II. MAN AND BIOSPHERE

INVESTIGATION OF NATURAL RADIOACTIVITY OF MINERAL WATERS IN SOUTHERN BULGARIA

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Abstract. The activity of naturally occurring radionuclides in mineral waters from certain most frequently used sources in Rhodope Mountains region, Southern Bulgaria has been measured with high precision by means of nuclear and radiochemical methods. The survey is pointed at radium-226 (^{226}Ra), lead-210 (^{210}Pb) and natural uranium (nat. U), which are most important from the point of view of public health in Bulgaria, because of their high toxicity and radiotoxicity in drinking water. Seventeen water sources were under investigation.

The activity concentrations of ^{226}Ra and ^{210}Pb varied from 13 to 168 mBq/L and from ≤ 1.8 to 104 mBq/L respectively. The concentration of nat. U in the mineral water was in the range ≤ 3 to 21 $\mu\text{g/L}$.

The annual effective doses were calculated for all investigated waters for adult inhabitants assuming yearly consumption of 730 litres.

The results have shown that all values of the annual effective dose of ingestion of mineral waters were below the individual dose criterion of 100 $\mu\text{Sv/y}$ reported by World Health Organization (WHO).

The obtained new results are used to assess the radiation status of the investigated waters. They will support timely and adequate measures to reduce the harmful impact of ionizing radiation on the population in cases of increased radioactivity

Key words: ^{226}Ra , ^{210}Pb , nat. U, mineral water.

INTRODUCTION

Water is the most important source of life and a major route by which radionuclides may enter the human body. Besides tap waters, mineral waters are the most consumed beverage. Bulgaria has more than 225 deposits of mineral springs having different temperature, chemical composition and radioactivity. More than 95% of all mineral water springs occur in Southern Bulgaria [1]. Therefore, their quality must be carefully and systematically controlled. Monitoring of natural radioactivity in drinking and mineral waters is important from radioecological point of view because of relatively high radiotoxicity of some of natural radionuclides and their importance in the study of cumulative radiation effects on human health [2]. Ground waters contain a number of both alpha emitters (such as $^{238,234}\text{U}$, ^{226}Ra , and ^{210}Po of the uranium-238 decay series) and beta emitters (such as ^{40}K , ^3H , ^{228}Ra of the thorium-232 decay series and ^{210}Pb of the uranium-238 decay series). Natural isotopes as ^{40}K and the nuclides from the ^{238}U and ^{232}Th series are the major source of internal and external exposure of human beings. Human activities such as mining, milling and processing of uranium ores and mineral sands, smelting of metalliferous ores, manufacturing of fertilizers, drilling of solid minerals, processing and burning of fossil fuels have raised the concentrations of naturally occurring radioactive materials in the environment [3, 4].

Water radioactivity depends on its origin and chemical composition. Radionuclides can penetrate

into ground water by processes of erosion and dissolution from rocks and minerals which form the aquifer or infiltration of rain water [5]. Radionuclide activities in mineral waters may vary in a wide range and are caused by complex solubility and transport processes in the aquifer. The hydrogeological conditions, the uranium and thorium content of the rock formations, solubility behavior and other chemical characteristics of the radionuclides are crucial for their presence in the water phase [3].

The determination of natural radioactivity in mineral water is very important from a radiological point of view. The ingestion of natural radionuclides from water poses a number of health problems and can give rise to an additional exposure dose to the stomach and to the whole body [2, 3, 6].

Recently national and EU regulations decrease the drinking water norms with the aim to strengthen consumers security concerning drinking water quality [7]. National legislation was fully harmonized with EU Directives. One of the main normative acts in this area is the Ordinance No. 9 of 16 March 2001 on the quality of water intended for drinking and domestic purposes [8].

Of all the radionuclides present in mineral waters, the isotopes of radium, uranium and lead are responsible for the major fraction of the internal dose received by humans from the naturally occurring radionuclides.

^{226}Ra ($T_{1/2} = 1600$ years) is one of the most radiotoxic and dangerous radionuclide in case of

ingestion due to its similar behavior to calcium leading to absorption of an appreciable fraction of radium and final deposition in bones [9]. The occurrence, natural distribution and concentration of ^{226}Ra in mineral waters are strongly correlated with the geological setting and the interactions of groundwater with radium bearing materials such as rocks, soil, ore bodies, etc. [10].

^{210}Pb ($T_{1/2} = 22.3$ years) is a naturally occurring member of the ^{238}U decay series. It could be found in various environmental samples. In groundwater, ^{210}Pb is found in an inorganic form, complexed with organic matter and adsorbed onto particles. ^{210}Pb plays an important role in human radiation exposure, since it possesses long residence times in the skeleton and thus highly contributes to the skeletal dose [11, 12].

