VII. RADIATION ECOLOGY

GROSS ALPHA AND BETA ACTIVITY IN MINERAL WATER FROM SOUTHWEST BULGARIA

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Abstract. The natural radioactivity levels of mineral water from certain most frequently used sources in Southwest Bulgaria were determined. The mineral water was investigated with regard to gross alpha and beta activity to determine whether the activity concentrations are below levels at which no further action is required. The measurements were made by Liquid Scintillation Spectrometer. The gross alpha and beta activity varied from ≤ 0.003 Bq/L to 0.671 Bq/L and from 0.054 and to 0.375 Bq/L, respectively. The values obtained in this study were compared with data reported by other authors in different countries and with reference values accepted for drinking water.
The annual effective doses were calculated for all investigated waters for adult inhabitants assuming yearly consumption of 730 litres. The results show that the annual effective dose of ingestion of these water samples excluding one, are below the individual dose criterion of 100 μSv/y according to the World Health Organization recommendations.

Keywords: gross alpha activity, gross beta activity, mineral water, annual effective dose.

INTRODUCTION

The mineral water is one of the most precious natural resources in Bulgaria. There are more than 240 deposits of mineral springs having different temperature, chemical composition and radioactivity [1, 2]. Most of them are low mineralized and are suitable for daily consumption, balneology (drinking balneology), recreation and others. Therefore, their quality must be carefully and systematically controlled. Natural mineral water may contain a number of alpha (^{238}U, ^{234}U, ^{232}Th, ^{226}Ra and ^{210}Po) and beta emitters (^{228}Ra and ^{210}Pb) from the natural decay series of uranium and thorium, the primordial isotope ^{40}K and artificial isotopes, such as ^{137}Cs and ^{90}Sr in various concentrations. The high-energy alpha and beta emitters have significant radiotoxicity. 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 many different factors like the type of the geological formation and hydrogeology of the region, the geochemistry of uranium and thorium, and the nature and concentration of other chemical constituents in the water. Radionuclides can penetrate into ground water by emanation processes, leaching and dissolution of mineral substances from rocks and minerals, which form the aquifer or infiltration of rain water [5, 6].

Gross alpha and beta activities are very useful parameters for the preliminary screening of water. Their content gives essential information about the natural radionuclides in water and their corresponding health hazards associated with water consumption [7]. The daily consumption of mineral water with high content of natural radionuclides may have a significant contribution to the internal natural radiation exposure of the population and the rules or directives made and maintained by national and international authorities become more and more important [8]. 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. The guidance levels defined on the Ordinance for total annual effective dose, gross alpha and gross beta activity in drinking water are 0.1 mSv/y, 0.1 Bq/l and 1.0 Bq/l, respectively [9].

The lack of any big research facility in Bulgaria like a high-flux nuclear reactor or an ion accelerator makes impossible the application of more precise and advanced methods for water quality investigations. Therefore, in the present study we apply the most accurate conventional analytic methods. The National cyclotron centre at the Institute for Nuclear research and Nuclear Energy – Bulgarian Academy of Sciences (INRNE – BAS), based on the TR-24 cyclotron, [10] will open a new horizon for such research, as well as for various applications in the field of nuclear physics and radiochemistry.

The aim of this study is to obtain a representative estimate of the concentration levels of gross alpha and beta activity in mineral water from certain most frequently used sources in Southwest Bulgaria in

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order to assess the radiation status of this water as well as the related dose to the population.

MATERIAL AND METHODS

Sampling. A total of fifteen water samples were collected from frequently used mineral water springs in Southwest Bulgaria. Water samples were collected using polyethylene bottles. 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. Determination of gross alpha and beta activity in mineral waters was performed by liquid scintillation counting. Acidified water samples to a volume of 1 000 mL were evaporated until dryness (avoiding boiling) and dissolved with 5 mL of 0.1M nitric acid. The solutions were then transferred to a 20 mL polyethylene scintillation vial, and mixed with 15 mL of Packard ULTIMA GOLD AB scintillation cocktail for measurement [15, 16]. 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. The counting efficiency was evaluated with ^{241}Am and ^{90}Sr/ ^{90}Y standards [11-13].

