BioRisk 17: 45–57 (2022)
doi: 10.3897/biorisk.17.77432
https://biorisk.pensoft.net

RESEARCH ARTICLE

A peer-reviewed open-access journal
BioRisk

Radiation status of soils from the region of the Eastern Rhodopes (Southern Bulgaria)

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Academic editor: Michaela Beltcheva | Received 1 November 2021 | Accepted 8 December 2021 | Published 21 April 2022

Citation: Hristozova M, Lazarova R (2022) Radiation status of soils from the region of the Eastern Rhodopes (Southern Bulgaria). In: Chankova S, Peneva V, Metcheva R, Beltcheva M, Vassilev K, Radeva G, Danova K (Eds) Current trends of ecology. BioRisk 17: 45–57. https://doi.org/10.3897/biorisk.17.77432

Abstract
Local values of natural radiation background in soils from unexplored regions in the Eastern Rhodopes were established. The impact of anthropogenic activity as a potential risk for increase in radiation background was assessed. Soil samples from areas near the liquidated lead-zinc mines – Madzharovo, gold mine – Ada Tepe, Krumovgrad, lead-zinc complex – Kardzhali, Neochim – Dimitrovgrad, deposits for extraction of gneiss, marble quarries, etc. were analyzed to study possible contamination. Specific activity of natural radionuclides ^{210}Pb, ^{238}U, ^{226}Ra, ^{235}U, ^{232}Th, ^{40}K and technogenic ^{137}Cs in the studied samples was determined by gamma spectrometric analysis with Multichannel analyzer DSA 1000, production of CANBERRA and HPGe-detector.

Keywords
Anthropogenic activity, natural and technogenic radionuclides, radiation monitoring of soils, radiation pollution, radioecology

Introduction

The Rhodopes are the largest mountain range in our country. The relief of the Eastern Rhodopes is mainly lowland and hilly with an average altitude of about 300 m. The main rocks are sedimentary and volcanic (andesites, rhyolites, tuffs, etc.), as the Eastern Rhodopes were occupied by a water basin with active underwater volcanism in the past. Deluvial and cinnamon soils are the most widespread. The soil-forming rocks are mainly granites, marble, gneiss and shale characterized by relatively high content of uranium and other natural radionuclides. The extraction of heavy and rare metals in the area, as well the production of some mineral fertilizers, carries the potential risk of further pollution of the environment with natural radionuclides.

The aim of the research was to study undisturbed soils, i.e. soils unaffected by industrial activity from areas in close proximity to industrial sites to assess the impact of anthropogenic activity on potential increase in radiation background. A large region was covered to collect initial data on the radiation status of the soils and for planning further studies in areas where high content of natural radionuclides was found.

The aim of the study was also to register the soil status in the region in terms of technogenic pollutant cesium-137.

A comparison was made with the radiation status of soils from other regions of Bulgaria.

Methods

Soil samples from representative points close to anthropogenically affected areas were collected and analyzed. Four expeditions were carried out to collect soil samples from areas near the liquidated Madzharovo lead-zinc mines, Ada Tepe gold mine, Krumovgrad, Kardzhali mining complex, Neochim, Dimitrovgrad, gneiss mining sites and marble quarries (Figures 1 and 2). Five samples were collected for each point according to BSS 17.4.5.01:1985.

Collection and preparation of samples were performed following ISO 18589–2,3 (2007) Sampling from 0 to 5 cm depth was carried out to monitor surface contamination and up to 20 cm to characterize the process in depth. Soil samples were air-dried, homogenized and ground and sieved through a 2 mm sieve. The Marinelli samples containers of 0,5 l volume and geometry 4π were used for performing gamma spectrometric analyses. The statistical reliability of the gamma analysis result is achieved through the duration of the measurements. The samples were measured from 19 to 24 hours.