Uranium is naturally occurring, radioactive heavy metal. It has three alpha emitting radionuclides: ^{234}U , ^{235}U and ^{238}U with a different atomic mass, different distribution and half-lives. More than 99 percent of uranium occurring in nature is ^{238}U [13]. It is widespread in nature, occurring in granites and various other mineral deposits. Uranium in water results from the weathering of rocks and soil [14]. Its concentration in ground and surface waters strongly depends on the geological matrix, as well as on chemical processes, such as ion exchange, sorption and precipitation [15, 16].

Independent investigations of the radioactive content of mineral waters, complementary to the national regulatory control, have been performed at Institute for Nuclear Research and Nuclear Energy, Bulgarian Academy of Sciences (INRNE, BAS) in order to confirm the recommended low level of radioactivity.

The aim of this study is to obtain a representative estimate of the concentration levels of ^{226}Ra , ^{210}Pb and nat. U in mineral waters from certain most frequently used sources in Rhodope Mountains region, Southern Bulgaria and to evaluate the radiological hazards caused by ingestion of these waters. The effective doses, resulting from the consumption of this water due to their natural radionuclide contents, were calculated.

MATERIAL AND METHODS

Study area. Rhodopes is the largest mountain in Bulgaria. The mountain lies in the Southern part of Bulgaria. It is divided in Eastern and Western Rhodopes. In geomorphological terms, the Rhodopes are part of the Rila-Rhodope massif, which is the oldest landmass on the Balkan peninsula.

A significant part of Bulgaria's hydropower resources are located in the western part of the mountain where there are many mineral water springs, the most famous being in Velingrad, Devin, Beden, Mihalkovo and others.

The western Rila-Rhodopes massif is built of Precambrian metamorphic and granite rocks, fractured by a dense system of seismically active faults. The metamorphic basin contains some large bodies of marble that act as hydrothermal reservoirs. Permeable terrigenous-clastic materials in the deep Neogene and Paleogene grabens also contain thermal waters [17].

Sampling. The samples of mineral water were directly collected from natural mineral water springs. Water samples were collected using polyethylene bottles. Two samples were collected at every sampling point. Conservation with concentration HNO_3 was provided, within 6 h after sampling.

The geographical distribution of localities, where mineral waters were investigated is shown in Fig. 1.

Fig. 1 Geographical distribution of localities, where mineral waters have been investigated

Analytical procedures. ^{226}Ra activity concentration in mineral water was determined by a combination of solvent extraction and LSC based on a radon extraction by a scintillation cocktail (Rotiscint eco from Roth/Karlsruhe with 20 % of cyclohexane) immiscible with water, followed by a measurement with a liquid scintillation counter of the alpha-emission of radon and of its decay products in radioactive equilibrium with the parent. The measurements were carried out using low-level Liquid Scintillation Counting (LSC) in a PACKARD TRI-CARB 2770 TR/SL LSC system, with pulse shape discrimination (PSD) for separation and acquisition of α and β events into different multi-channel analyzers [13].

^{210}Pb activity concentration A radiochemical procedure, based on extraction chromatography with a crown ether extractant, was applied to determination of ^{210}Pb . Pb carrier and ^{209}Po tracer were added in order to correct for chemical recoveries after separation and purification. Purified Pb was isolated as PbSO_4 , dried and weighed to determine the chemical recovery. It was then dissolved in 2 mL of 0.05 M HCl and mixed with 18 mL of Packard InstaGel XF liquid scintillation cocktail in a 20 mL glass vial and measured after in-

growth of its daughter radionuclide ^{210}Bi , using a Packard Tri-Carb 2770 TR/SL liquid scintillation analyzer [2, 11].

Uranium concentration. It was determined on the basis of the ability of uranium melted with sodium fluoride to produce light under UV illumination. The intensity of induced luminescence is proportional to the concentration of Uranium [13, 18].

Annual effective dose. The annual effective dose was estimated using the hypothesis that any person from the public would drink at most 730 l/year from any given spring.

RESULTS AND DISCUSSION

The measurement of natural radioactivity in mineral waters from certain most frequently used sources in Rhodope Mountains region, Southern Bulgaria was carried out. The results of the activity concentrations of ^{226}Ra and ^{210}Pb and nat. U concentration in mineral waters are given in Table 1. For comparison with the ^{226}Ra and ^{210}Pb data, the nat. U concentrations in mg/L are converted in specific activity in mBq/L.

