Radiological Monitoring of Waters in Bulgaria

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Abstract

The radiological indicators of drinking and groundwaters from different regions of Bulgaria were tested and analyzed in 2024 in connection with the regular monitoring of waters in Bulgaria. Higher uranium content than the norm regulated in the national legislation was found in separate samples from the districts of Burgas, Plovdiv, and Smolyan.

Increased alpha activity was determined in drinking and groundwaters from the districts of Burgas, Pernik, Yambol, Plovdiv, Haskovo, and Smolyan. This is probably due to both the specific hydro-geological structure of the aquifers and the presence of sites related to the underground uranium mining carried out in the past in the country. The analysis found that in most cases the higher alpha activity is due to the contribution of uranium, which exhibits low radiotoxicity and after calculating the indicative dose, which did not exceed 0.1 mSv per year, the waters were determined as safe for consumption from radiological point of view. In cases where uranium concentration in the studied samples was not sufficient to justify the higher alpha activity, the waters were analyzed for the presence of polonium-210, due to the high radiotoxicity of the radionuclide. Only in one of the waters from the Smolyan region, it was higher than the secondary concentration determined in the regulations, which is probably due to the fact that the water was from a new drilling and contained sediment particles, since polonium-210 usually settles in sediments.

In general, the monitoring of waters from different regions of Bulgaria carried out in 2024 found that in the majority of the samples the radiological indicators did not exceed the parametric values set out in the national legislation.

Key words: radioactivity of waters, radon, uranium, tritium, gross alpha and beta activity

Introduction

Radiological monitoring of water is based on determining the content of natural and technogenic radionuclides in them. These indicators are dynamic and depend on a number of factors such as erosion processes, amount of precipitation, dissolution processes from aquifers, etc. In addition, radioactive contamination of soils, and hence of waters, can occur as

a result of anthropogenic activities such as uranium mining (Yordanova et al., 2015), mining of minerals, metals (Hristozova et al., 2022) coal (Tsolova et al., 2022) and electricity production through coal-fired thermal power plants (Lazarova et al., 2020), as well as in the production and agricultural use of phosphate fertilizers (Ghosh et al., 2008).

In the national legislation, radiological criteria for water quality in Bulgaria are regulated by Regulation 9 on the quality of water intended for drinking and domestic purposes, Regulation 1 on the study, use and protection of groundwater and the Regulation on the requirements for bottled natural mineral, table and spring waters intended for drinking purposes.

The paper describes the study and analysis of the radiological indicators of 130 drinking and 170 groundwaters from different regions in Bulgaria in connection with the regular monitoring of waters in Bulgaria.

Materials and Methods

Content of radon, tritium, natural uranium (< 0.03 mg/l), gross alpha (< 0.1 Bq/l), and beta activity (< 1 Bq/l), in drinking waters were measured according to Regulation 9 from different regions of Bulgaria and the risk of additional dose exposure from the consumption of the tested waters was assessed by determining the total indicative dose (< 0.1 mSv/year).

Groundwaters was analyzed under Regulation 1 for radiological indicators gross alpha (< 0.5 Bq/l) and beta activity (< 1 Bq/l), and natural uranium (< 0.06 mg/l) and according to Regulation 9, in cases where the ground water sources were intended for drinking and domestic purposes.

Testing Laboratory of Radioecology and Radioisotope Research at ISSAPP "Nikola Poushkarov" is accredited for determining content of radioactive elements in waters, soils, plants and foods under BDS EN ISO/IEC 17025:2018.

Specific activity of ^{222}Rn was measured gamma-spectrometrically by the line of ^{214}Bi at 609 kV. Multi-channel analyzer DSA 1000 (CANBERRA) with pure Ge detector, 30% efficiency and 1.8 keV resolution was used.

Determination of tritium activity in water samples was carried out according to the method for liquid scintillation counting according to developed and validated laboratory procedure (Naidenov et al., 2010).

Gross alpha and beta activity were determined according to ISO standard procedures BDS EN ISO 9696/2017 and BDS EN ISO 9697/2019 respectively based on the preparation of planchets with dry residue of aliquot part of water samples measured on low-background gas proportional alpha/beta counter. The results are validated by annual participation in interlaboratory comparisons, proficiency test schemes and regular internal quality control.