The specific activity of natural radionuclides ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K and technogenic ^{137}Cs in soil samples was determined by gamma spectrometric analysis following ISO 18589-3. A DSA 1000 Multi-Channel Analyzer, CANBERRA, with ultra-pure germanium detector, with 35% efficiency and 1.8 keV resolution was used allowing simultaneous and direct measurement of a large number of gamma emitters with energies from 50 to 2000 keV. ^{238}U was measured by the daughter product ^{234}Th (63.3 keV and 92.3 keV). ^{226}Ra was determined by the maximum energy peak at 186.3 keV, with correction for ^{235}U (185.6 keV), ^{210}Pb – by the gamma line at 46.6 keV. ^{232} Th was determined by the daughter product ^{228}Ac (911.0 keV)

Figure 1. Eastern Rhodopes – map of the studied area.

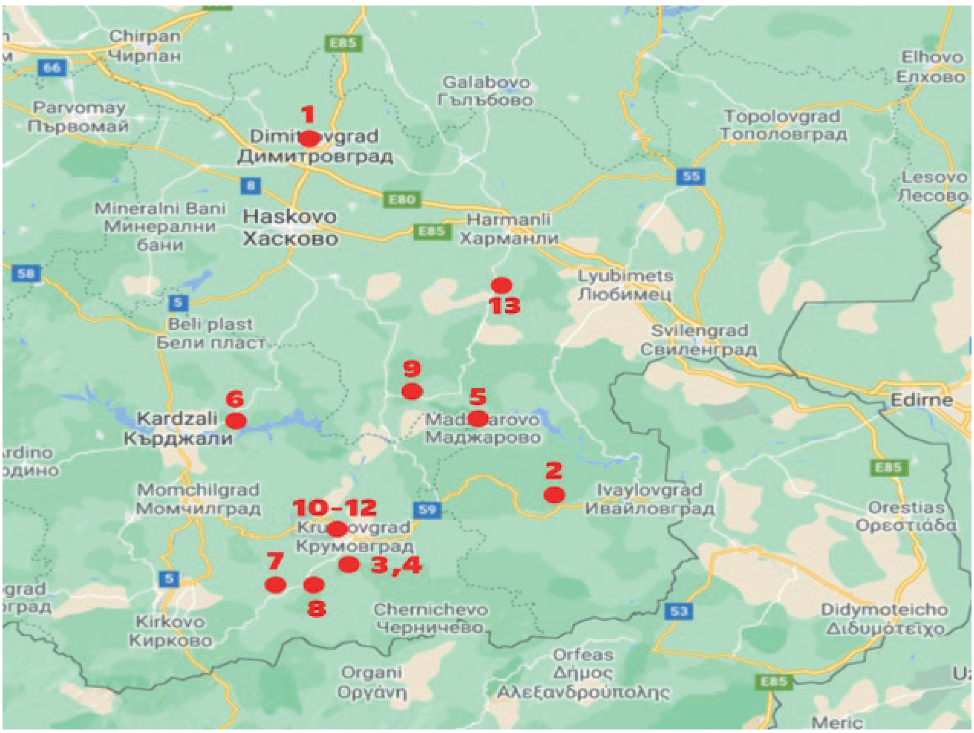

Figure 2. Ada Tepe gold mine.

and ^{40}K – by the 1461 keV full energy peak. Technogenic ^{137}Cs was measured by the gamma line at 661.6 keV.

To assess the radiation risk to the population in the studied areas radium equivalent activity (Ra_{eq}) and external hazard index (H_{ex}) were calculated by the formulas given below.

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K, \text{ (Beretka and Mathew 1985)},$$

where A_{Ra}, A_{Th} and A_K are the activities in Bq/kg of ^{226}Ra, ^{232}Th and ^{40}K respectively. The limit value is 370 Bq/kg.

$$H_{ex} = A_U / 370 + A_{Th} / 259 + A_K / 4810 \leq 1, \text{ (Krieger 1981)},$$

where A_U, A_{Th} and A_K are activities in Bq/kg of ^{238}U, ^{232}Th and ^{40}K, respectively.

