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Content of natural and technogenic radionuclides in Bulgarian soils

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Abstract

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This study presents data for the content of natural radionuclides from uranium-radium and thorium series, potassium-40 and technogenic cesium-137 measured in soils from North and South Bulgarian regions in the period 2017-2018.

Analysis of results of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K content showed the values were within the range cited in the literature as typical for the soils from the respective regions. Slightly higher values were observed in the Rhodopes and Mesta River valley due to the presence of rocks such as gneisses, shales, granites, etc., containing higher contents of natural radionuclides, on which the soils in the area were formed.

Comparing the data with those obtained in previous years, it was concluded that there were no significant changes in the content of cesium-137 in the surface layer of soils and the decrease in activity is mainly due to the radioactive decay and not to significant migration along soil horizon or extraction from the soil by plants.

From the results obtained, it can be concluded that no contamination was observed in recent years, either as a result of nuclear accidents or uncontrolled release of radioactive waste.

Keywords: natural and technogenic radionuclide; radiological monitoring of soils

Introduction

The study of the content and behavior of uranium and other naturally occurring radioactive elements in soils is essential, as they, along with their daughter products, form the main part of dose load on living organisms and humans. The majority of natural radionuclides are long-lived alpha-emitters and besides being in the group of highly radiotoxic elements, they are toxic as chemical elements.

In addition to natural radioactive elements, technogenic radionuclides also contribute to dose load on population and can cause extremely serious adverse effects on human health in an event of a serious emergency. Chernobyl (1986) and Fukushima (2011) accidents were examples for this from the recent past. This requires the regular monitoring of radioecological situation in the country and in particular of soils as a reservoir and main source of radionuclide input into human body along the soil-plant-human chain.

In this regard, the purpose of the present study was to determine the current radiological status of soils from North and South Bulgaria by 2018.

Materials and Methods

The regular radiological monitoring of soils in Bulgaria is carried out following a sampling strategy described elsewhere (Yordanova et al., 2014). The monitoring is conducted on every two years – soils from North Bulgarian regions are sampled during the first year and on the next– soils from South Bulgaria are collected.

In order to assess the impact of Kozloduy Nuclear Power Plant (NPP) on the radioecological status of soils in North Bulgaria, soil samples from the monitoring network points along the Danube river basin (east and west from the NPP, from Novo Selo, Vidin to Silistra) are collected along with soil samples from the 30 km exclusion zone around the NPP.

To monitor the pollution of the Southern regions in the country, soils from the Rhodopes and Mesta River valley are sampled.

Preparation of samples is carried out according to Bulgarian State Standard (BSS) 17.4.5.01 and BSS ISO 18589-2.

Soil samples were homogenized, dried at 80°C and sieved through a 2 mm sieve. Specific activity of natural ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K and technogenic ^{137}Cs was determined by gamma-spectrometric analysis under method validated in the Laboratory (Naydenov et al., 2010). A DSA 1000 multi-channel analyzer product of CANBERRA and ultra-pure Ge-detector were used. Measurements were carried out according to ISO 18589-3 using gamma spectrometer with 25% efficiency and 1.8 keV resolution for 1332 keV energy peak of ^{60}Co. Reliability of results is confirmed by regular participation in interlaboratory comparisons and proficiency tests. Measurement time varied between 19 and 24 hours.

^{226}Ra was determined by the full energy peak at 186.3 keV, with correction for ^{235}U (185.6 keV). The activity of ^{238}U was determined by the daughter product ^{234}Th (63.3 keV and 92.3 keV). For ^{232}Th, the ^{228}Ac (911.0 keV) and ^{208}Tl (583.3 keV) gamma lines were used. The specific activity of ^{137}Cs was determined by the energy peak at 661.62 keV.

Results and Discussion

Gamma-spectrometric analysis of soils from North Bulgaria

Content of natural radionuclides ^{238}U, ^{235}U, ^{226}Ra, ^{232}Th, ^{40}K and technogenic ^{137}Cs in soils (0-5 cm layer) from North Bulgaria (30 km zone of Kozloduy NPP and along the Danube river basin) for 2017 are presented in Table 1. The average activity concentration of each radionuclide in 0-5 cm soil layer and the respective standard deviation in absolute value were calculated.

