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INVESTIGATION OF ^{238}U, ^{234}U AND ^{210}PO CONTENT IN SELECTED BULGARIAN DRINKING WATER

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Abstract. The radioactivity of selected sources of drinking water in Southern Bulgaria was investigated using ^{238}U, ^{234}U and ^{210}Po activity measurements and dose calculation, respectively. The activities of ^{238}U, ^{234}U and ^{210}Po varied from 226 to 826 mBq/L, 274 to 1623 mBq/L and < 0.6 to 25.5 mBq/L, respectively, being lower then derived concentrations for radioactivity in water intended for human consumption of the considered radionuclides, given in EC Directive 2013/51/EURATOM. In some drinking waters the mass concentration of natural uranium exceeded the set maximum chemical concentration level of 0.030 mg/L.

A radioactive disequilibrium between ^{234}U and ^{238}U in water was detected.

Based on the radionuclide activity concentrations total annual effective ingestion doses for adults, as well as contribution of each particular radionuclide to the total doses, were assessed and discussed. The lowest contribution to the annual effective doses was found for ^{210}Po and the highest for ^{234}U. The results show that the annual effective doses of residents are below the reference level of 100 μSv/y according to the recommendations of the World Health Organization. The obtained new results are used to assess the radiation status of the investigated water.

Keywords: drinking water; natural radioactivity; ^{238}U; ^{234}U; ^{210}Po; annual effective dose

Introduction

Drinking water contains a number of naturally occurring radionuclides from both uranium-radium (^{238}U – ^{226}Ra) and thorium (^{232}Th) decay chains, potassium (^{40}K), tritium (^{3}H), radon (^{222}Rn) and its daughter products polonium (^{210}Po) and lead (^{210}Pb) and artificial radionuclides (^{137}Cs, ^{134}Cs, ^{90}Sr, etc.) coming from the fallout from atmospheric nuclear weapons testing and the accidents at nuclear reactors (Altıkulaç et al. 2015).

The determination of natural radioactivity in drinking 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 (Joksić et al. 2007; Rožmarić et al. 2012; Zehringer 2019).

Of all the radionuclides present in drinking water, the radionuclides of uranium, radium, polonium, lead, and short-lived ^{222}Rn are responsible for the major fraction of the internal dose received by humans from the naturally occurring radionuclides (Outola et al. 2008).

Recently national and EU regulations decreased the drinking water norms with the aim to strengthen consumer's security concerning drinking water quality (WHO, 2011). World Health Organization (WHO) guidelines for drinking water and Directive 2013/51/EURATOM set parametric values of 0.1 mSv/y for annual effective dose, 2.8 Bq/L for ^{234}U, and 3 Bq/L for ^{238}U. National legislation was fully harmonized with EU Directives (Ordinance No 9, 2001, last corrected 2018). In the same national legislation, the maximum permitted level for uranium based on its chemical toxicity is set as 0.030 mg/L which means that if in the samples the ^{238}U activity is above 0.38 Bq/L the limit value is exceeded.

According to national and international legislation, when drinking water has a gross alpha activity above the recommended screening level of 0.1 Bq/L, monitoring of specific radionuclides is required. The radionuclides to be measured shall be defined taking into account all relevant information about likely sources of radioactivity. When the concentrations may lead to indicative dose above 0.1 mSv or the uranium mass concentration is above set maximum concentration value, remedial actions should be taken to improve the quality of the water to a level which complies with the requirements for the protection of human health (Figure 1).