Results

The data of the content of ^{40}K, ^{210}Pb, 235U, ^{238}U, ^{232}Th and ^{226}Ra, as well as technogenic ^{137}Cs in the soil samples from the studied region of the Eastern Rhodopes are presented in Table 1 in Bq/kg dry weight and combined standard uncertainty. The table indicates the anthropogenically affected areas near where the soil samples have been taken.

The results are discussed in the context of possible increase in background radiation exposure due to continuing human activity in the areas.

The radiation hazard was assessed by calculating the radium equivalent activity (maximum permissible level – 370 Bg/kg) and the external hazard index presented (maximum permissible level – 1) in Figures 3 and 4.

Table 1. Content of radionuclides in soils from the region of the Eastern Rhodopes in Bq/kg dry weight.

No	Sampling	Depth cm	^{210}Pb	^{137}Cs	^{40}K	^{238}U	^{226}Ra	^{235}U	^{232}Th
1	Neochim, Dimitrovgrad	0–5	34±5	28±1	670±10	41±4	30±3	1.7±0.5	38±3
2	Quarry, village of Cherni rid	0–5	55±6	< 1	850±20	50±5	73±8	2.5±0.5	47±4
3	Ada Tepe mine, Krumovgrad, bypass road	0–5	60±5	26±1	360±10	18±3	21±4	1±0.5	25±2
4	Ada Tepe mine, Krumovgrad	0–5	-	-	1000±10	9±4	-	-	-
5	Madzharovo mine,	0–5	52±6	5±1	1432±20	55±6	92±10	2.5±1.0	40±3
6	LZC, Kardzhali	0–5	64±7	1±0.5	750±20	56±7	65±8	2.6±0.5	70±8
7	Marble quarry, Golyama Chinka Village	0–5	26±4	12±1	260±10	30±5	52±7	1.5±0.5	22±2
8	Fossils after the village of Kandilka	0–5	45±5	7±2	300±10	42±5	53±6	2±0.5	16±2
9	Arable soil the village of Razhenovo	0–5	60±5	6±1	850±20	62±5	70±8	3±0.5	82±7
10	Arable soil, Krumovgrad	0–5	50±8	26±4	26±4	40±5	36±5	2±0.5	30±4
11 - 1	Arable soil, Krumovgrad (yard)	0–5	36±6	87±4	540±10	28±4	42±5	1.5±0.5	35±4
11 - 2	Arable soil, Krumovgrad (yard)	5–10	52±8	76±3	570±10	35±6	33±5	1.8±0.5	33±3
11 - 3	Arable soil, Krumovgrad (yard)	10–20	57±10	76±3	570±10	38±5	30±4	1.7±0.5	34±3
12	Undisturbed soil, Krumovgrad	0–5	28±5	7±1	540±20	35±5	25±4	1.6±0.5	35±5
13	Arable soil, Leshnikovo village	0–20	45±6	35±3	670±20	35±6	31±6	2±0.5	40±3
14	min and max value	0–20	26 - 64	< 1 - 87	26 - 1432	9 - 62	21 - 92	1 - 3	16 - 82

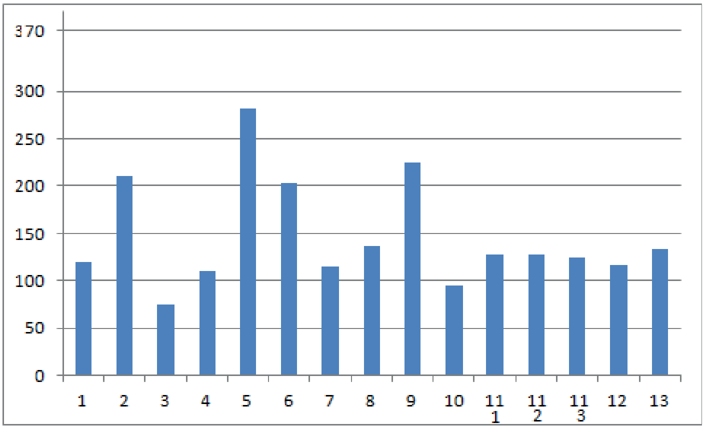

Figure 3. Radium equivalent activity (Ra_{eq}).

