Natural Radionuclides in Soils from Selected Regions in Bulgaria Affected by Natural and Anthropogenic Processes

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Abstract

The content of natural radionuclides ^{40}K, ^{235}U, ^{238}U, ^{232}Th and ^{226}Ra in soils from two regions of past uranium mining in Bulgaria were determined and analyzed and results of radiation monitoring summarized. Soil samples from three other regions in the country: North Bulgaria - plane region (around "Kozloduy" Nuclear Power Plant and the Danube River) – 40 samples; semi-mountainous - (Sofia field) – 5 samples and South Bulgaria – mountainous (Rhodope massif) – 25 samples, were analyzed for comparison.

Evaluation of possible radiation hazard to the population in the respective regions was made by calculatingthe average radium equivalent activity (Ra_{eq}) and the external hazard index (H_{ex}). It was found that content of natural radionuclides in the studied soils, both plain and mountainous, is within normal background levels and is not hazardous to the population.

Key words: radionuclides ^{40}K, ^{235}U, ^{238}U, ^{232}Th and ^{226}Ra, radiation monitoring of soils, radioecology

Introduction

Anthropogenic impact on environment has severely increased in recent decades leading to serious ecologic problems. Areas affected by mining industry which are highly contaminated are of particular interest due to the hazards for human health and the state of the landscape. Such is the catchment area of the left tributary of the river Taina, municipality of Novi Iskar, where underground and open uranium mining were carried out in the "Iskra" section and the area along Mesta river valley around Eleshnitsa mine. (Misheva at al., 2009, Yordanova at al., 2011 a, Filchev and Yordanova, 2011, Yordanova at al., 2015)

Water catchment of the river Taina occupies an area of 4,775 km^2. The altitude of the catchment varies from 500 m at the flowing in the Iskar river to 964 m in the highest part of the catchment area. The main soil types in the area of research are leached Cinnamonic forest soils - 45.8% and Brown forest soils -36.37%. The remaining 17.4% of the catchment is covered with Anthropogenic soils, reflecting mainly the human impact on the river basin.

The village of Eleshnitsa is located 12 km North-east of Bansko at the southern foot of North-west of the Rhodope massif "Belakite" along the two banks of Zlataritsa river lower stream. The river Zlataritsa flows into the Mesta river, which passes near Eleshnitsa. Warm air from Aegean Sea passes along Mesta valley.

Fig. 2. *Engineering activity on the closure of Eleshnitsa Mine*

Both the Belakis and other surrounding mountain massifs and plane areas are covered with pine trees and partly with beech and oak forests. The land of the village is relatively large. It covers about 300 km^2 of predominantly mountainous and semi-mountainous area. To the west and northwest it is bordered by the land of the village of Babiak, to the north by Gul-tepe peak, and to the East and Southeast by the land of the villages of Gostun and Obidim. The Mesta river and the land of the town of Dobrinishte and the town of Bansko serve for south and southwest border respectively .

In Eleshnitsa, mining was carried out under the classical method. In plant "Zvezda" only flotation was carried out and the enrichment in Buhovo.

The objective of the present study was to determine the content of natural radionuclides ^{40}K, ^{235}U, ^{238}U, ^{232}Th and ^{226}Ra in soils from the two regions of past uranium mining in Bulgaria – "Iskra" section, Novi Iskar and Eleshnitsa and to evaluate possible radiation hazard to the population in these regions.

Materials and Methods

Soil samples from 10 points in the area of the river Taina, Iskra section, and 11 points along Mesta river valley, Eleshnitsa mining area, were collected and analyzed.

Soil samples from three other regions of the country: North Bulgaria – plain area (around Kozloduy Nuclear Power Plant and along the Danube River) - 40 samples; semi-mountainous (Sofia field)- 5 samples and South Bulgaria - mountainous (the Rhodope massif) –25 samples, were also taken and determined for comparison. Collection and preparation of samples was carried out in accordance with BDS 17.4.5.01 and ISO 18589-2. Soil samples were homogenized, dried at 80 ° C and crushed through 2 mm sieve. They were analyzed for contents of natural radionuclides ^{40}K, ^{235}U, ^{238}U, ^{232}Th and ^{226}Ra in the Testing Laboratory of Radioecology and Radioisotopic Research at ISSAPP "N.Pushkarov ". The laboratory has been accredited by EA "BAS" under BDS EN ISO / IEC 17025: 2006 since 2002, and it has been declared by the Ministry of Agriculture and Food as National Reference Laboratory in the field of radiometry since 2007. Validated interlaboratory methodologies were used (Naidenov et al., 2001).

