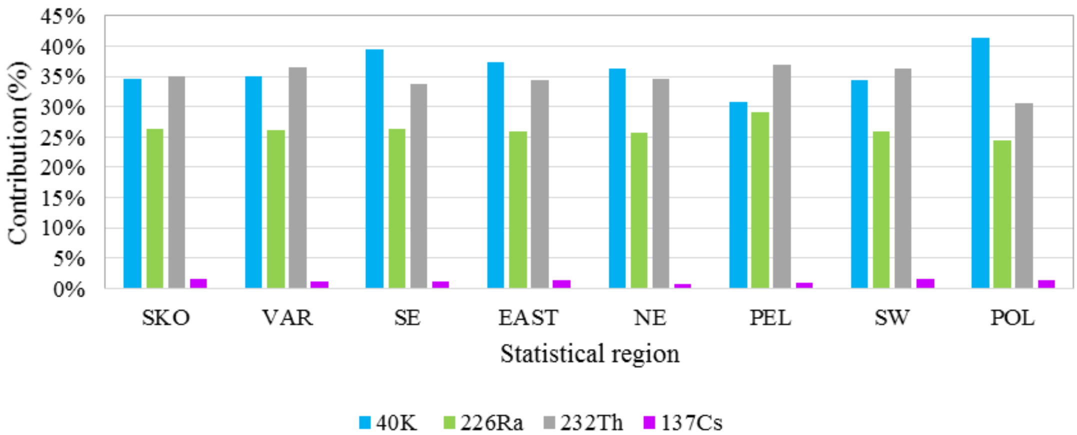

Figure 2. Different contributions of the ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs into total absorbed dose rate through 8 statistical regions

Table 3. Descriptive statistic of the annual effective doses due to Natural and artificial radionuclides in soil

	Annual effective dose D_E	
	Natural radionuclides	^{137}Cs
No. of observations	213	213
Minimum (μSv/y)	15	0.01
Maximum (μSv/y)	215	6.55
Arithmetic mean (μSv/y)	83	1.42
Standard deviation (μSv/y)	29	1.14
Geometric mean (μSv/y)	78	1.01
Geometric standard deviation	1.44	2.58

a.)

b.)