Results and Discussion

• *Drinking waters*

- Radon

Despite the fact that the radiation risk from exposure to radon through the consumption of drinking water is much lower than that caused by inhalation of indoor radon,

European legislation and the corresponding Bulgarian Regulation 9 introduced the parametric value of 100 Bq/l for this radionuclide in drinking waters. When the set parametric value exceeds 100 Bq/l and is up to 1000 Bq/l, water supply organizations and state health control authorities conduct jointly study and assess whether and what corrective actions should be taken, taking into account the risk to human health, without putting the drinking and domestic water supply at risk. Corrective actions are taken in all cases when the radon concentration exceeds 1000 Bq/l, in order to ensure radiation protection.

In 2024, over 110 waters were tested for radon content. Only in two of the waters (from Blagoevgrad and Smolyan districts), radon activity was above the parametric value of 100 Bq/l. In the first one, this is probably due to the fact that the water is underground from a new drilling. A control study will be conducted on it in 2025. The second water is from the village of Banite, Smolyan district, which is spa resort and is located in the Rhodope Mountains. The higher radon content in it is probably due to the water aquifers in the Rhodope Mountains characterized by higher content of uranium and radium and radon respectively as a product of the radioactive decay of uranium-radium family enters the water through processes of erosion and dissolution from rocks and minerals formed in the aquifer. In the remaining waters, the concentration of the radionuclide was under the control level of 100 Bq/l, and in most of the samples, it was below the minimum detectable activity of 10 Bq/l.

- *Tritium*

Natural levels of ^{3}H in environmental samples, mainly the result of the interaction of cosmic rays with the atmosphere, increased because of the nuclear tests conducted from 1945 to 1963. However, the activity of ^{3}H in the atmosphere has decreased (half-life = 12.33 years) and today its levels are comparable to those before the nuclear tests.

Another source of tritium in the environment are nuclear power plants (NPPs), with tritium contamination around NPPs being found mostly in nearby surface waters.

As far as legislation is concerned, the parametric value for the tritium content in drinking water is 100 Bq/l, and elevated levels of tritium may be due to the presence of other artificial radionuclides, and if the tritium concentration exceeds its parametric value, an analysis for the presence of other artificial radionuclides is required.

116 water samples were analyzed for tritium content. The activity of the radionuclide in all of them was lower than the parametric value, and in the majority of the samples, the values were below the minimum detectable activity of 10 Bq/l. These results showed that no technogenic pollution of the tested waters was observed.

- *Natural uranium, gross alpha and beta activity*

In addition to being radioactive, natural uranium is also toxic as a chemical element and its ingestion can lead to internal human organs damage. The maximum permissible concentration of uranium in drinking water is 0.03 mg/l.

Elevated levels of natural uranium were found in separate samples from the districts of Burgas, Plovdiv, and Smolyan. Figure 1 presents the results for the number of samples with higher levels of natural uranium from the total number of samples tested during the year.