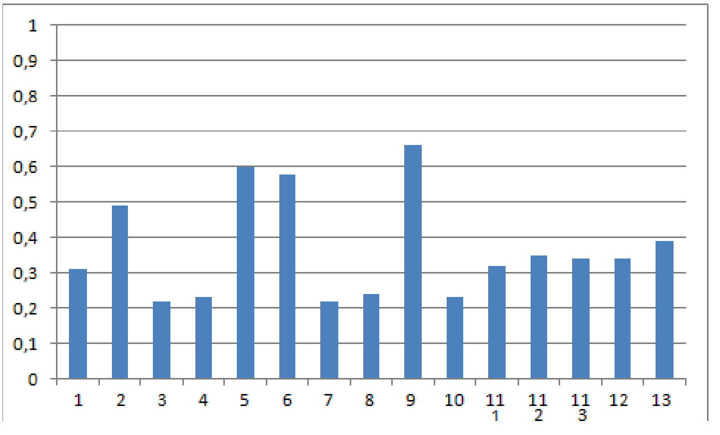

Figure 4. External Hazard Index (H_{ex}).

Discussion

The study covers unexplored areas of the Eastern Rhodopes affected by a variety of human activities, the result of which may cause additional radiation pollution to the soil. Sampling was carried out in close proximity to such areas.

Natural radioactivity

Radioactivity levels in the environment depend on geological aspects, mainly on the composition of rocks and soil, where natural radionuclides are found in varying concentrations (Raykov 1978; Négrel et al. 2018). The Rhodopes are rich in uranium and other ore deposits. Exposure to various human activities can increase the proportion of natural radioisotopes in the effective dose. This can pose a potential risk to humans and living organisms, as 75% of the radiation received by mankind is due to natural sources of radiation. (Ghiassi-Nejad et al. 2001).

A comparison is made between the radioisotopes content in the soils from the studied areas and the average values of natural radionuclides in undisturbed soils from different regions of the world, Bulgaria, the Western Rhodopes, and the Sofia field. (Yordanova et al. 2005, 2015; Lazarova et al. 2019).

The publications of the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR 2000) report average values of the content of natural radionuclides in Bq/kg in undisturbed soils from different regions of the world. The data is presented in Table 2.

For the study region, the specific activity of the natural radionuclides ^{238}U, ^{226}Ra, ^{232}Th, ^{40}K, ^{210}Pb in Bq/kg was in the following range: ^{238}U – 9÷62; ^{226}Ra – 11÷92; ^{232}Th –16÷82; ^{40}K – 260÷1432 and ^{210}Pb – 26÷64 (Figs 5–7).

Content of natural radionuclides in the studied soils was within the background amounts and was comparable to global averages (Figs 5–7). The differences between the Eastern and Western Rhodopes could be explained by the predominance of sedimentary rocks in the Eastern Rhodopes. Sedimentary rocks, especially of biogenic origin, have a very low content of radioactive elements.

Table 2. Content of ^{238}U, ^{226}Ra , ^{232}Th and ^{40}K in Bq/kg in various regions of the world.