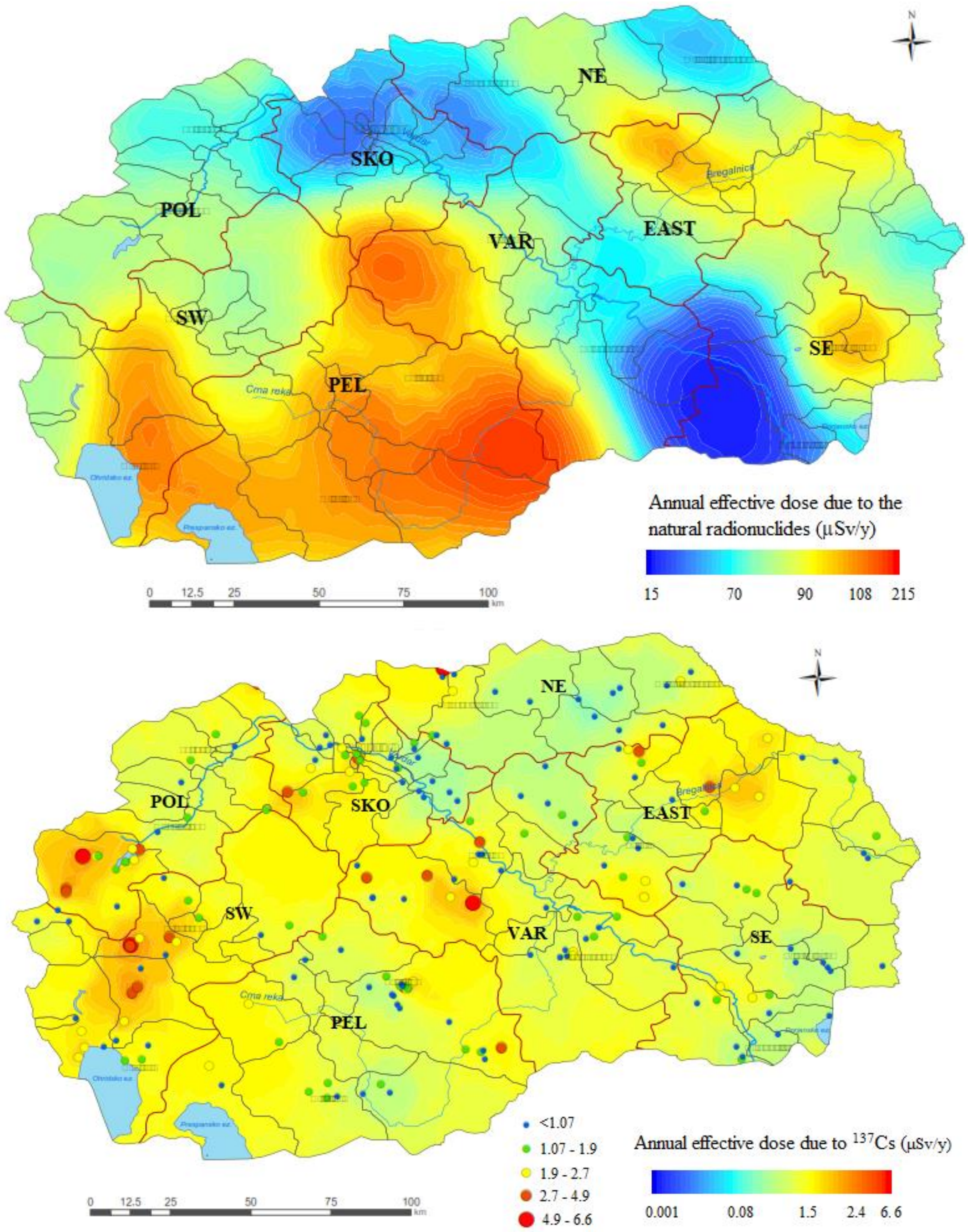

Figure 3. Interpolated maps of the annual effective doses due to the natural radionuclides (a) and ^{137}Cs (b) specific activities in the soil

As can be seen from Table 3 and Figure 3, the annual effective dose received due to the exposure to natural radionuclides is much higher than the annual effective dose due to ^{137}Cs, i.e., the contribution of ^{137}Cs in the total external exposure of the population is negligible.

On the map in Figure 4, the values of the external hazard index for each sampling location are shown. The estimated hazard index values (H_{ex}) ranged from 0.07 to 1.05. The value of the external hazard index exceeded 1 only at one location.

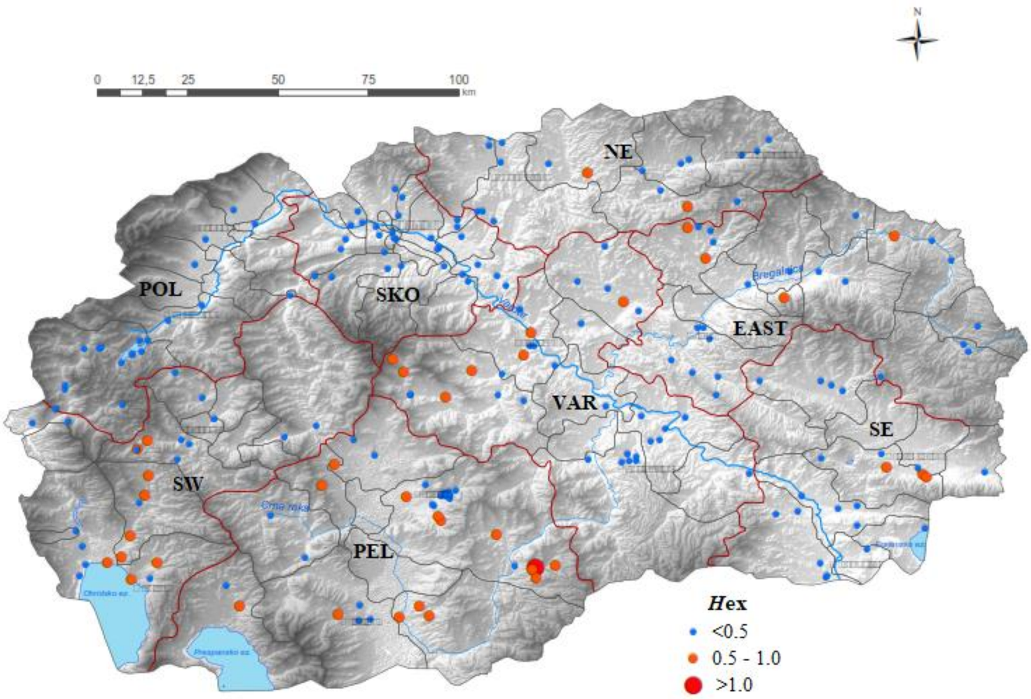

Figure 4. External hazard index (H_{ex}) values through the territory of the Republic of Macedonia

DISCUSSION

External exposure due to gamma radiation emitted by ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs in the soils appear at different levels across to the country territory. In general, the contribution of natural radionuclides (^{226}Ra, ^{232}Th, ^{40}K) and ^{137}Cs to the total gamma dose rate are different (Table 1). Specifically, the mean contributions of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs into the total gamma dose rate are 26%, 35%, 35% and 1.27% respectively.

