

RADIUM CONTENT IN BULGARIAN MINERAL WATERS

by

Rositza M. KAMENOVA-TOTZEVA*, **Alexandar V. TOTZEV**, and **Radostina M. KOTOVA**

National Center of Radiobiology and Radiation Protection, Sofia, Bulgaria

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This study presents the results of ^{226}Ra content in 104 Bulgarian mineral waters as well as the investigated correlations between the physicochemical parameters, geological structure, and specific activity of ^{226}Ra . The results show that the ^{226}Ra activities range from $<30\text{ mBqL}^{-1}$ to 4900 mBqL^{-1} with a mean value of 156 mBqL^{-1} . The highest concentrations of ^{226}Ra were measured in mineral waters originating from the carbonate strata, as the dominant geological structure. Thus it is well connected with the estimated moderate positive correlation between Ca^{2+} , Mg^{2+} , and Ra^{2+} cations in the water. The estimated annual dose shows large dispersion of values ranging from $6\text{ }\mu\text{Sv}$ to $1000\text{ }\mu\text{Sv}$ with a mean value of $32\text{ }\mu\text{Sv}$. The results demonstrate the need for further investigation of every mineral water, but greater attention should be given to the mineral waters with low pH and high content of Ca^{2+} and Mg^{2+} .

Key words: ^{226}Ra , mineral water, correlation, dose assessment

INTRODUCTION

Radium belongs to the group of radionuclides with high radiotoxicity. The ^{226}Ra follows the calcium metabolism in the human body and is deposited partly in the bone tissue. The ^{226}Ra could cause bone, cranial and nasal tumors [1].

^{226}Ra is essentially soluble in water. It enters groundwater by the dissolution of materials in water layers, removal of rock or soil surfaces and is released from minerals by radioactive decay [2]. In its passage within the Earth's crust, mineral waters come into contact with large surfaces of radioactive eruptive rocks such as granites, quartz porphyry and basalt which contains radium [3]. The activity concentrations of radium depend on the geological structure of the aquifer and distribution of the parent element in the rock matrix [3-5].

The solubility of the parent element and the solubility of the radionuclide itself influence the concentrations of the radionuclides in mineral waters [6].

In addition to the geological structure of the aquifer there are many other factors, such as physicochemical parameters controlling the content of ^{226}Ra water.

Radium is highly soluble in waters with low pH and its concentrations frequently are connected with the total dissolved salts and sulfates. It is reported that a positive correlation between the minerals content and the level of natural radionuclides in water exists [7, 8]. The correlation between the radium content in the water and contents of chlorides, calcium, potassium, and bromine was observed in the literature [9, 10-13]. Those

components become typical elements in the salt composition of groundwater. On the other hand, it was reported that the content of radium does not depend on the dissolved uranium in the water [14, 15].

It is well known that the content of radium in the water depends on the chemical composition of the water more than the content of radium in the rocks of the aquifer [16, 17].

The issue of the relationship between the total dissolved salts in the water, water conductivity, pH and other physicochemical parameters is not yet fully understood.

The specific activities of ^{226}Ra in the mineral waters vary widely. Due to the high radiotoxicity of radium isotopes, especially ^{226}Ra numerous studies were conducted and evaluations in terms of implementation of radiation protection for the population were made.

With regard to public health, the purpose of this study was to determine the specific activities of ^{226}Ra in Bulgarian mineral waters as well as to estimate annual effective doses due to the ingestion of ^{226}Ra .

The occurrence of elevated concentrations of ^{226}Ra and the correlation between ^{226}Ra concentrations and physicochemical parameters in mineral water samples were also presented.

MATERIALS AND METHODS

Bulgaria is divided into three major hydrogeological units – Lower Danube Basin, Intermediate Region and Rila-Rhodopes Region. The Lower Danube Artézian Basin encompasses the Moesian Plate,

* Corresponding author; e-mail: r.totzeva@ncrrp.org

Balkan Foreland tectonic zone and northern part of the Balkan tectonic zone. The Intermediate Region covers the southern part of the Balkan zone and Sredna gora tectonic zone. The Rila-Rhodopes Region comprises the Rila-Rhodopes massif. Three types of reservoirs are found in the country – stratified, fractured and mixed (water from a fractured reservoir is secondarily accumulated in a younger sediment reservoir).

The Moesian Plate has a Caledonian-Hercynian basement and a cover of Upper Paleozoic and Mesozoic sediments. The main geothermal reservoirs in the platform area are situated in the carbonate strata of the Malm-Valanginian, Middle Triassic and Upper Devonian age.