	^{40}K	^{238}U	^{226}Ra	^{232}Th
World average	400	35	35	30
Europe	40÷1650	2÷330		2÷190
Bulgaria	40÷800	8÷190	12÷210	7÷160
average	(400)	(40)	(45)	(30)

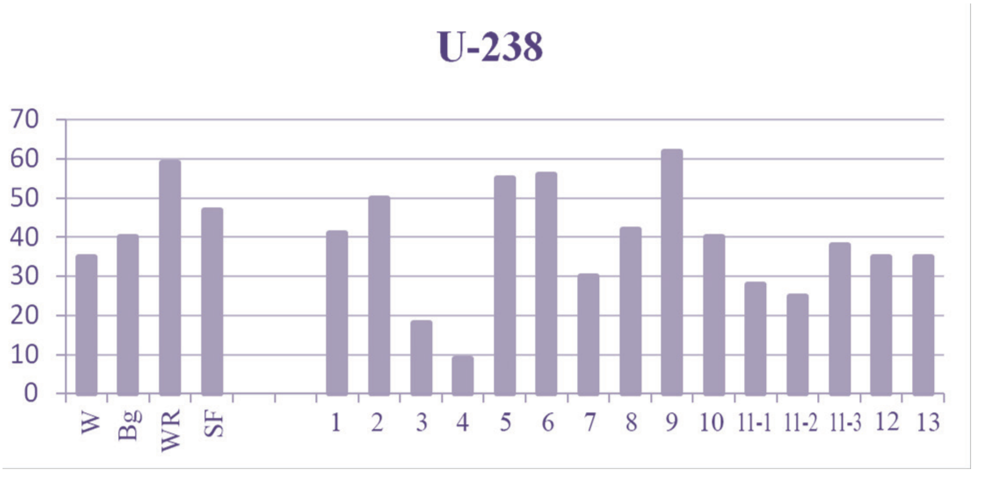

Figure 5. Content of ^{238}U in soil samples – world average (W), average for Bulgaria (Bg), average for the Western Rhodopes (WR), average for the Sofia field (SF) and in the studied samples.

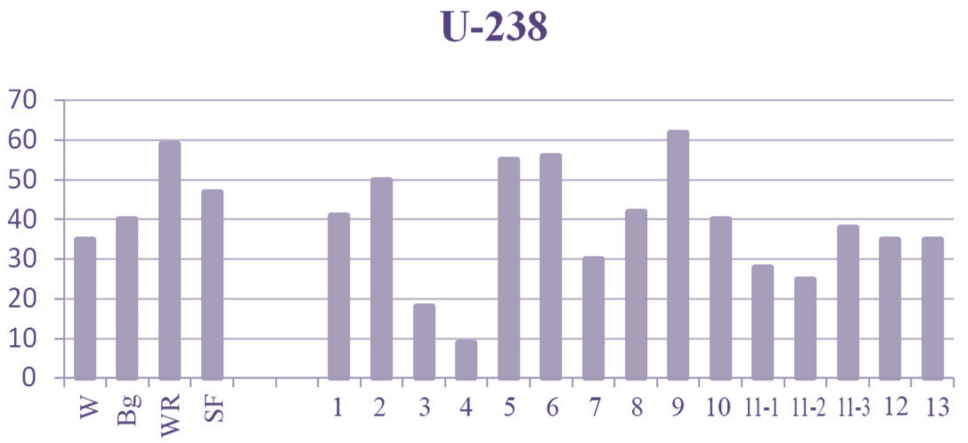

Figure 6. Content of ^{226}Ra in soil samples – world average (W), average for Bulgaria (Bg), average for the Western Rhodopes (WR), average for the Sofia field (SF) and in the studied samples.