Activity concentration of natural ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in soil samples was determined by gamma-spectrometric analysis with Multi Channel Analyzer DSA 1000, CANBERRA, and ultra-clear Ge detector. Measurements were carried out according to the requirements of ISO standard 18589-3, using gamma spectrometer with a pure Ge detector with 20% efficiency and 1.8 keV resolution for the ^{60}Co line of 1332 keV energy. The credibility of results is confirmed by regular interlaboratory comparisons and proficiency testing schemes. The time for measuring one sample varies between 19 and 24 hours.
^{226}Ra was determined by the full energy absorption peak of 186.3 keV, with correction for ^{235}U (185.6 keV). Activity of ^{238}U was determined by daughter product ^{234}Th (63.3 keV and 92.3 keV). Gamma-lines of ^{228}Ac (911.0 keV) and ^{208}Tl (583.3 keV) were used for ^{232}Th.

Results and Discussion

Figures 1 and 2 present the results from the analyzes of the content of natural radioactive elements ^{40}K, ^{235}U, ^{238}U, ^{232}Th and ^{226}Ra at 10 points of Taina river area and 11 points along the Mesta river in Eleshnitsa mining area.

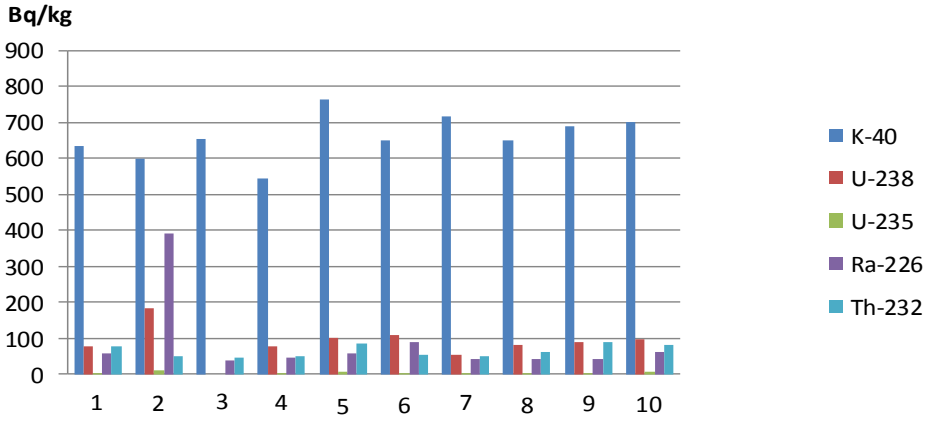

Fig. 1 *Content of ^{40}K, ^{235}U, ^{238}U, ^{226}Ra and ^{232}Th in soil samples from 10 points in the area of the river Taina.*

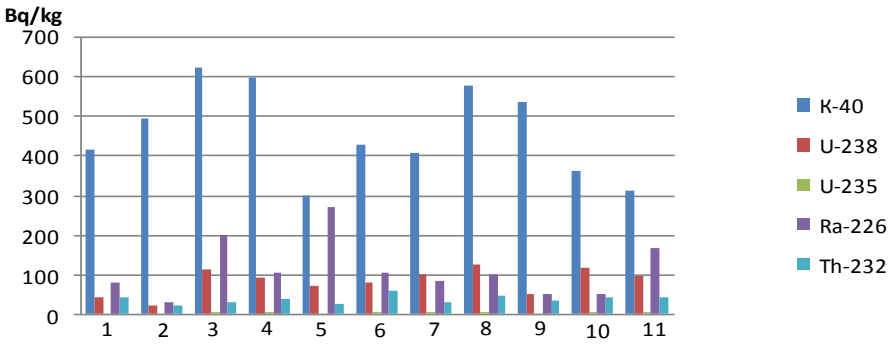

Fig. 2. *Fig.2. Content of ^{40}K, ^{235}U, ^{238}U, ^{226}Ra and ^{232}Th in soil samples from 11 points along the Mesta river valley, Eleshnitsa mining area.*

The average results from the analysis of natural radionuclides contents in the soils from separate areas of research are presented in Table 1. The standard deviation obtained is presented in absolute terms and percentage in brackets.