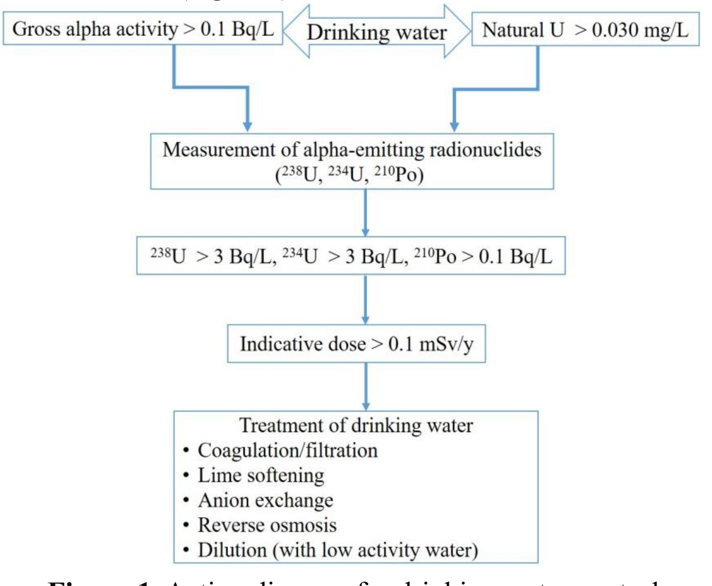

Figure 1. Action diagram for drinking water control

The determination of the radionuclides of uranium as well as ^{210}Po in water is of primary importance to human health due to the high toxicity and radiotoxicity of uranium and polonium (Zehringer 2019).

Uranium is heavy naturally occurring radioactive element. It is widespread in the Earth's crust. Uranium is harmful to human health, especially hazardous for kidneys due to high radioactivity (alpha particle emission due to radioactive decay) and first of all its toxic chemical properties (Rožmarić et al. 2012; Sekudewicz and et al. 2019; Zapecza & Szabo 1986). It has three alpha emitting radionuclides: ^{234}U, ^{235}U and ^{238}U with a different atomic mass that have different distribution and half-lives. More than 99 percent of uranium occurring in nature is ^{238}U.

Usually, uranium isotopes (^{238}U and ^{234}U) are the most abundant radionuclides in water because of the great mobility and the long half-life (4.47×10^9 years for ^{238}U and 2.45×10^5 years for ^{234}U), which makes these radionuclides long-term hazardous (Nuhanović et al. 2015). The ^{238}U isotope and the less frequent ^{234}U occur naturally in the (IV) oxidation state in granites and various other minerals such as pitchblende, monazite and lignite sands and phosphates of uranium, which are components of various types of rocks (Abojassim & Mohammed 2017). As a result of the rocks weathering, the uranium oxidizes to the (VI) oxidation state through which it can be dissolved in water (Sekudewicz & Gąsiorowski 2019).

Human activity, such as mining, coal combustion, fertilizer production, inappropriately stored radioactive waste and other activities, can contribute to elevated content of uranium isotopes in drinking water (Nuhanović et al. 2015; Outola et al. 2008).

Occasionally, larger quantities of ^{234}U than ^{238}U are observed in water, and this phenomenon may be related to rock weathering. $^{234}U/^{238}U$ activity ratio in natural water is an important indicator of the origin of the uranium in the studied sample. Commonly observed disequilibrium between ^{234}U and ^{238}U in water is a result of nuclear recoil effects and extensive rock/water interactions (Nuhanović et al. 2015; Sekudewicz & Gąsiorowski 2019).

Particular attention should be paid to the naturally occurring ^{210}Po, as one of the most radiotoxic substances to humans. ^{210}Po is a radionuclide of the ^{238}U decay series, with half-life of 138.4 d (Ahmed et. al. 2018). Therefore, it is important to study the concentrations of this radionuclide in drinking water.

Measurements of natural radioactivity in drinking water have been performed in many parts of the world, mostly for assessment of the doses and risk resulting from water consumption (Beyermann et al. 2010; Ortega et al. 1996; Outola et al. 2008; Radenković et al. 2015; Rožmarić et al. 2012).

In Bulgaria, few data are available concerning the occurrence of natural radionuclides in drinking water. The only data for natural radioactivity levels in drinking water already published concerns natural uranium, ^{226}Ra and ^{210}Pb, as well as gross alpha and gross beta activity (Kamenova-Totzeva et al. 2015; Slavchev et al. 2020). However, activity concentration levels of uranium and polonium isotopes in drinking water in Bulgaria and the radiological impacts of the ingestion of this water have not been reported previously.