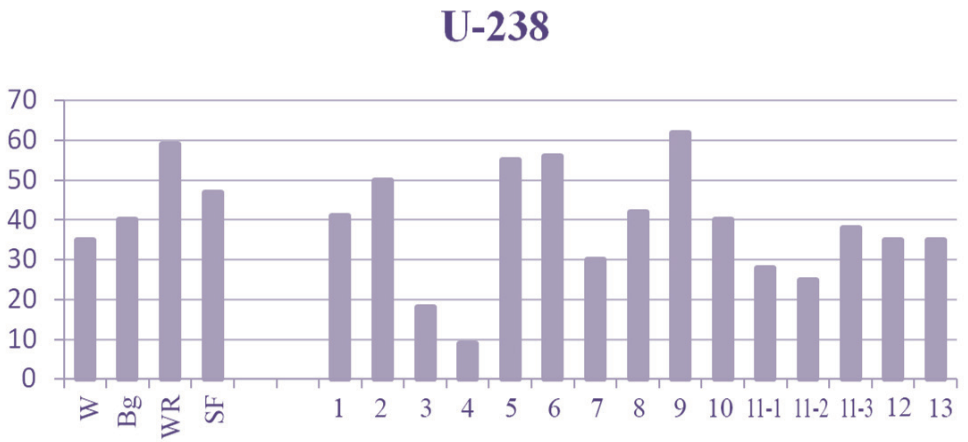

Figure 7. Content of ^{232}Th in soil samples – world average (W), average for Bulgaria (Bg), average for the Western Rhodopes (WR), average for the Sofia field (SF) and in the studied samples.

Potassium is an important element. It is found in all living organisms. The importance of the radioactive isotope ^{40}K is mainly due to its long half-life (1.28 × 109 years) and its ubiquity.

World average specific activity of ^{40}K (activity per unit mass of soil) is 370 Bq/kg, ranging from 100 to 700 Bq kg (Mcaulay and Moran 1988).

Content of ^{40}K in Bulgarian soils varies significantly – from 40 to 800 Bq/kg. For the studied area it ranges from 30 to over 1400 Bq/kg (Fig. 8). This wide range is characteristic of cinnamon forest soils, predominant in the studied region, due to the great variety of soil-forming rocks in these soils. In the soil samples from the Ada Tepe

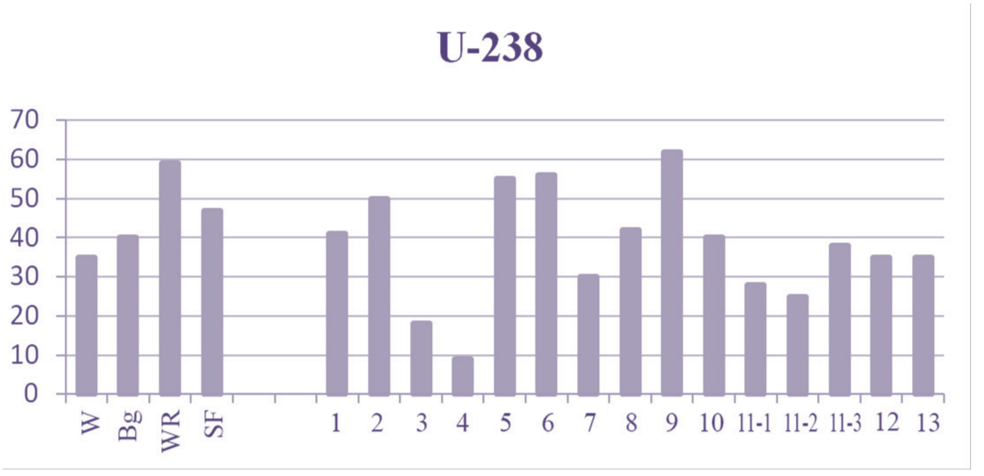

Figure 8. Content of ^{40}K in soil samples – world average (W), average for Bulgaria (Bg), average for Western Rhodopes (WR), average for the Sofia field (SF) and in the studied samples.

gold mine and the Madjarovo mine, the measured values were two times higher than the world average. This can be explained by the presence of sedimentary rocks rich in organic remains.

A shift in the radioactive equilibrium between ^{238}U and ^{226}Ra and higher content of radium was detected in some of the soil samples. It may be due to the lower mobility of radium in soils, the low humus content and the greater solubility of uranium salts allowing U migration along the soil profile.

No significant differences were found in the radiation status of arable and non-arable soils. The results of the studied radionuclides correspond to the average or slightly above average values, typical for the geographical latitude of Bulgaria and were within the values cited in the literature as normal for the respective regions.