From the results obtained it can be seen that the content of natural radioactivity in soils from North Bulgaria is relatively homogeneous and is within the range typical for the

Table 1. Content of ^{238}U, ^{235}U, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs (Bq/kg) in soils (0-5 cm depth) from North Bulgaria (Kozloduy NPP area and the Danube river basin) (2017)

Sampling point	^{137}Cs	^{40}K	^{238}U	^{235}U	^{226}Ra	^{232}Th
Vratsa	5±1	390±20	56±6	2.5±0.5	35±5	50±3
Sofronievo	3±1	640±30	40±5	1.8±0.5	30±6	40±5
Butan	7±1	650±40	35±6	1.5±0.5	25±3	48±4
Hairedin	7±1	690±40	40±5	1.8±0.5	43±5	40±5
Septemvriitsi	12±1	660±30	50±6	2.3±0.5	30±5	40±4
Bazovets	7.5±1.5	660±30	55±8	2.5±0.5	40±5	50±6
Glozhene	9±1	580±30	40±5	1.8±0.5	30±5	36±5
Kozloduy NPP (catchment)	2±1	620±30	55±6	2.5±0.5	45±5	45±5
Kozloduy NPP (Akatsieva gorichka)	10±1	410±20	35±6	2.0±0.5	42±5	35±5
Kozloduy (Boteva aleya)	14±1	850±40	48±5	2.5±0.5	40±5	54±5
Kozloduyport	12±1	520±30	27±4	≤1	28±4	30±4
Dolni Tsibar	6±1	680±30	36±4	2.0±0.5	35±4	40±5
Leskovets	24±1	440±20	40±6	1.8±0.5	43±5	40±5
Byala Slatina	13±1	600±30	46±6	2.0±0.5	44±6	36±5
Knezha	15±0.1	600±30	37±5	2.0±0.5	63±5	67±5
Dekov	4.2±0.5	620±30	49±5	2.5±0.5	49±6	45±4
Lozitsa	4.3±0.5	660±30	57±6	3.0±0.6	56±6	67±5
Belene NPP	2.0±0.2	410±20	30±5	≤ 1	35±5	46±5
Oresh	6±1	620±30	48±5	2.2±0.5	46±5	46±5
Svishtov (Sviloza)	20±2	500±20	41±5	3.0±0.5	56±8	41±4
Average +/−SDV	10.4±7.3	602.3±109.8	41.8±8.2	2.0±0.5	40.2±9.5	40.1±9.6

soils of the region. No significant deviation from the radioactive equilibrium U-238: Ra-226 as members of one radioactive family was found.

Comparing the data for the activity concentration of ^{238}U, ^{226}Ra and ^{232}Th in the samples under study with those obtained in previous years (Figure 1), no significant changes were observed. The results were within the range of the values cited in the literature as typical for the region concerned (UNSCEAR 1982, UNSCEAR 2000).

Standard deviation calculated for Cs-137 activity concentration in 0-5 cm soil layer (Table 1) shows the heterogeneity of surface contamination which has been observed in all the years after Chernobyl accident (Yordanova et al., 2014).

The average data for the specific activity of cesium-137 in different years (Figure 1) indicated that radionuclide content in soils resulted from the global fallout and the Chernobyl accident. No additional contamination due to the operation of Kozloduy NPP or transboundary transfer of radioactive materials was observed.

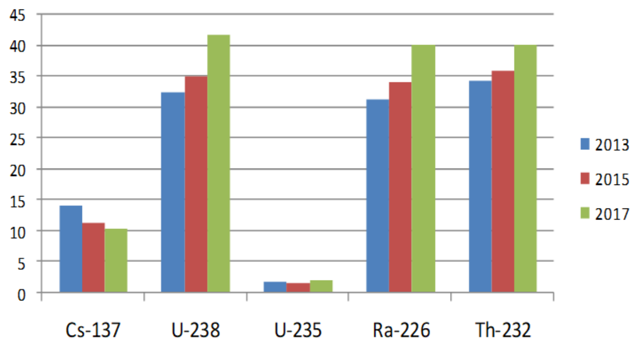

Fig. 1. Content of radioactive elements [Bq/kg] in soils from Kozloduy NPP area and the Danube river basin

Gamma-spectrometric analysis of soils from South Bulgaria – the Rhodopes and Mesta River valley

The results of the gamma-spectrometric analysis of soil samples (0-5 cm layer) from the Rhodopes and the Mesta river valley are presented in Tables 2 and 3 respectively.

Comparing the average results obtained for natural radionuclides – ^{238}U, ^{226}Ra and ^{232}Th with data from previous years (Figure 2), no significant changes were found. The values were within the range cited in the literature as normal for the regions concerned. Slightly higher values were found in soils from South Bulgaria due to the presence of rocks like gneisses, slate, granite and others containing higher content of natural radionuclides, on which the soils in the area were formed (Álvarez et al., 2018; Muthamilselvanet al., 2018; Montes et al., 2012).