The aim of this study is to determine the activity concentrations of ^{238}U, ^{234}U and ^{210}Po, as well as $^{234}U/^{238}U$ activity ratio in drinking water collected from selected settlements located in Southern Bulgaria. In order to evaluate potential health hazards, doses due to ingestion of this water were estimated to assess the contribution of these radionuclides to public exposure from natural radioactivity.

Material and Methods

Sampling

Drinking water samples were directly taken from the public water supplies of the town of Parvomay and the villages of Byala reka, Bryagovo, Dragoynovo, Padarsko, Babek, Bolyarino, Karadzalovo, Borets, Vinitsa, Zelenikovo and Vladimirovo situated in the Upper Thracian Lowland, Southern Bulgaria. The locations of the 14 sampling points are shown in Figure 2.

The samples were collected in 10 L polypropylene bottles. The sampling was done from faucets which are high enough to put a bottle underneath, without contacting the mouth of the container with the faucet.

Before sampling the tap is turned on to a steady stream for 2 – 3 minutes to remove any stagnant water in the plumbing network and the bottle and cap are rinse three times with sample water. The bottle should be filled to within one to two centimeters from the top. Then, the drinking water samples were acidified with nitric acid, to prevent losses by sorption of the studied radionuclides onto the vessel walls.

Figure 2. Geographical locations of the sampling points

Radiochemical methods

Analyses of natural radionuclides ^{238}U, ^{234}U and ^{210}Po were performed by radiochemical procedures summarized in Table 1 and described in more detail below.

Table 1. Summary of methods used for drinking water analysis

Radionuclide	Tracer	Analytical method	V (L)	MDA (mBq/L)
^{238}U, ^{234}U	^{232}U	$Fe(OH)_3$ precipitation, extraction chromatography, microcoprecipitation or electrodeposition, alpha spectrometry	2	1
^{210}Po	^{209}Po	extraction chromatography, spontaneous deposition, alpha spectrometry	1	0.4

Determination of uranium isotopes

The activity concentrations of ^{238}U and ^{234}U were separated from other radionuclides using extraction chromatography and alpha spectrometry. The radiochemical procedure adopted for ^{238}U and ^{234}U determination is described in more detail by Rožmarić et al. (2012). A 2 L water sample was used for the analysis, which was acidified with concentrated HCl and H_2O_2 to pH of approximately 1. Fe (III) (Fe^{3+}) carrier as $FeCl_3$ for uranium co-precipitation and ^{232}U tracer for determination of recovery were added.

Radionuclides were concentrated from the water sample as $Fe(OH)_3$ co-precipitation at pH 9 – 10 using NH_4OH. The precipitate was filtered through a 0.45 µm polypropylene filter, rinsed with water (to pH= 7) and dissolved in 3 M HNO_3. The pure uranium fraction was obtained by use of Eichrom UTEVA resin which was preconditioned in 3 M HNO_3. After the interfering elements were removed by washing the column with 3 M HNO_3, 9 M HCl and 0.5 M $H_2C_2O_4$/5 M HCl, uranium radionuclides were eluted with 0.01 M HCl. The source for alpha spectrometric measurement was prepared by microcoprecipitation with NdF_3 and filtration on a polypropylene disk (0.1 µm). In some cases, electrodeposition was used to produce an alpha source with better spectrometric quality.

^{210}Po determination

^{210}Po was determined by alpha spectrometry after radiochemical separation of polonium from the other alpha radionuclides present in water. The preparation of the ^{210}Po sample was performed using 1L samples. A radiochemical procedure, based on extraction chromatography with a crown ether extractant, was applied to separate simultaneously the lead and polonium fractions. Pb carrier and ^{209}Po tracer were added in order to correct for chemical recoveries and sample was evaporated and dissolved in 2M HCl acid. Separation of polonium from lead was performed on Eichrom Sr spec resin preconditioned with 2 M HCl. ^{210}Po was eluted from the column with 6 M HNO_3 and obtained polonium fraction were evaporated to dryness. Polonium source for alpha spectrometric measurement was prepared by self-deposition on a copper disk from 2M HCl solution (pH=1) with addition of 100 ml of distilled water. Spontaneous deposition of polonium was carried out at 50° C for 4 h. The disk was rinsed with water and ethanol, and dried at room temperature (Rožmarić et al. 2012).