Table 1. Activity concentrations of ^{226}Ra and ^{210}Pb , nat. U concentrations and the corresponding activity concentration in certain Bulgarian mineral waters

Sample code	Mineral source	^{226}Ra [mBq/L]	^{210}Pb [mBq/L]	nat. U [$\mu\text{g/L}$]	nat. U [mBq/l]
MW1	Devin, well number 3	51 ± 4	1.9 ± 0.3	≤ 3	≤ 75
MW2	Devin, well number 5	74 ± 5	2.3 ± 0.4	≤ 3	≤ 75
MW3	Mihalkovo, well 1aVP	168 ± 18	36 ± 3	6.9 ± 0.8	173 ± 20
MW4	Mihalkovo, well VKP	152 ± 15	31 ± 5	6.1 ± 0.7	153 ± 18
MW5	Lenovo	58 ± 6	5.1 ± 0.7	3.4 ± 0.4	79 ± 10
MW6	Rakitovo well number 5	18 ± 2	4.8 ± 0.5	4.1 ± 0.4	103 ± 10
MW7	Rakitovo, well number 7	23 ± 3	≤ 1.9	≤ 3	≤ 75
MW8	Rakitovo, well number 9a	28 ± 3	≤ 1.8	≤ 3	≤ 75
MW9	Bratsigovo, well number 2	89 ± 7	71 ± 9	10 ± 1	250 ± 25
MW10	Bratsigovo, well number 5	13 ± 2	104 ± 8	21 ± 2.3	525 ± 58
MW11	Velingrad, well number 2	76 ± 7	37 ± 5	3.1 ± 0.4	78 ± 10
MW12	Velingrad, well number 5	81 ± 6	27 ± 2	3.1 ± 0.4	78 ± 10
MW13	Velingrad, spring Kremachna Banya	109 ± 10	42 ± 5	≤ 3	≤ 75
MW14	Velingrad, spring Veliova Banya	155 ± 19	38 ± 4	≤ 3	≤ 75
MW15	Velingrad, spring Vlasa	41 ± 5	13 ± 2	≤ 3	≤ 75
MW16	Velingrad, Syarna Banya well	75 ± 9	5.1 ± 0.8	≤ 3	≤ 75
MW17	Belovo	65 ± 5	71 ± 9	4.1 ± 0.4	103 ± 10

The results show that activity concentrations of ^{226}Ra in water samples were in range 13 – 168 mBq/L. The highest values were found in mineral waters from Mihalkovo, Smolyan region (MW3) and Velingrad, spring Veliova Banya (MW14). This may be explained by the high TDS (total dissolved solids) value of water from Mihalkovo. It was found that the higher the TDS value of water, the higher the activity concentration of the measured ^{226}Ra [16]. The reason for the high value of a concentration of the mineral water from Velingrad, spring Veliova Banya is due to the presence of granitic formations in this area. The content of ^{226}Ra in all samples is below the recommended level of 500 mBq/L [8].

According to data of this study, ^{226}Ra activity concentrations in investigated mineral waters are comparable with data reported for other countries [5, 19, 20]. Activity concentration values of ^{226}Ra published earlier [13, 18] for some mineral waters in Bulgaria are similar to the results obtained here.

The mean ^{226}Ra activity concentration in mineral waters from Rhodope Mountains region was 80 mBq/L. This value is higher than the published values for the concentration of ^{226}Ra in mineral waters in other countries such as Italy, Sweden, Portugal, France and Germany, and lower than the obtained values for Romania and Argentina [5, 21].

^{210}Pb activity concentrations ranged from ≤ 1.8 to 104 mBq/L (Table 1). The values obtained for mineral waters varied over a wide range. In general, the measured activity levels were lower than the limit established for water for human consumption (200 mBq/L). The obtained ^{210}Pb activity concentrations do not differ significantly from the data reported for other countries [2, 22, 23].

A high degree of radioactive disequilibrium was noted between ^{226}Ra and its daughter product ^{210}Pb present in some water samples. Disequilibrium between ^{210}Pb and its parent isotope ^{226}Ra is due to diffusion of the intermediate gaseous isotope ^{222}Rn . If diffusion of ^{222}Rn is negligible, ^{210}Pb is likely to be in equilibrium with its progenitor ^{226}Ra because the other four intermediates have all shorter half-life of only seconds or minutes. The loss of ^{210}Pb might be caused by chemical precipitation, biological activity or other factors [11, 22].

In the investigated waters the concentration of nat. U varied from ≤ 3 to 21 $\mu\text{g/L}$ (Table 1). The highest concentration of uranium was detected in samples from Bratsigovo (MW10). It can be seen from Table 1 that concentrations of nat. U measured in 8 water samples (47% of the total water samples)

are below the minimum detectable concentration (MDC). The uranium concentration in the water depends on many different factors like the type of the geological formation of the region, the nature and concentration of other chemical constituents in the water and chemical processes, such as ion exchange, sorption and precipitation [24, 25]. In oxidizing conditions, uranium forms soluble stable complexes, e.g. carbonates, phosphates, etc. and can move for long distances, while in reducing conditions (absence of air) uranium precipitates, forming concentrated secondary deposits [23].