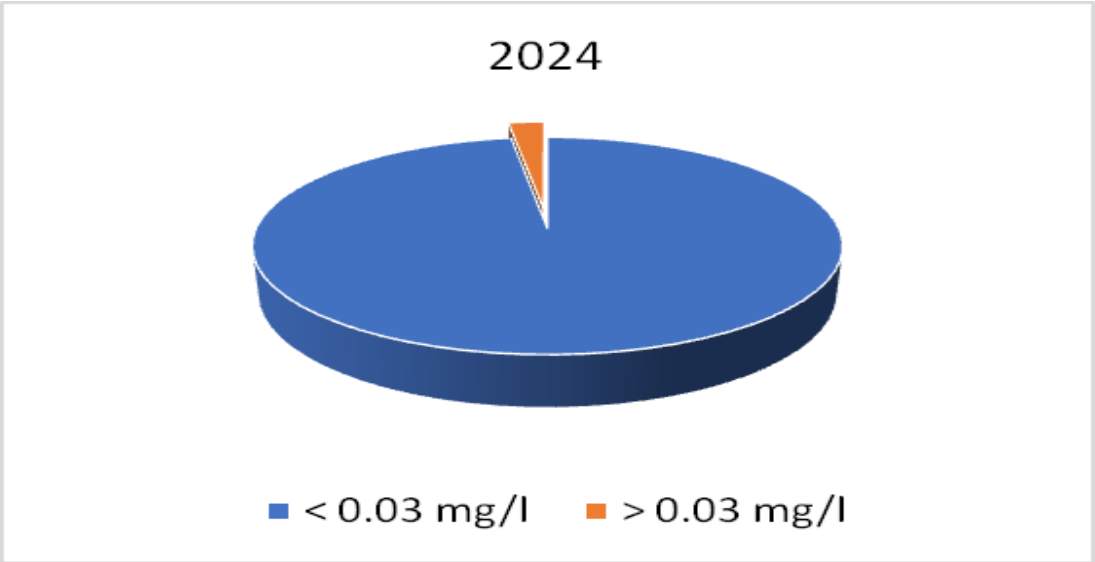


Figure 1. *Content of natural uranium in drinking waters in 2024*

Alpha activity in waters is mainly due to the content of the isotopes of uranium (^{234}U, ^{235}U and ^{238}U) and ^{226}Ra, which are alpha emitters. Beta activity is usually mainly due to the content of ^{40}K and the short-lived beta emitters of the ^{238}U family. In all samples, beta activity was below the control level of 1 Bq/l. When gross alpha and beta activity are lower than the control levels, it is assumed that the indicative dose is below 0.1 mSv per year and the consumption of these waters does not lead to additional dose exposure. When gross alpha activity exceeds the parametric control value of 0.1 Bq/l, additional studies have to be carried out to identify specific alpha emitters.

The results of the studies of gross alpha activity of drinking waters are presented in Figure 2.

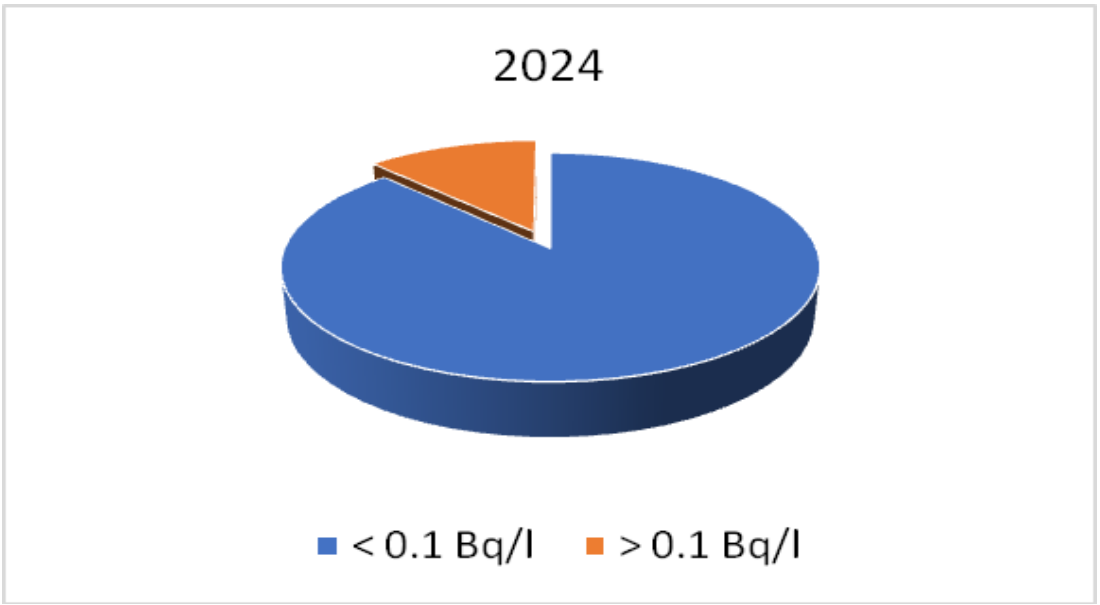


Figure 2. *Gross alpha activity in drinking waters in 2024*

The analysis showed the increased alpha activity in the majority of the samples studied is due to the contribution of natural uranium, which exhibits low radiotoxicity. The indicative