Instrument

Uranium and polonium radionuclides were identified and measured by means of high resolution ORTEC Octete Alpha Spectrometric system equipped with 8 chambers and ion implanted type ULTRA-SATM detectors with 300 mm^2 active surface. The energy resolution (FWHM) for ^{241}Am, 5.486 MeV line is 20 keV for 4 cm source to detector distance for all detectors. Energy calibration, as well as, efficiency calibration for source geometry is done by mixed radionuclides standard containing ^{238}U, ^{234}U, ^{239}Pu and ^{241}Am with known activity, and for geometry of electroplated sources. The efficiency calibration is performed with ^{241}Am Amersham standard (Dimova et al. 2003). Typical alpha spectrum of the uranium isotopes and ^{210}Po is shown in Figure 3.

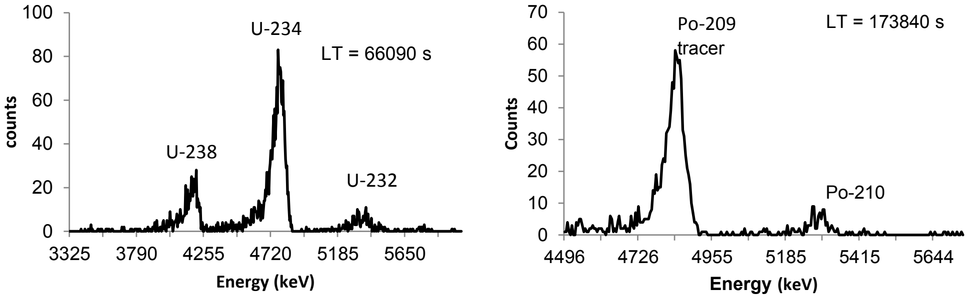

Figure 3. Alpha spectrum of the drinking water samples, U (left) and Po (right panel)

Annual effective dose

For the total annual effective dose calculation, activity concentrations of the radionuclides in Bq/L, dose coefficients of 0.045, 0.049, and 1.2 μSv/Bq for ^{238}U, ^{234}U ^{210}Po, respectively and annual water consumption of 730 L for adults were used (ICRP 1996; Rožmarić et al. 2012; WHO 2011).

Results and discussion

Activity concentrations

The activity concentrations of ^{238}U, ^{234}U and ^{210}Po in drinking water samples collected from selected sources in Southern Bulgaria are presented in Figures 4 and 5.

Table 2 shows the range of results, arithmetic mean (AM) and geometric mean (GM) of ^{238}U, ^{234}U and ^{210}Po activity concentrations in drinking water samples. Figure 3 presents activity concentrations of ^{238}U and ^{234}U in the investigated waters. All drinking water samples have gross alpha activity above recommended screening level of 0.1 Bq/L. Therefore, continuous monitoring of alpha radionuclides in those waters is required.

As can be seen from Table 2 and Figure 4 the concentrations of ^{238}U and ^{234}U in drinking waters varied from 226 to 826 mBq/L with an average of 477 mBq/L and 274 to 1623 mBq/L with an average of 718 mBq/L, respectively. The AM is slightly larger than the GM.