Technogenic radioactivity

Radioactive contamination introduces new elements into the ecosystem. As a result of nuclear tests in the 1960s and the Chernobyl accident, ^{137}Cs entered the environment. For the study area, the specific activity of the technogenic radionuclide is in the range (1–87) Bq/kg.

After the Chernobyl nuclear accident, the southern part of Bulgaria was most affected. Four to five times higher activity concentrations were measured in the soils of Southern Bulgaria than in those of the Northern part (Yordanova et al. 2007). The Rhodopes have received significant amounts of radioactive ^{137}Cs. Through the rains, it entered the soil, bound to the surface soil layer and was redistributed into the ecosystem, where it remained for a long time due its long half-life.

From the data on the specific activity and dynamics of ^{137}Cs it can be seen that the soil pollution was non homogeneous as a result of air transport. It varied between

3 and 1700 Bq/kg, even within small areas (tens of square meters) (Yordanova et al. 2007). This was also confirmed by the present studies where its content was ranging from <1 to 87 Bq/kg (Fig. 9). The total technogenic γ-activity in the soils of Bulgaria has increased between 10 and 300 times after the Chernobyl accident. For ^{137}Cs this excess was 3–10 times, in some cases reaching up to 50 times above the characteristic background values. BFSA (2012).

Until 1986, the average value of ^{137}Cs specific activity in Northern Bulgaria was 10 Bq/kg and in Southern Bulgaria – 26 Bq/kg. (Naydenov and Staneva 1987a, b). After 1990, cesium-137 could be detected in all soil samples. (Yordanova et al. 2014). In 1996, due to the high inhomogeneity, the average values of the isotope varied between 160 and 280 Bq/kg for Southern Bulgaria and between 40 and 60 Bq/kg – for Northern Bulgaria. (Yordanova et al. 2007). According to the data of the Executive Agency for Environment (2016), the highest values were registered in the Western Rhodopes (Chetroka region – 261/kg and the town of Laki – 189 Bq/kg;). 35 years after the Chernobyl accident, the present study recorded specific activity of cesium-137 from <1 to 87 Bq/kg. This decrease of ^{137}Cs activity in the surface soil layer was mainly due to its radioactive decay.

The Eastern Rhodopes are rich in water. Sites such as the Ada Tepe gold mine, the Madzharovo mines, the Kardzhali Lead-Zinc Complex, are located in close proximity to settlements or to large water sources. A study on the radiological impact of uranium mining on surface waters and sediments closure shows that the migration of U_{nat}, ^{226}Ra, ^{210}Pb and ^{232}Th through surface water is one of the major pathways for contamination spread. (Ivanova et al. 2015). Most of the sites are near surface water bodies and are potentially dangerous for groundwater bodies. This poses an environmental risk in case of possible pollution, not only for humans but also for all living organisms, as the Eastern Rhodopes have the greatest species variety in Bulgaria and is one of the richest terri-