The results show a negligible contribution of the ^{137}Cs in the total external gamma exposure of the population in the Republic of Macedonia (Table 2 and Table 3). As mention previously, the ^{137}Cs in soils are different in origin: nuclear weapon tests and Chernobyl accident. Considering that no published results related to ^{137}Cs activity in the country before the Chernobyl accident makes the evaluation of the origin's relevant contributions demanding. Overall, people living in the western part of the country receive a higher dose compared to those living in the east (Figure 3b). It could be a result of the complex dispersion pattern of cesium in the environment but also linked to higher levels of precipitation in the western part of the country.

Otherwise, the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in the soils are associated to the formation of each lithological area, particularly to its content in the rock from which the soils originate [20]. Commonly, the geology of the Republic of Macedonia comprises a variety of complex of sedimentary, metamorphic and volcanic rocks with different age and mineral content. The highest gamma dose rate was related to the soil sample from Mariovo that belongs to the Pelagonia region (Figure 3a). Higher gamma dose rates are found in South West, as well, compared to other statistical regions (Table 2). The South West statistical region appertains into the Western Macedonian geotectonic zone while as the Pelagonija statistical region belongs to the Western Macedonian zone and Pelagonian Massif. Both geotectonic zones comprised rocks of volcanic origin [25]. The contribution of the natural radionuclides into doses varied within regions as well as. For example, maximum contributions due to such radionuclide into a total absorbed dose were found to be in PEL (29%) due to ^{226}Ra; VAR (37%) and PEL (37%) due to ^{232}Th and POL (41%) due to ^{40}K.

The mean gamma dose rate for the whole country of 65 nGy/h was similar to that of 62.8 nGy/h reported for the neighboring country Serbia [26] but higher than the dose of 54 nGy/h obtained from survey in Kosovo and Metohija [27] and dose of 46.2 nGy/h assessed for population in the Thessaloniki city in Greece [28].

The mean annual effective dose value of 78 μSv/y due to natural radionuclides in the soils of the Republic of Macedonia is slightly higher than the worldwide value of 70 μSv/y reported in UNSCEAR reports [1–3].

CONCLUSION

The gamma exposure of the population due to ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs contained in the soils of the Republic of Macedonia have been analyzed. The gamma doses emitted from ^{137}Cs was negligible compared to the dose originated from natural radionuclides. The contribution of the natural ^{226}Ra, ^{232}Th and ^{40}K in the total dose of the population in the Republic of Macedonia were 26%, 35% and 35% respectively. In general, in the regions where bedrocks are from the volcanic origin the exposure due to ^{226}Ra, ^{232}Th, ^{40}K in soil is higher in comparison with other parts of the country. The obtained results are comparable with the results reported in the literature from neighboring countries.

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ПРОЦЕНКА НА РИЗИКОТ ОД РАДИОНУКЛИДИТЕ ВО ПОЧИТЕ НА РЕПУБЛИКА МАКЕДОНИЈА

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Во последната деценија се направени многу студии за мерење природна радиоактивност во региони на Република Македонија. Сепак, информациите за експозицијата на терестријалните зрачења и следствено процената на ризикот од нив не беа достапни. Во оваа студија, процената на ризикот беше направена врз основа на специфичните активности на ^{226}Ra, ^{232}Th, ^{40}K и ^{137}Cs во површинската почва. Резултатите укажуваат на тоа дека надворешните гама-дози што потекнуваат од природните радионуклиди во почвите се повисоки во однос на оние од ^{137}Cs. Брзината на апсорбираната доза во воздухот варира во интервали: од 4,3 до 57 nGy / h (од ^{226}Ra); од 3,9 до 88 nGy / h (од ^{232}Th); од 3,3 до 58 nGy / h (од ^{40}K); од 0.01 до 5.3 nGy / h (од ^{137}Cs). Проценетите средни годишни ефективни дози што потекнуваат од природните радионуклиди и ^{137}Cs во почвите на Република Македонија се 78 μSv и 1.01 μSv, соодветно. Вредностите на индексот на надворешна опасност Hex општо укажуваат на низок ризик од гама-зрачење за населението кое живее во Република Македонија

Клучни зборови: надворешна гама доза; индекс на надворешна опасност