The Sredna gora zone is a heterogeneous hydrothermal region where unstratified (fault-fractured), stratified and mixed hydrothermal systems are present. Hydrothermal circulation takes place in the fractured massif of granite and metamorphic rocks and in the Upper Cretaceous volcano – sedimentary deposits. Thermal reservoirs are formed also in many postorogenic Neogene – Quaternary grabens filled up with terrigenous deposits.

The western part of the Rila-Rhodopes massif is built of Precambrian metamorphic and granite rocks, fractured by a dense set of seismically active faults. Unstratified hydrothermal systems with thermal waters of low salinity, meteoric origin and of the highest temperature up to 100 °C are found in this area. The metamorphic basin contains some large bodies of marble that act as hydrothermal reservoirs. Permeable terrigenous-clastic rocks in the deep Neogene and Paleogene grabens also contain thermal waters [18].

Bulgaria has a total of more than 225 hydrothermal sources of mineral waters with total capacity of 5000 Ls⁻¹. In southern Bulgaria there are 148 and in northern Bulgaria there are 77. According to their temperature, mineral waters are cold (to 37 °C), warm (37 °C to 60 °C) and hot (over 60 °C). The majority of them have a temperature between 37 °C and 50 °C.

Sampling was done for the period of 5 years (2011-2016). This study presents the results for 104 Bulgarian mineral waters, which show a different degree of mineralization and different hydrogeological units. On the basis of the access to data for physicochemical parameters, for correlation purposes 56 mineral waters, representing different cases were valued.

Sampling was done in polyethylene bottles. Conservation with concentrated HNO₃ was provided in the laboratory, within 6 to 8 hours after sampling. Filtering of the samples was not done, because they were very clear and did not show any type of suspended matter.

The ²²⁶Ra determination was performed by measurement of its daughter ²²²Rn after equilibrium in an ionizing chamber using Alpha Guard PQ 2000 PRO. The procedure includes several radiochemical steps.

Radium was deposited from the solution as radium-barium sulphate. The sulphate was transformed into carbonate after melting with sodium carbonate. Radium carbonate was transformed into chloride after digesting with HCl into a radon bubbler. The radon bubblers were purged and sealed, flooded with pressurized aged air for 20 min and stored for 20 to 30 days. After these emanation processes, the growing of ²²²Rn during the storage time which was equal to ²²⁶Ra activity was measured. The percentage of established equilibrium was used for the calculating of the final results. The efficiency of the ionizing chamber was 73.44 %. The uncertainty of measurement was 4.76 % ($k = 2$). The minimum detectable activity (MDA) was 0.001 BqL⁻¹. The volume of the chamber was 600 ml and the volume of the system was 913 mL.

Physicochemical analyses were done by the Accredited Laboratory from the National Specialized Hospital for Physical Therapy and Rehabilitation. The results have been taken from the State Certificates of mineral waters, published in the public registry of the official web page of the Ministry of Health [19].

The results for the total dissolved solids (TDS), anions (F⁻, Cl⁻, SO₄²⁻), cations (Na⁺, Ca²⁺, Mg²⁺), pH and temperature (T) of the investigated mineral waters were correlated with radium content by the statistical software tool IBM SPSS.

Annual effective doses (ID) from ingestion of mineral water with ²²⁶Ra were estimated using the most conservative hypothesis that one person is drinking the same mineral water the entire time on the basis of 2 L per day consumption for adults.

RESULTS AND DISCUSSION

Specific activity of ²²⁶Ra in mineral waters

The results for ²²⁶Ra specific activity of 104 analyzed Bulgarian mineral waters are presented in tab. 1 and fig. 1. The results represent approximately 46 % of all mineral waters in Bulgaria. The distribution of the results is shown in fig. 2.

The results show that the ²²⁶Ra activities range from <30 mBqL⁻¹ (MDA) to 4900 mBqL⁻¹ with a mean value of 156 mBqL⁻¹ ($n = 104$). The value of the kurtosis (100) means that the distribution is leptokurtic, *i. e.* there are higher frequencies of values near the mean. If we exclude the maximum value from the assessment the mean value becomes 107 mBqL⁻¹ with Std. Deviation 62 mBqL⁻¹ ($n = 103$). When comparing the radium specific activities values observed in the present study, excluding the highest value, with

Table 1. Statistical results for ²²⁶Ra content in mBqL⁻¹

Min.	Max.	Mean	Std. dev.	Median	Kurtosis		
Statist.	Statist.	Statist.	Std. err.	Statist.	Statist.	Std. err.	
30	4900	155.8	46.5	474	94	100.19	.47