Portable pH meter Sension 156 was used for the measurements of physicochemical parameters: chemical reaction (pH) and total dissolved solids (TDS).

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 gross alpha and beta activity in mineral water from certain most frequently used sources in Southwest Bulgaria was carried out. The activity concentrations for gross alpha and beta are shown in Fig. 2.

In Table 1 some statistical results obtained for the studied mineral water are presented.

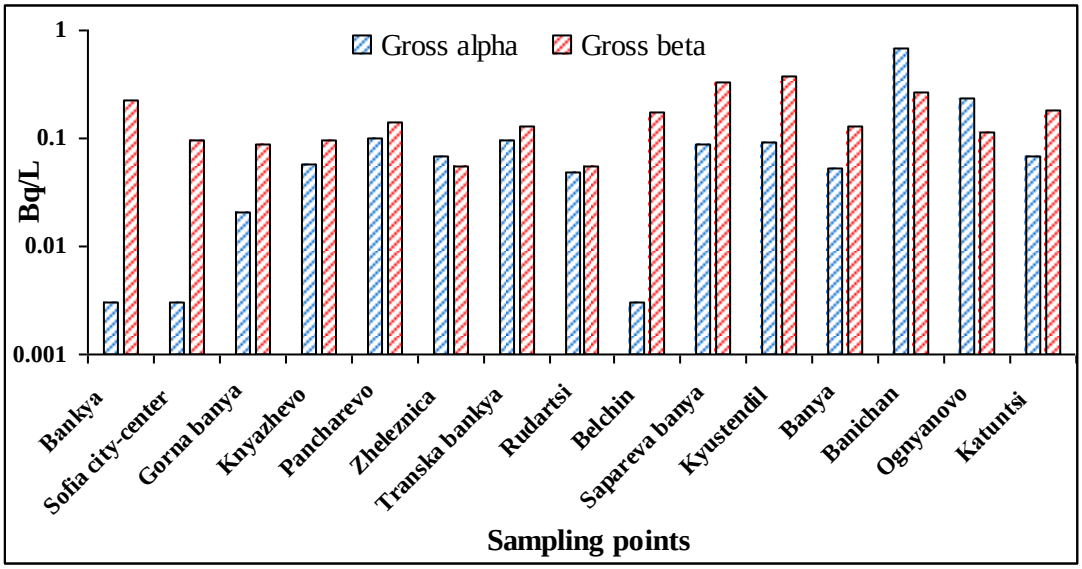

Fig. 2. Gross alpha and beta activity in certain mineral water springs from Southwest Bulgaria

Table 1. Statistical results for gross alpha and beta activity [Bq/L], physico-chemical parameters and total annual effective dose [μSv/y] for the mineral water under investigation

Parameter	Min	Max	Mean	Median	Standard deviation
Gross alpha activity [Bq/L]	< 0.003	0.671	0.107	0.067	0.166
Gross beta activity [Bq/L]	0.054	0.375	0.164	0.129	0.097
pH	7.34	9.74	8.83	9.07	0.75
TDS [mg/L]	132	636	342	295	151
Total annual effective dose [μSv/y]	10.2	164	38.4	23.8	37.9

The values of gross alpha activity ranged from ≤ 0.003 Bq/L to 0.671 Bq/L with average value of 0.107 Bq/L and median of 0.067 Bq/L. The large standard deviation indicates that gross alpha activity in mineral water greatly vary. The difference between the mean and the median values was large, indicating that most samples had a low gross alpha activity. In 4 from the 15 analyzed samples, gross alpha activity is lower than minimum detectable activity (MDA) of 0.003 Bq/L. The lowest values were registered in samples from Bankya, Sofia city-center and Belchin. The maximum gross alpha activity of 0.671 Bq/L was determined in the sample from Banichan. We established that, for 13% of mineral water studied, the gross alpha activities are over the recommended level (0.1 Bq/L). The main reason is the different local, geological and hydrological conditions of the original sources of the waters investigated. Southern Bulgaria is characterized by a wide variety of relief and geological structures. There are mountains with granite and metamorphic rocks, lignites, uranium-rich minerals of uranite, carnotite, and phosphate deposits [14]. Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks [15].