Table 2. Activity concentrations (mBq/L) of ^{238}U, ^{234}U and ^{210}Po in certain Bulgarian drinking water from Southern Bulgaria

Activity concentrations (mBq/L)	^{238}U	^{234}U	^{210}Po
Range	226–826	274–1623	< 0.6–25.5
Arithmetic mean	477	718	5.4
Geometric mean	440	626	3.8

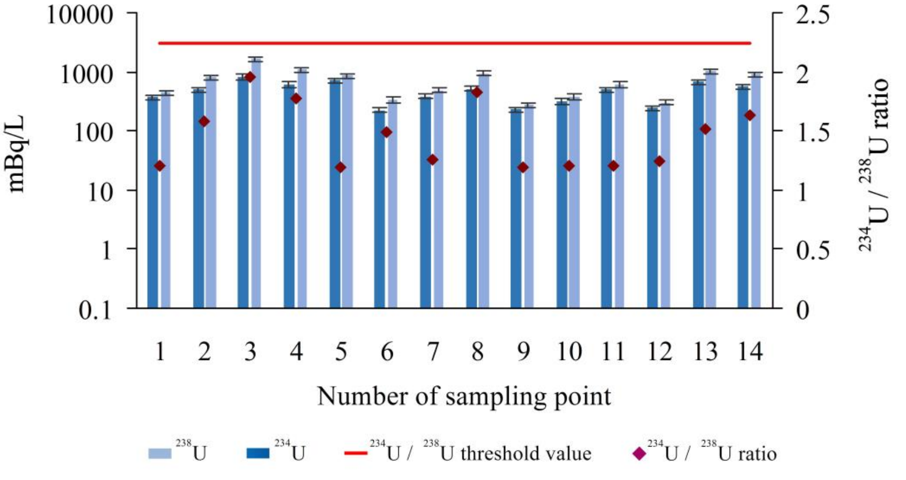

Figure 4. Activity concentration of ^{238}U and ^{234}U and $^{234}U/^{238}U$ ratio in drinking water samples.
The threshold activity value of ^{238}U as 3 000 mBq/L is given.

The highest activity concentration of uranium isotopes was detected in sample 3 (Bryagovo). In some water samples the calculated mass concentration of ^{238}U exceeded the maximum value of 0.030 mg/L. The study area is located in the Upper Thracian Uranium Ore Region, where uranium mining was carried out in the past. The ore region is characterized by exogenous uranium deposits formed in the Bartonian-Quaternary complex compound by sedimentary and less volcano-sedimentary rocks. Different granitoids and high grade metamorphic rocks are the sources of uranium. The ore bodies are localized within sandstone aquifer, less in aleurolite, tuff-sandstone, rare in clay, within reducing or neutral conditions (Popov et al. 2016). The uranium activity 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 (Ortega et al., 1996). The hydro-chemical composition of water varies from hydrocarbonate-sodium to hydrocarbonate-sodium-calcium, sulfate-hydrocarbonate-calcium, rarely sulfate-sodium and chloride-sulfate-sodium depending on the lithological and landscape conditions (Popov et al., 2015). In oxidizing conditions, uranium forms soluble stable complexes, e.g. uranyl-carbonate, uranyl-sulfate and hydroxyl-uranyl complexes, which are highly mobile and define the migration and concentration in exogenous conditions, while in reducing conditions (absence of air) uranium precipitates, forming concentrated secondary deposits (Outola et al. 2008; Popov et al. 2016; Zapecza & Szabo 1986).

It is observed that the activity concentration of ^{234}U in drinking water samples is higher than the activity concentration of ^{238}U. A state of radioactive disequilibrium between ^{234}U and ^{238}U in water was detected. Usually the $^{234}U/^{238}U$ activity ratio in natural water is in the range of 0.5-1.2, but it can reach 30 in extreme cases (Nuhanović et al. 2015). In this study $^{234}U/^{238}U$ activity ratios were found to vary between 1.19 and 1.96 (Figure 4). It is established that radionuclides produced by alpha decay are more readily driven out from rock because alpha decay causes the atom to recoil, which reduces atom stability in the lattice, i.e. ^{234}U activity concentration in water is higher than, or equal to, that of the parent ^{238}U because alpha decay-induced recoil can expel ^{234}U from rock (Zapecza & Szabo 1986).