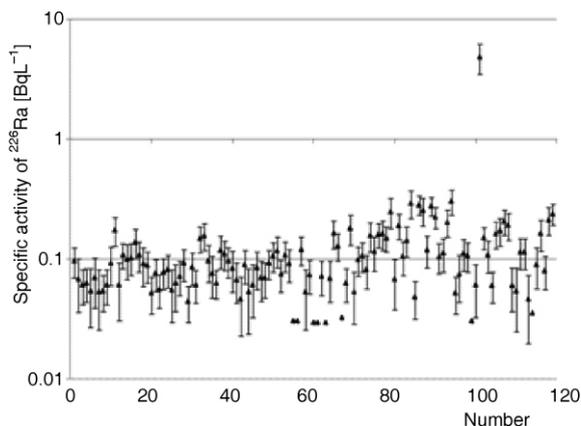


Figure 1. Specific activity of ^{226}Ra in Bulgarian mineral waters in $[\text{BqL}^{-1}]$

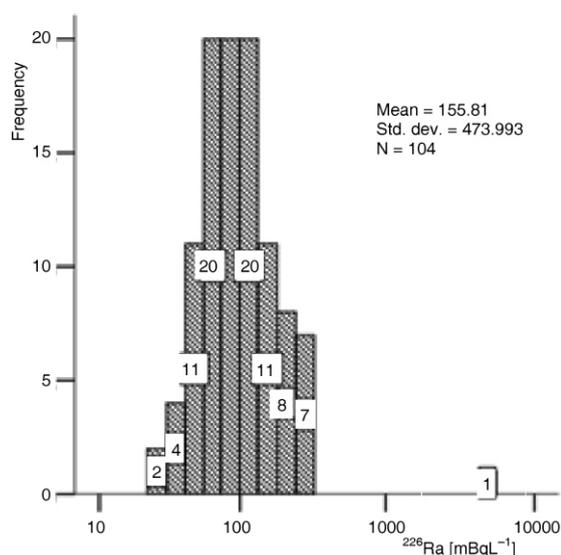


Figure 2. Histogram with the distribution of the ^{226}Ra content in Bulgarian mineral waters

those reported for other countries (tab. 2), it was observed that these results are of the same order of magnitude as those reported for Croatia, Greece, France, Tunisia, Morocco and Hungary and higher than those determined in waters from Serbia, Romania, Spain, Turkey and Poland.

Table 2. The comparison of ^{226}Ra content in mineral waters from different countries

Country	^{226}Ra $[\text{mBqL}^{-1}]$	Literature source
Croatia	87-6200	[20]
Tunisia	34-3900	[21]
France	588-2287	[22]
Morocco	9-3696	[23]
Greece	300-5000	[24]
Hungary	4,3-2940	[25]
Romania	60-480	[4]
Serbia	10-530	[26]
Spain	2-1370	[27]
Turkey	100-1200	[12]
Poland	4-758	[13]
Bulgaria	30-4900	This study

Table 3. Statistical results for ^{226}Ra content in mBqL^{-1}

Geothermal reservoir (dominant structure)	Min. statist.	Max. statist.	Mean	
			Statist.	Std. err.
Sediments	30	113	64	34
Granite and metamorphic rocks	30	152	78	31
Carbonate strata	30	240	127	63
Effusive rocks	47	110	73	23
Gneisses	76	177	112	36

Geological structure and radium content

For the assessment of possible correlation between the geological structure of the water table and radium content 56 mineral waters were investigated. The distribution of the number of the investigated waters considering the geothermal reservoirs origin is: to the platform area situated in the carbonate strata – 19, to the platform area situated in the fractured massif of granite and metamorphic rocks – 16, to the platform area situated in the sediments – 5, to the platform area built mainly by the effusive rocks – 9, and to the platform area built mainly by gneisses – 7.

The obtained results for ^{226}Ra content in different geological reservoirs are presented in tab. 3.

The highest concentrations of ^{226}Ra were measured in the mineral waters originating from the carbonate strata, as the dominant geological structure. The lowest concentrations of ^{226}Ra were registered in mineral waters originating from geothermal reservoirs built in effusive rocks and sediments as the dominant geological structure. On the basis of the standard error of the mean values, there are obvious statistically high differences within the groups. The limited number of investigated samples in the groups and the fact that we investigated only the dominant material in the geological structure could give only the pilot information about a possible correlation between ^{226}Ra content and the geological structure.