dose calculated with the corresponding radionuclide concentration and the dose coefficient for uranium was below 0.1 mSv per year, indicating that the water is not dangerous for human health from radiological point of view. Radium, which is more radiotoxic, has relatively low solubility in water and does not form soluble complexes. Uranium isotopes (^{234}U and ^{238}U) are generally the most abundant radionuclides in waters, since under oxidizing conditions uranium forms soluble stable complexes (e.g. carbonates) and can migrate over long distances (Nuccetelli at al., 2012)

Drinking waters with alpha activity higher than the control level were from the districts of Burgas, Pernik, Yambol, Plovdiv and Haskovo. The increased concentration of natural uranium and gross alpha activity in these samples was probably due to both the specific hydro-geological structure of the aquifers and the presence of sites related to the closed uranium mining from natural uranium deposits.

In cases where the concentration of uranium in the studied samples was not sufficient to justify the higher alpha activity, the waters were analyzed for the presence of polonium-210 according to the new methodology introduced into the practice of the Laboratory in 2023 under BDS EN ISO 13161:2020.

Polonium-210, which is also an alpha emitter from the uranium-238 family, is distinguished by the highest radiotoxicity and respectively, the secondary norm for it in Regulation 9 is the lowest – 0.1 Bq/l. In all tested drinking waters, the content of Po-210 was below the minimum detectable activity. Respectively, the indicative dose did not exceed 0.1 mSv/year, which indicated that the consumption of these waters would not lead to additional dose exposure.

• ***Groundwaters***

The groundwaters were analysed for the content of natural uranium (< 0.06 mg/l), gross alpha (< 0.5 Bq/l) and beta activity (<1 Bq/l) according to Regulation 1 and Regulation 9, in cases where the water sources were intended for drinking and domestic purposes. In all tested waters, no values for uranium above the norm of 0.06 mg/l and for alpha activity above 0.5 Bq/l were recorded according to Regulation 1, but since most of the samples required calculation of the indicative dose, they were tested in accordance with the norms and parametric values of the radiological indicators according to Regulation 9.

The results of the tests performed are given in Figures 3 and 4.