The results obtained in this study are compared with the reported values from other countries in the world (Table 3).

Listed values show the extremely wide activity concentration range of ^{238}U and ^{234}U from < 0.4 to 3 934 mBq/L and from < 0.4 to 964 mBq/L, respectively. Natural radionuclide concentrations in drinking water can be very different due to geographical and geological factors. The measured ^{238}U activity concentrations are higher than those observed in Italy, Greece, Belgium and Poland and lower than those observed in Germany and India. Results obtained for ^{234}U are higher than those given in the literature.

Table 3. Comparison of the activity concentrations of ^{238}U and ^{234}U in drinking waters from different countries

Country	^{238}U [mBq/L]	^{234}U [mBq/L]	References
Germany	8.6 – 3 934	–	Beyermann et al. 2010
Italy	< 0.4 – 161	< 0.4 – 211	Forte et al. 2007
Greece	4.08 – 95.32	3.88 – 160.13	Samaropoulos et al. 2012
India	30.7 – 3 848	–	Shenoy et al. 2012
Belgium	0.3 – 16.8	0.4 – 22.7	Vasile et al. 2016
Poland	1.0 – 725	2.4 – 964	Walencik et al. 2010
Bulgaria	226 – 826	274 – 1623	This study

The results of the measured ^{210}Po activity in drinking water samples are shown in Table 2 and Figure 5. The values obtained are in the range < 0.6 – 25.5 mBq/L with an average of 5.41 mBq/L and can be regarded as the lowest among all analyzed radionuclides.

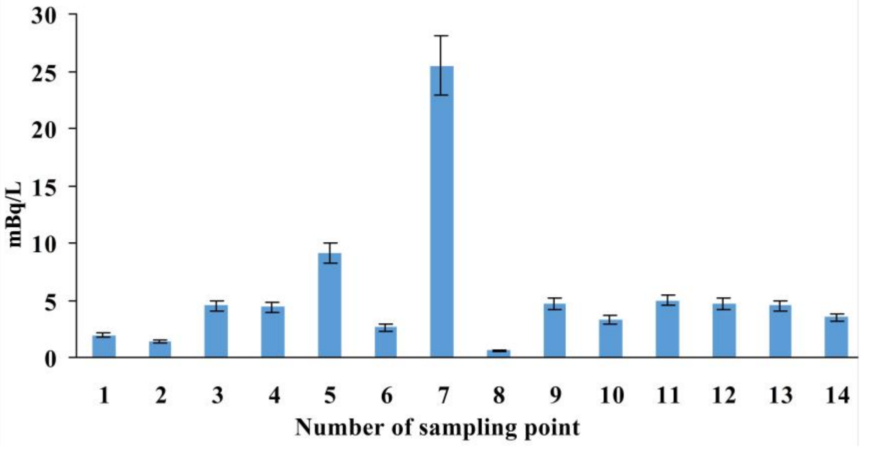

Figure 5. Activity concentration of ^{210}Po in drinking water samples

The content of polonium and its parents in groundwater are related to the quantity and seasonal diversity of precipitation, the infiltration time and the type of rocks through which the water flows etc. In groundwater, the concentration of ^{210}Po is usually less than 40 mBq/L (Sekudewicz & Gąsiorowski 2019).

The results obtained in this study are in agreement with other investigations (Ahmed et al. 2018, Sekudewicz & Gąsiorowski 2019, Kavitha et al. 2017, Walsh et al. 2014). For example, activity concentrations of ^{210}Po up to 114.2 mBq/L was measured in tap water samples in Western Australia (Walsh et al. 2014).

Annual effective doses

In order to estimate the radiological hazard to members of the public from ingested ^{238}U, ^{234}U and ^{210}Po, the expected total annual effective doses were calculated on the basis of the results for activity concentration of these radionuclides. The dose reference level of 100 µSv/y has been used for comparison with our results. The results of the evaluation of the total annual effective doses are shown in Figure 6.