Correlations between physicochemical parameters and ^{226}Ra

The chemical compositions and specific activity of ^{226}Ra in the mineral waters analyzed (tab. 4) indicate a broad variation for all parameters. The TDS ranged from a few mgL^{-1} to 10892 mgL^{-1} . The results of pH, temperature, anions and cations demonstrate the difference of the mineral waters composition.

The concentration of ^{226}Ra in water can be influenced by a number of factors, such as geological compositions of the water table, the mineral contents of the water and chemical behavior of the nuclide [28].

Pearson correlation coefficients (two tailed) between physicochemical parameters and the ^{226}Ra content quantified in the investigated mineral waters were

Table 4. Physicochemical parameters in mineral water

Parameter	Min. statist.	Max. statist.	Mean statist.	Std. dev. statist.
$T [^{\circ}\text{C}]$	13	98	39	20
pH	6.41	9.95	8.48	0.95
TDS [mgL^{-1}]	1	10892	658	1368
Cl^{-} [μgL^{-1}]	2230	4680320	93210	566374
F^{-} [μgL^{-1}]	110	24500	3775	3984
Na^{+} [μgL^{-1}]	3010	2729000	134701	332412
Ca^{2+} [μgL^{-1}]	400	741480	38901	99921
Mg^{2+} [μgL^{-1}]	30	231040	13965	33263
SO_4^{2-} [μgL^{-1}]	8760	2133600	129435	273531
^{226}Ra [mBqL^{-1}]	30	240	95	49

estimated and presented in tab. 5. We observed a moderate and positive relationship between Ca^{2+} , Mg^{2+} and ^{226}Ra (r , .404 and r , .417, respectively). This could be due to the similar chemical behavior between two valence cations from the second group of the Periodic table (Ra^{2+} , Ca^{2+} and Mg^{2+}). A weak positive correlation was observed between TDS, SO_4^{2-} , Na^{+} , and ^{226}Ra (r , .361, r , .363 and r , .312, respectively). A weak negative correlation was established between pH and ^{226}Ra (r , -.390). No correlation was observed between ^{226}Ra and temperature, Cl^{-} and F^{-} .

Annual effective doses

Mineral waters are frequently used by people for drinking purposes. For this reason an assessment of the effective dose was done.

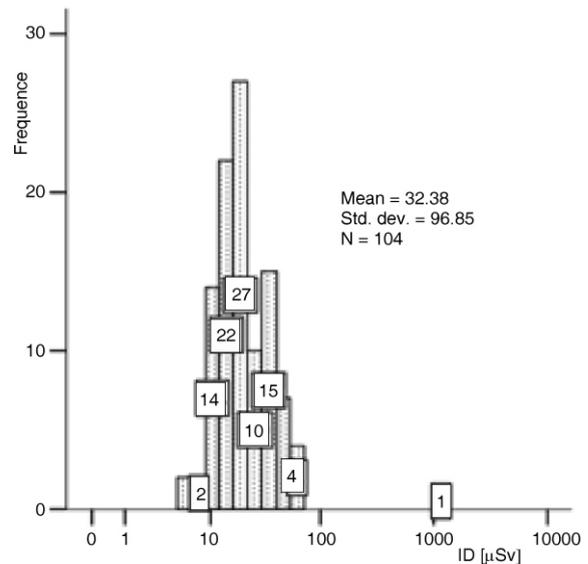
Annual effective dose (ID) from ingestion, due to ^{226}Ra present in mineral water for adults was estimated taking into account that all waters will be used permanently for drinking with 2 l/day consumption per person; . The dose conversion factor for ^{226}Ra is $2.8 \cdot 10^{-04} \text{ mSvBq}^{-1}$ [29].

The following formula was used to calculate the annual effective dose

$$ID = A \cdot V \cdot E$$

where A is the activity concentration of ^{226}Ra [BqL^{-1}], V – the annual intake of mineral water [L] and E is the dose conversion factor of the ^{226}Ra [mSvBq^{-1}].

The values of the estimated annual effective dose, due to ^{226}Ra present in mineral water show a broad variation ranging from 6 μSv to 1000 μSv . The mean value is 32 μSv with a Std. Error of 9 μSv . The

**Figure 3. Histogram with the distribution of the ID due to ^{226}Ra content in mineral waters**

median value is 19 μSv . The frequency distribution of the annual effective dose (ID) is presented in fig. 3.

The calculated dose, with the exception of the highest value is much lower than the Individual Dose Criterion (IDC) for the public 1.0 mSv recommended by the World Health Organization for all groups of the population [30]. WHO defined in the Guidelines for Drinking Water Quality that the additional risk to health from exposure to an annual dose of 0.1 mSv, associated with the intake of radionuclides from drinking water, is considered to be very low.