A higher concentration of natural radioactivity was measured along the Mesta River valley, near Eleshnitsa (^{238}U – 150±40 Bq/kg, ^{226}Ra – 280±50 Bq/kg), an area of past uranium mining and processing. The data also showed significant deviation of ^{238}U/^{226}Ra ratio indicating anthropogenic impact. Similar results were also reported in previous studies of the area (Yordanova et al., 2011; Yordanova et al., 2015; Lazarova et al. 2017).

The standard deviation calculated for the activity concentration of ^{137}Cs, showed significant variation indicating the heterogeneity of the pollution.

More than 30 years have passed since the Chernobyl accident in 1986 causing the main ^{137}Cs soil contamination.

Table 2. Results of gamma-spectrometric analysis of soil samples (0-5 cm depth) from the Rhodopes [Bq/kg] (2018)

Sampling point	^{137}Cs	^{40}K	^{238}U	^{226}Ra	^{235}U	^{232}Th
Chepelare	170±10	620±30	73±10	40±18	2±1	57±7
Rojen area	45±4	450±20	60±16	56±10	2±1	44±5
Snezhanka	138±10	920±40	130±10	70±10	5±1	68±6
Smolyan (fishpond)	7±1	950±40	118±6	77±010	5±1	80±8
Smolyan (Kashlite)	56±5	800±40	102±10	80±8	5±2	80±8
Smolyan Lakes	103±5	720±30	55±6	70±7	3±1	62±6
Smolyan (Kriva Reka)	82±4	820±40	54±6	72±7	3±1	64±6
Stoykite	87±4	800±40	32±5	36±6	2±1	30±3
Shiroka Laka	30±2	1020±50	46±5	65±10	2±1	80±4
Stickel	34±2	700±40	31±4	36±5	2±1	48±4
Solyshta	27±1	720±40	28±3	35±5	< 1	40±4
Borino	84±4	950±30	30±3	40±10	1±0.5	38±4
Dospat (Kasaka)	56±3	620±30	32±6	74±10	2±1	68±6
Batak (dam lake)	169±8	480±20	58±6	68±10	2±1	47±5
Velingrad	73±4	900±50	35±10	40±10	2±1	70±7
Average +/−SDV	77±50	765±172	59±33	57±17	3±1	58±16

Table 3. Results of gamma-spectrometric analysis of soil samples (0-5 cm depth) from Mesta River valley [Bq/kg] (2018)

Sampling point	^{137}Cs	^{40}K	^{238}U	^{226}Ra	^{235}U	^{228}Th
Yundola	104±5	880±40	43±8	32±	2±1	45±4
Yakoruda	12±1	900±50	80±10	56±10	3±1	70±7
Eleshnitsa	12±1	1020±50	150±40	280±50	10±2	60±6
Dobrinishte	12±1	840±40	80±20	100±20	5±1	100±20
Bansko	22±1	700±40	65±10	75±10	3±1	64±6
Predel	30±2	900±50	40±10	56±10	2±1	74±7
Belitsa	44±2	760±30	60±15	80±10	3±1	52±6
Cherna Mesta	7±1	720±30	50±5	54±10	2±1	48±4
Average +/−SDV	30±32	840±108	71±35	92±79	4±3	64±18

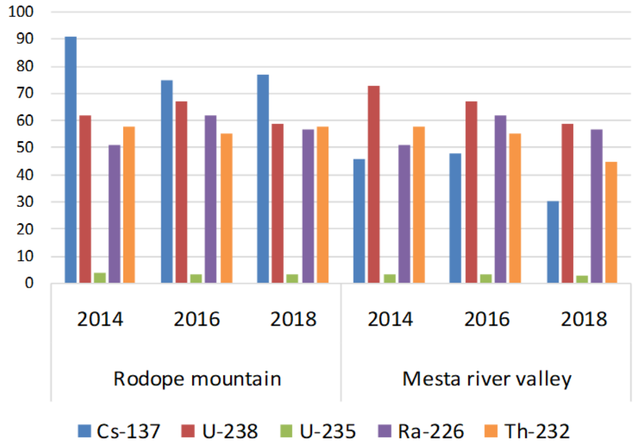

Fig. 2. Content of radioactive elements in soils from the Rhodopes and Mesta river valley

^{137}Cs half-life is 30.15 years, therefore it was of interest to compare the radionuclide content found in recent years with that measured before the accident.