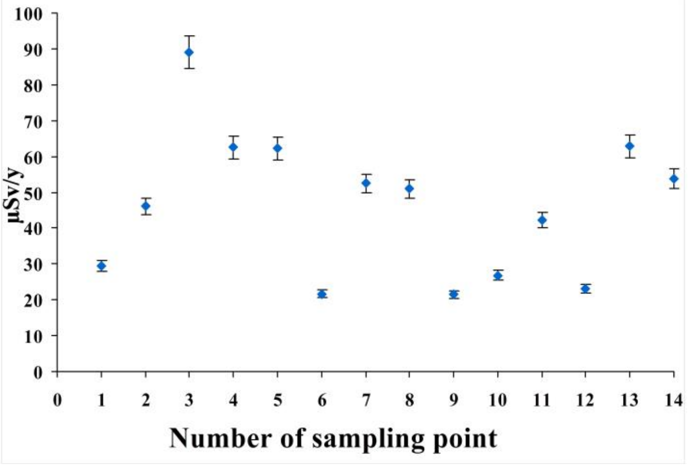

Figure 6. Expected cumulative annual effective doses due to ^{238}U, ^{234}U and ^{210}Po activity concentrations for the waters under investigation

The total annual effective doses received by the population as a result of ingestion of drinking water was in the range 23.1 – 89.1 µSv/y. The average annual effective dose estimated for all samples was 46.1 µSv/y. It is evident that the calculated doses vary over wide range, but all values are below the reference level of 100 µSv for one year's consumption of drinking water. Consequently, the health hazards related to ^{238}U, ^{234}U and ^{210}Po in drinking water are expected to be negligible. The values of the total annual effective doses for adult received from the consumption of analyzed drinking water are in good agreement with the results obtained by us in previous studies (Slavchev et al. 2019) for drinking water in the Central and Southern regions of Bulgaria and those obtained by (Kamenova-Totzeva et al. 2015) for drinking water samples from Southwest Bulgaria (0.0175 µSv/y – 95.5 µSv/y).

Contribution of each radionuclide to the total annual dose is given in Figure 7.

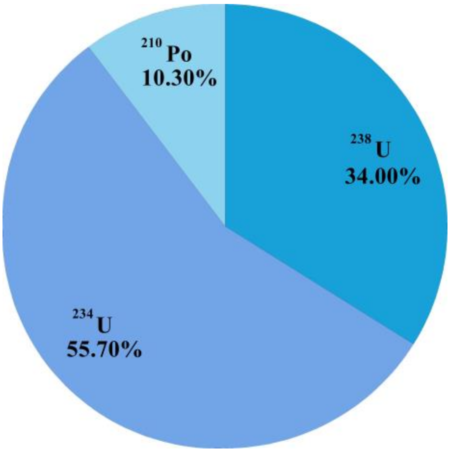

Figure 7. Contribution of each analyzed radionuclide to the total annual effective ingestion dose in drinking water

As seen from the obtained results, it is obvious that the highest contribution to the total effective dose in investigated water comes from ^{234}U (up to 56 %). ^{238}U dose contribution is around 34 % for drinking water. The lowest contribution was found for ^{210}Po (up to 10 %).

Based on the results obtained in this study, we can conclude that the main contribution to the formation of the total annual effective dose is due to ^{234}U.

Conclusions

Investigations of the radioactivity levels of ^{238}U, ^{234}U and ^{210}Po in selected drinking water sources from Southern Bulgaria were carried out.

The values show that the highest activity concentrations were due to ^{234}U. The results are comparable to results from other studies around the world.

A state of radioactive disequilibrium between ^{234}U and ^{238}U in water was detected.

The mass concentrations of the uranium exceeded the guideline value set by WHO, 2011, based on uranium chemical toxicity in drinking water of 0.03 mg/L in some of the analyzed samples.

The total annual effective ingestion doses for adults were assessed from the activity concentrations measured in this study. In all cases, the estimated doses were below the WHO recommended guidance level of 100 μSv/y for the consumption of drinking water.

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

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