In the majority of the examined cases the dose is less than 1% of the total effective dose received from all natural sources (2.4 mSv) presented by The the United Nations Scientific Committee on the Effects of Atomic Radiation [31]. The comparisons demonstrate the insignificance of doses from ingestion of mineral water with ^{226}Ra in comparison with other sources of exposure. The water with the highest value for ^{226}Ra content ($4.9 \cdot 1.38 \text{ BqL}^{-1}$) is prohibited for human consumption.

CONCLUSIONS

The diversity of the geological origin of Bulgarian mineral waters constitutes the main factors for the

Table 5. Pearson correlation coefficients (r), related to particular physicochemical parameters and ^{226}Ra content

		$T [^{\circ}\text{C}]$	pH	TDS [mgL^{-1}]	Cl^{-} [μgL^{-1}]	F^{-} [μgL^{-1}]	Na^{+} [μgL^{-1}]	Ca^{2+} [μgL^{-1}]	Mg^{2+} [μgL^{-1}]	SO_4^{2-} [μgL^{-1}]
^{226}Ra [mBqL^{-1}]	Pearson correlation	.198	-.390**	.361**	.264*	.106	.312*	.404**	.417**	.363**
	Sig. (2-tailed)	.143	.003	.006	.049	.439	.019	.002	.004	.006
	N	56	56	56	56	56	56	56	45	55

*Correlation is significant at the 0.05 level (2-tailed); **Correlation is significant at the 0.01 level (2-tailed)

different physicochemical parameters of the waters. The highest concentrations of ^{226}Ra were measured in the mineral waters originating from the carbonate strata, as the dominant geological structure. The lowest concentrations of ^{226}Ra were registered in mineral waters originating from geothermal reservoirs built in effusive rocks and sediments as the dominant geological structure. Thus it is well connected with the estimated moderate positive correlation between Ca^{2+} , Mg^{2+} and Ra^{2+} cations in the water. The quantity of TDS, SO_4^{2-} and Na^+ can influence weakly the ^{226}Ra contents. The pH can influence weakly the ^{226}Ra contents with a negative correlation.

The activities of ^{226}Ra range from a few mBqL^{-1} to 4900 mBqL^{-1} . After excluding the maximum value, the mean value becomes $107 \pm 62 \text{ mBqL}^{-1}$.

The estimated annual dose shows a wide variety of values ranging from $6 \mu\text{Sv}$ to $1000 \mu\text{Sv}$ with the mean value of $32 \pm 9 \mu\text{Sv}$. All results, except one are much lower than the values recommended by the WHO for adults.

The results from this study demonstrate the need for investigation of every single mineral water in the territory of the country, giving more attention to mineral waters with low pH and high content of Ca^{2+} and Mg^{2+} .

AUTHORS' CONTRIBUTIONS

Theoretical and numerical analyses were carried out by R. M. Kamenova-Totzeva, and A. V. Totzev and R. M. Kotova analyzed and discussed the results. The manuscript was written and the figures were prepared by all the authors.

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Росица М. КАМЕНОВА-ТОЦЕВА, Александар В. ТОЦЕВ, Радостина М. КОТОВА

САДРЖАЈ РАДИЈУМА У МИНЕРАЛНИМ ВОДАМА БУГАРСКЕ

У овом раду приказани су резултати испитивања садржаја ^{226}Ra у 104 минералне воде у Бугарској, као и испитивање корелације између физикохемијских параметара, геолошке структуре и специфичне активности ^{226}Ra . Резултати показују да се активност ^{226}Ra креће у опсегу мањем од 30 mBqL^{-1} до 4900 mBqL^{-1} , са средњом вредношћу од 156 mBqL^{-1} . Највеће концентрације ^{226}Ra измерене су у водама које настају из карбонатних слојева као најдоминантније геолошке структуре. Ово је у доброј вези са процењеном умерено позитивном корелацијом између Ca^{2+} , Mg^{2+} и Ra^{2+} катјона у води. Процењена годишња доза показује велику дисперзију резултата у опсегу од $6 \mu\text{Sv}$ до $1000 \mu\text{Sv}$ уз средњу вредност од $32 \text{ } 9 \mu\text{Sv}$. Резултати указују на потребу за даљим испитивањима свих минералних вода, с тим да би већа пажња требало да буде посвећена минералним водама са ниском рН вредношћу и већим садржајем Ca^{2+} и Mg^{2+} .

Кључне речи: ^{226}Ra , минерална вода, корелација, процена дозе