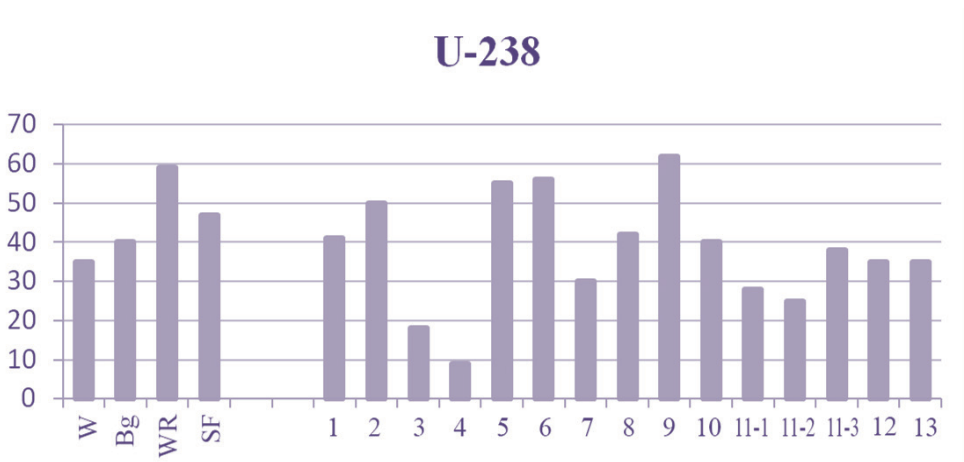

Figure 9. Content of ^{137}Cs in soil samples – average for Western Rhodopes (WR), average for the Sofia field and in the studied samples.

tories in aspect of biodiversity in Europe (Ministry of Environment and Water 2013). Many Balkan endemics and highly endangered species inhabit the region. The studied areas are close to regions of the European ecological network Nature 2000 and are of growing interest for tourism. In this regard a further research and assessment of the radiological status of ground and surface water in the region could be recommended.

The radium equivalent index (Ra_{eq}) and the external hazard index (H_{ex}) were used to assess the results obtained with respect to potential radiation hazard to the population. The data for H_{ex} do not exceed the permissible upper limit 1. Thus, with an external hazard index below 1 and low Radium equivalent activity, in relation to natural radionuclides in the soil, the study area of the Eastern Rhodopes is within normal background amounts and does not pose a radiation hazard for the population and biota in the area.

Conclusion

The analysis of data obtained showed the natural radionuclides content in studied soils does not differ considerably from the average values for our latitudes cited in the literature.

The measured ^{137}Cs content in the samples was as a result of the global fallout and the Chernobyl accident.

No additional pollution and impact of industrial activities on the content of radionuclides was found.

The External Hazard Index (Hex) showed the content of the studied radionuclides was not dangerous for the biota in the region from radiological point of view.

Due to the systematic use of unregulated drinking water sources in the region, a recommendation is given for the radiological assessment of ground and surface water in the studied areas.

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Supplementary material 1

Figure S1

Authors: Milena Hristozova, Radoslava Lazarova

Data type: jpg file

Explanation note: Content of 238U, 232Th and 226Ra in soil samples – world average (W), average for Bulgaria (Bg), average for the Western Rhodopes (WR), average for the Sofia field (SF) and in the studied samples.

Link: https://doi.org/10.3897/biorisk.17.77432.suppl1

Supplementary material 2

Figure S2

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Data type: jpg file

Explanation note: Content of 238U, 232Th and 226Ra in soil samples – world average (W), average for Bulgaria (Bg), average for the Western Rhodopes (WR), average for the Sofia field (SF) and in the studied samples.

Link: https://doi.org/10.3897/biorisk.17.77432.suppl2

Supplementary material 3

Figure S3

Authors: Milena Hristozova, Radoslava Lazarova

Data type: jpg file

Explanation note: Content of 238U, 232Th and 226Ra in soil samples – world average (W), average for Bulgaria (Bg), average for the Western Rhodopes (WR), average for the Sofia field (SF) and in the studied samples.

Link: https://doi.org/10.3897/biorisk.17.77432.suppl3

Supplementary material 4

Figure S4

Authors: Milena Hristozova, Radoslava Lazarova

Data type: jpg file

Explanation note: Content of 40K in soil samples – world average (W), average for Bulgaria (Bg), average for Western Rhodopes (WR), average for the Sofia field (SF) and in the studied samples.

Link: https://doi.org/10.3897/biorisk.17.77432.suppl4

Supplementary material 5

Figure S5

Authors: Milena Hristozova, Radoslava Lazarova

Data type: jpg file

Explanation note: Content of 137Cs in soil samples – average for Western Rhodopes (WR), average for the Sofia field and in the studied samples.

Link: https://doi.org/10.3897/biorisk.17.77432.suppl5