The results obtained for gross beta activity in mineral water varied between 0.054 and 0.375 Bq/L, with an average value of 0.172 Bq/L and a median value of 0.129 Bq/L. The highest value of gross beta activity of 0.375 Bq/L was found in the sample from Kyustendil. The data obtained for mineral water ranged over a wide range. In general, the measured activity levels were lower than the limit established for water for human consumption (1 Bq/L). For 69% of the analyzed samples, the gross alpha activity is lower than the gross beta activity, trend also reported by other authors [16].

The results obtained in this study were compared with similar measurements in other places in the world (Table 2).

Table 2. Comparison of gross alpha and beta activity in mineral water from different countries

Country	Gross alpha [Bq/L]	Gross beta [Bq/L]	References
Hungary	< 0.008–3.34	0.035–2.6	17
Italy	< 0.004–0.28	< 0.025–0.93	18
Turkey	< 0.007–0.90	0.013–0.85	19
Vietnam	0.005–0.12	0.001–0.19	20
Brazil	< 0.01–0.11	< 0.05–1.60	21
Bulgaria	≤ 0.003–0.67	0.054–0.38	This study

The measured gross alpha activity in our investigation is higher than those observed in Italy, Brazil and Vietnam and lower than those observed in Hungary and Turkey. Our results for the gross beta activity in mineral water are generally lower than those given in the literature. The values of gross alpha and beta activity published earlier [12,13] for some mineral water sources in Bulgaria are similar to the results obtained here.

The pH of the water samples varied from 7.3 to 9.7, with an average value of 8.85 according to the mineral content of water. This is affected by the type of Earth layers, from which the water is drawn. Water, percolating through a hard rock like granite, will have a higher mineral content, and will thus tend to be more acidic.

The average value of TDS was 328 mg/L and it ranged from 132 to 636 mg/L. 87 % of the of mineral water samples are slightly mineralized (TDS ≤ 500 mg/L) and suitable for everyday use. TDS is the term used to describe the inorganic salts and small amount of organic matter present in water. The principal constituents are usually calcium, magnesium, sodium, and potassium cations, chloride, sulfate and nitrate anions.

The annual effective dose associated with radiation exposure through the ingestion of water was estimated to assess the health risk for the population. In order to calculate the annual effective dose, we considered that alpha gross activity is due to ^{226}Ra because more than 50% of the annual effective dose is due to the its contribution and beta gross activity due to ^{228}Ra and ^{210}Pb, which are radionuclides with the highest effective conversion factors CF [16, 22]. The expected total annual effective dose was calculated for adults (older than 17 years). The dose reference level of 100 μSv/y has been used for comparison with our results. In Table 1, the minimum, maximum, median and standard deviation values of total annual effective dose are given. We established that the total annual effective doses received by the population as a result of ingestion of mineral water were in the range 10.2 – 164 μSv/y, with an average value of 38.4 μSv/y. It is evident that the calculated doses varied over wide range, but all values, excluding one are below the recommended level of reference dose level of 100 μSv for one year's consumption of mineral water.

The data obtained from gross alpha and beta measurements can provide basic information for consumers and competent authorities, regarding the internal exposure risk due to water intake.

CONCLUSIONS

Investigations of gross alpha and beta activity in the mineral water samples collected from certain most frequently used sources in Southwest Bulgaria were carried out to check the compliance with national and international regulation and obtain the data on the natural radioactivity levels of investigated mineral water.

The content of gross alpha activity in all samples, excluding two mineral water samples, is below the recommended level of 0.1 Bq/L by WHO and Bulgarian regulation. For all selected water sources, the values of gross beta activity do not exceed the limit of 1 Bq/L.

The results showed that the annual effective dose of ingestion of the investigated mineral water

in all samples, excluding one are lower than the recommended value of 100 μSv/y as reported by WHO.

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 water samples, as well as, the related dose to the population.

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