The results obtained in this study are relatively low compared to some literature data [26] and well below the WHO guidance levels of 30 $\mu\text{g/L}$, respectively [7].

Annual effective doses due to ingestion of the mineral waters were calculated to evaluate the radiological hazard to the local population. Internal exposure, arising from the intake of ^{226}Ra , ^{210}Pb and nat. U through the ingestion of mineral waters, was evaluated taking into account the radium, lead and uranium content of waters and the yearly average consumption of 730 liters (around 2 l/day). The expected total annual effective doses were calculated for adults (older than 17 years). For radionuclide concentrations below the detection limit, the minimum detectable activity (MDA) was considered. The dose reference level of 100 $\mu\text{Sv/y}$ has been used for comparison with our results. The annual effective dose was calculated separately from ^{226}Ra , ^{210}Pb and U content. Table 2 shows a comparative overview of values of the annual effective dose from the already mentioned radionuclides in mineral waters.

In the prevailing number mineral water samples, the annual effective dose originating from ^{226}Ra is higher than that from ^{210}Pb and nat. U. Based on these results, we can conclude that the main contribution to the total annual effective dose is due to ^{226}Ra .

Estimated total annual effective doses for mineral waters are presented in Fig. 2.

We established that the total annual effective doses received by the population as a result of ingestion of mineral water were in the range 8.12 – 58.1 $\mu\text{Sv/y}$. It is evident that the calculated doses vary over wide range, but all values are below the recommended level of reference dose level of 100 μSv for one year's consumption of mineral waters. Consequently, the health hazards related to ^{226}Ra , ^{210}Pb and nat. U in mineral waters are expected to be negligible.

Table 2. Expected annual effective doses for investigated mineral waters, calculated separately on the basis of ^{226}Ra , ^{210}Pb and U activity measurements

Sample code	Mineral source	Annual effective doses [$\mu\text{Sv/y}$]		
		^{226}Ra	^{210}Pb	nat. U
MW1	Devin, well number 3	10.4	0.96	2.46
MW2	Devin, well number 5	15.1	1.16	2.46
MW3	Mihalkovo, well 1aVP	34.3	18.1	5.68
MW4	Mihalkovo, well VKP	31.1	15.6	5.03
MW5	Lenovo	11.9	2.57	2.60
MW6	Rakitovo well number 5	3.68	2.42	3.38
MW7	Rakitovo, well number 7	4.70	0.96	2.46
MW8	Rakitovo, well number 9a	5.72	0.91	2.46
MW9	Bratsigovo, well number 2	18.2	35.8	8.21
MW10	Bratsigovo, well number 5	2.66	52.4	17.2
MW11	Velingrad, well number 2	15.5	18.6	2.56
MW12	Velingrad, well number 5	16.6	13.6	2.56
MW13	Velingrad, Kremachna Banya spring	22.3	21.2	2.46
MW14	Velingrad, Veliova Banya spring	31.7	19.1	2.46
MW15	Velingrad, Vlasa well	8.38	6.55	2.46
MW16	Velingrad, Siarna Banya well	15.3	2.57	2.46
MW17	Belovo	13.3	35.8	3.38
Mean		15.3	14.6	4.13
Range		2.66-34.3	0.91-52.4	2.46-17.2

Fig. 2 Expected cumulative annual effective doses due to ^{226}Ra , ^{210}Pb and U activities for the waters under investigation

CONCLUSIONS

Investigations of natural radionuclides in mineral water sources in Southern Bulgaria were carried out. The results show that the activity concentrations of ^{226}Ra and ^{210}Pb ranged from 13 to 168 mBq/L and from ≤ 1.8 to 104 mBq/L, respectively and do not exceed the guidance levels recommended by WHO and Bulgarian regulation. The concentrations of nat. U in all samples were below the limit of 30 $\mu\text{g/L}$.

We did not observe a correlation between the activity concentrations of ^{226}Ra and ^{210}Pb in the investigated waters.

The expected total annual effective doses for adults were calculated from the measured ^{226}Ra , ^{210}Pb and U activities. The results showed that the annual effective dose of ingestion of these mineral water samples is lower than the recommended value of 100 $\mu\text{Sv/y}$.

According to the results of our study, it is evident that the investigated mineral waters are suitable for human consumption without any radiological hazard. The obtained new results are used to assess the temporary radiation status of the investigated waters, as well as, the related dose to the population.

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