In studies carried out before the accident (Naydenov, 1986; Naydenov&Staneva, 1987) the following average values of cesium-137 in soils were reported: North Bulgaria – 10 Bq/kg, South Bulgaria – 26 Bq/kg. The comparison showed the radiological status of soils in the lowlands of North Bulgaria was comparable to that of the pre-accident period. In semi-mountainous and mountainous regions of South Bulgaria (Figure 2), the values for ^{137}Cs were two to three times higher. The reason is probably two to three times higher levels of ^{137}Cs deposition in these areas in 1986.

Conclusions

Radiological monitoring of soils from North and South Bulgaria was carried out and content of natural radionuclides ^{40}K, ^{235}U, ^{238}U, ^{232}Th and ^{226}Ra and the technogenic ^{137}Cs were determined for the period 2017-2018. Activity concentration of natural radionuclides under study was within the values cited in literature for the respective regions. Slightly higher values found in the Rhodopes massif and the Mesta River valley were logical due to the presence of soil forming rocks such as gneisses, shales, granites, etc. containing higher contents of natural radionuclides.

From data obtained for the content of cesium-137 it can be concluded that no contamination with the radionuclide was observed in recent years, either as a result of accidents or uncontrolled release of radioactive waste.

References

Álvarez, J., López, L., Parra, F., Bello, C., Anzil, P., Salvatore, M., Scarlatta, L., Ferreyra, P., Miyno, S., Felkai, E., Lira, R., Hanly, A., Cuney, M. & Mercadier, J. (2018). New studies of uranium deposits related to granites in Argentina. International Atomic Energy Agency (IAEA), No. IAEA-CN-261.

BSS 17.4.5.01. (1985) Nature Protection. Soil. General Requirements for Sampling.

BSS ISO 18589-2. (2007). Measurement of Radioactivity in the Environment–Soil. Part 2: Guidance for the Selection of the Sampling Strategy, Sampling and Pre-Treatment of Samples.

ISO 18589-3 (2015) Measurement of radioactivity in the environment – Soil – Part 3: Test method of gamma-emitting radionuclides using gamma-ray spectrometry

Lazarova, R., Yordanova, I. &Staneva, D. (2017) Natural radionuclides in soils from selected regions in Bulgaria affected by natural and anthropogenic processes. *Journal of Soil Science, 2 (2)*, 106-111.

Muthamilselvan, A. & Saravanavel, J. (2018) Intrinsic Uranium concentration of crystalline rocks in parts of Central Tamil Nadu, India. *JETIR, 5 (8),*1-8.

Montes, M. L., Mercader, R.C., Taylor, M.A. &Desimoni, J.R. (2012). Assessment of natural radioactivity levels and their relationship with soil characteristics in undisturbed soils of the northeast of Buenos Aires province, Argentina. *Journal of Environmental Radioactivity,105,* 30-39.

Naydenov, M. (1986) Content of men-made radionuclides in soils from the region around Kozlodouy NPP, Scientific Reports and Announcements, Bulgarian Academy of Agriculture, Sofia, Bulgaria, 50-68 (Bg).

Naydenov, M. & Staneva, D. (1987). Composition and specificity of the contamination on the territory of the country after the accident in Chernobyl NPP, Scientific Reports and Announcements, Bulgarian Academy of Agriculture, Sofia, Bulgaria, 63-69 (Bg).

Naydenov, M., Yordanova, I., Staneva, D. &Misheva, L. (2010) Procedure for determination of natural and technogenic gamma-emitters in soils, waters and other agricultural samples. In: *Procedures for determination of radioactive elements in environmental samples*. National Center for Agricultural Science, Sofia, 5-11.

UNSCEAR (2000). Sources, Effects and Risks of Ionizing Radiation. Annex B. New York.

UNSCEAR (1982). Ionizing Radiation: Sources and Biological Effects. Annex E. New York.

Yordanova, I., Banov, M., Misheva, L., Staneva, D.& Bineva, Ts. (2015). Natural radioactivity in virgin soils and soils from some areas with closed uranium mining facilities in Bulgaria. *Open Chemistry*, *13*, 600–605.

Yordanova, I., Misheva, L., Staneva, D., Bineva, Ts. & Hadjiyanakiev, Y.(2011). Natural Radioactivity in Soil and Water from Areas with Closed Uranium Mining Facilities in Bulgaria. Scientific Reports of International Conference "100 Years Bulgarian Soil Science", Sofia, 876-881 (Bg).

Yordanova, I., Staneva, D., Misheva, L., Bineva, Ts., Banov, M.(2014). Technogenic radionuclides in undisturbed Bulgarian soils, *Journal of Geochemical Exploration*, *142*, 69-74.

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