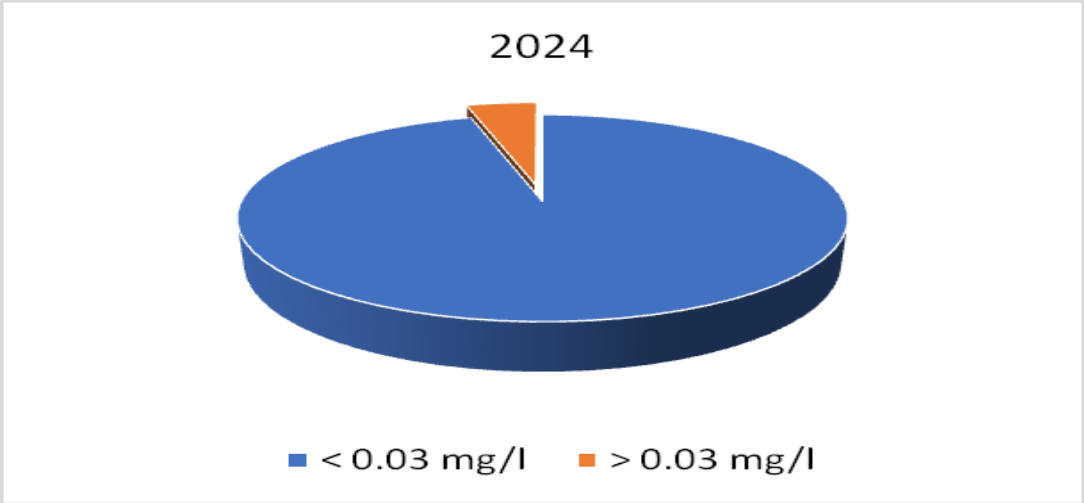


Figure 3. *Content of uranium in underground waters in 2024*

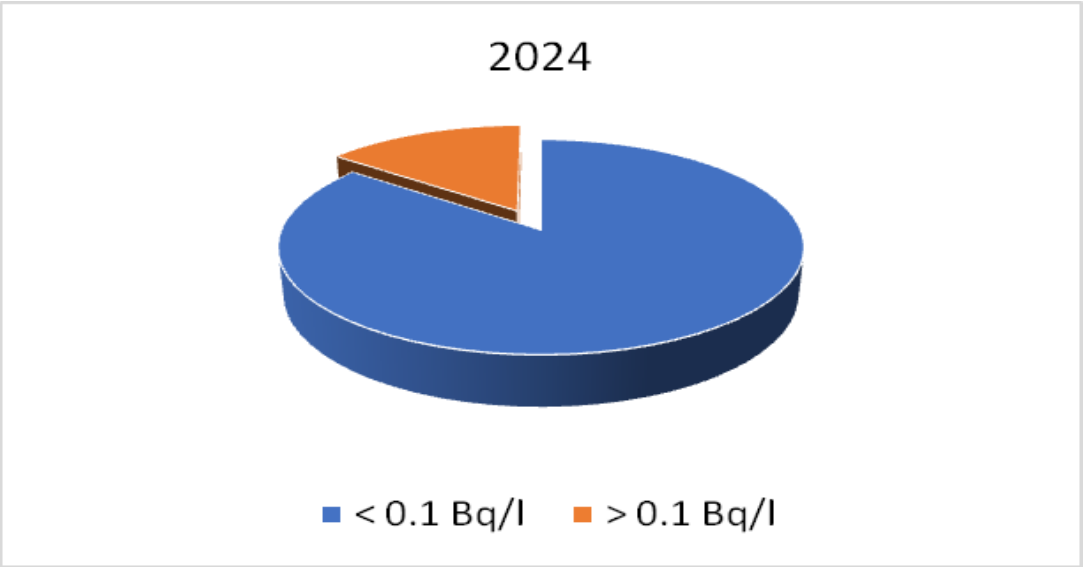


Figure 4. *Gross alpha activity in drinking waters in 2024*

- *Uranium content in groundwaters*

Of all the groundwater samples tested, natural uranium content of over 0.03 mg/l were determined in 5 samples from Plovdiv and Smolyan districts and did not meet the requirements of Regulation 9. Accordingly, they could not be used as drinking waters due to the high toxicity of uranium as a chemical element.

- *Gross alpha and beta activity*

Gross alpha activity was over 0.1 Bq/l in 25 of the tested groundwater samples. After the analysis, it was found that in most of them the increased alpha activity was due to the natural uranium content in them and they are not dangerous from a radiological point of view. The calculated total indicative dose was below 0.1 mSv, which indicates that the water can be used for drinking and domestic purposes.

One of the waters from Nedelino was tested for Po-210 content, since the alpha activity in it exceeded the natural uranium content. The content of Po-210 in it was 0.13±0.03 Bq/l, making it hazardous for consumption from radiological point of view because it exceeded the secondary concentration for the radionuclide of 0.1 Bq/l. The sample was from new groundwater drilling. The higher content of the radionuclide in it is probably due to the presence of sediment particles in the water, since polonium-210 has low solubility in water and usually settles in sediments (Persson and Holm, 2011). Thus, its presence in surface waters is not usual, but it could be present in higher concentrations in groundwater. A control study of the respective water from the same water source will be carried out in 2025.

Conclusion

The monitoring of waters from different regions of Bulgaria carried out in 2024, showed that in the majority of the samples the radiological indicators did not exceed the parametric values set out in the national legislation.

In the majority of the samples, the increased alpha activity was due to the uranium content. The indicative dose was calculated. It was below the control level, which indicated

that these waters were not hazardous to human health from radiological point of view.

The waters in which uranium was not sufficient to justify the increased alpha activity were tested for polonium-210 content due to the high radiotoxicity of the radionuclide. In all waters except one sample from a new drilling, the activity of the radionuclide did not exceed the secondary concentration under Regulation 9. The content of the radionuclide in the water has to be monitored regularly through periodic tests.

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