Table 1. *Average values of natural radionuclides content in soils from five regions of the country in Bq.kg^{-1} dry weight.*

	^{238}U [Bq.kg^{-1}]	^{226}Ra [Bq.kg^{-1}]	^{232}Th [Bq.kg^{-1}]	^{40}K [Bq.kg^{-1}]
North Bulgaria (plane area)	33±8(24%)	32±10(31%)	34±9 (26%)	456±106(23%)
Sofia field (semi-mountainous)	26±11(42%)	31±10(31%)	40±11(27%)	447±97(22%)
South Bulgaria (Rhodope mountain-mountainous)	62±24(39%)	46±22%(48)	58±16(27%)	686±215(31%)
Mesta river valley(Eleshnitsa mining area-semi-mountainous)	73±23(31%)	51±17(33%)	57±16(28%)	735±122(16%)
River Taina catchment, "Iskra" section	98±35(36%)	35±20(57%)	37±20(54%)	458±108(24%)

Values presented in the table for radionuclides studied correspond to average values characteristic for the latitude of Bulgaria estimated and reported by International Atomic Agency, Vienna (UNSCEAR, 1993, Kinova, 1997, Yordanova at al., 2005, Yordanova at al., 2011 b). The average values reported are 40 Bq.kg^{-1} dry weight for ^{238}U, ^{226}Ra and ^{232}Th, and 580 Bq.kg^{-1} for ^{40}K respectively. Slightly higher values found in Rhodope mountain and Mesta river valley are due to the presence of rocks as gneiss, shale, granite, etc. with higher content of natural radionuclides on which soils in the area have been formed (Montes. et al., 2012).

Evaluating radiation hazard

Radium equivalent activity index (Ra_{eq}) and external hazard index (H_{ex}) were used to evaluate the results obtained in terms of radiation hazard to the population in the areas of research.

Radium equivalent activity was calculated by the following formula (Beretka, Mathew, 1985):

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (1)$$

Where A_{Ra}, A_{Th} and A_K are the activities in Bq.kg^{-1} of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The calculation is based on the assumption that 370 Bq.kg^{-1} of ^{226}Ra, 259 Bq.kg^{-1} of ^{232}Th and 4810 Bq.kg^{-1} of ^{40}K produce the same gamma-ray dose. The limit value is 370 Bq.kg^{-1}.

External hazard index was estimated by the following formula (Krieger, 1981):

$$H_{ex} = A_U / 370 + A_{Th} / 259 + A_K / 4810 \leq 1 \quad (2)$$

Where A_U, A_{Th} and A_K are the activities in Bq.kg^{-1}of ^{238}U, ^{232}Th и ^{40}K respectively.
The results obtained are presented in Table 2.

Table 2. *Average values of Ra_{eq} [Bq.kg^{-1}] and H_{ex} in soils from the five regions of the country*

	Ra_{eq} Bq.kg^{-1}	H_{ex}
North Bulgaria(plane area)	115.73	0.31
Sofia field (semi-mountainous)	122.62	0.32
South Bulgaria (Rhodope mountain- mountainous)	181.76	0.54
Mesta river valley (Eleshnitsa mining area- semi-mountainous)	189.10	0.57
River Taina catchment, “Iskra” section	123.45	0.49
Maximumvalue	370	1

The values of the radiation hazard evaluation of gamma background resulting from natural radionuclides content in the soils from studied areas confirm the population safety from radiological point of view.

Conclusion

Content of natural radionuclides radionuclides ^{40}K, ^{235}U, ^{238}U, ^{232}Th and ^{226}Ra in soils from two regions of past uranium mining (Iskra section, Novi Iskar and Eleshniza, the Mesta river valley) were determined along with soil samples from three other regions of the country used for comparison. Results of radiation monitoring in the five regions of study were summarized. Values for natural radionuclides under research correspond to average values characteristic for the latitude of Bulgaria reported by the International Atomic Agency, Vienna. Slightly higher values found in the Rhodope mountain in south Bulgaria and Mesta valley are due to the presence of rocks as gneiss, shale, granite, etc. with higher content of natural radionuclides on which soils in the area have been formed.

The radium equivalent activity index (Ra_{eq}) and external hazard index (H_{ex}) indicate that the contents of the natural radionuclides in soils from the studied regions, both plane and mountainous, is within the normal background quantities and does not pose radiation hazard to the population.

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