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MEASUREMENTS OF GROSS ALPHA AND BETA ACTIVITY IN DRINKING WATER FROM GALATI REGION, ROMANIA

VIOLETA PINTILIE[1,2], ANTOANETA ENE[3,*], LUCIAN P. GEORGESCU[3], LUMINITA MORARU[3], CATALINA ITICESCU[3]

[1]Department of Public Health Galati, Ionizing Radiation Laboratory, 12B Rosiori St., 800066 Galati, Romania; E-mail: violetapintilie18@gmail.com
[2]"Dunarea de Jos" University of Galati, Doctoral School of Engineering, 47 Domneasca St., 800008 Galati, Romania
[3]"Dunarea de Jos" University of Galati, Faculty of Sciences and Environment, Department of Chemistry, Physics and Environment, 47 Domneasca St., 800008 Galati, Romania
**Corresponding author*: aene@ugal.ro

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Abstract. In this study gross alpha and gross beta activities have been determined in 136 drinking water samples collected in 2014 from all over the territory of Galati County, Romania. Water sources that have been used for producing drinking water in Galati are: drilled wells, spring water and surface water (from the Danube River). The gross alpha activity and gross beta activity ranged from < 6.00 to 85.24 mBq L^{-1}, and from < 25.00 to 438.85 mBq L^{-1}, repectively. There was not registered any exceeding of the maximum allowed limits for gross alpha activity (100 mBq L^{-1}), and gross beta activity (1000 mBq L^{-1}) stipulated in the national legislation. The values obtained in this study were compared with those presented in similar studies from Romania and other regions of the world and with average values reported in Romania in 2014. The highest values, both for gross alpha activity and gross beta activity were determined in drinking water which had the main source the drilled wells. For 84.6% of the analysed samples, the ratio between gross alpha activity and gross beta activity is lower than 1. Assuming that the entire gross alpha and beta activity is due to the most radiotoxic natural radionuclides ^{210}Po (alpha emitter), and ^{228}Ra (beta emitter), the calculated averages of the effective doses due to water ingestion by adults (age >17 years) were 19.43 μSv y^{-1} and 38.17 μSv y^{-1}, respectively, which are lower than the recommended WHO value of 100 μSv y^{-1}.

Key words: gross alpha activity, gross beta activity, water, equivalent effective dose, Galati, Romania.

1. INTRODUCTION

Water is an indispensable component in our lives and the managing of the water resources is a national and international problem. Water bears the radioactive, microbiological and chemical fingerprints of the crossed environment, being a very important transporter of these elements. A major influence on the composition and mobility of components from water has the regional geology [1, 2]. The quality of the water destined to human consumption should be strictly controlled in a certain region as regards the physico-chemical, biological and radioactive parameters [1–4].

The radioactivity of drinking water is an environmental factor which contributes to the population exposure to ionizing radiations and the activity of monitoring the water radioactive content is the responsability of the national public health systems by ensuring the maintaining of the effective dose by ingestion in the provided limits.

The population usage of drinking water represents a way of the population exposure to the ionizing radiations by ingestion the radionuclides existing in it. UNSCEAR estimates that the natural sources contribution to the effective dose is 2.4 mSv y^{-1} (in this dose value being contained the value of 0.3 mSv y^{-1} due to the usage of food and water) [5].

Owing to the complexity of the aquifer structure, variety of the geological background, fluctuations in radionuclide content of soil and bedrock in different layers, depth of water extraction and the large number of physical and chemical processes involved in the transfer of radionuclides from environment to groundwater or fresh water, it is necessary to perform regular measurements of radioactivity parameters of different types of drinking water in each region using simple, low-cost and reliable methods [3, 6–11]. The most commonly used radiometric methods of drinking water analysis for routine monitoring are the screening methods based on the measurement of gross alpha and gross beta activity [11–14] according to ISO methods [15, 16]; in the case that the measured values of radioactivity do not exceed the screening values adopted by World Health Organization (WHO) [17], no further radiological investigation for specific radionuclides is required [7, 13, 18, 19].

The aim of the paper is to determine the levels of gross alpha and beta activity of drinking water from various sources in Galati region, Romania, and to assess the effective doses due to water ingestion by adult members of the public. To the best of our knowledge, there is no study performed so far in this region on drinking water radioactivity.

2. EXPERIMENTAL

2.1. STUDIED AREA AND SAMPLING

The analysis of the drinking water was carried out in Galati County, an important zone in Lower Danube Euroregion and Black Sea basin, situated in the South-East Development Region of Romania, at the confluence of the Danube with its main tributaries in the Lower Danube River basin, Siret and Prut Rivers, at the border between Romania and Republic of Moldova in the western part [20], between 45°25' and 46°10' North latitude, 27°20' and 28°10' East longitude (Fig. 1). Galati County is a confluence zone between the Covurlui Plateau in the north part (50% of the county surface), Tecuci and Covurlui plains (34%) and Lower Siret and Prut (16%) in the south part [21]. It has an area of 4,466.3 km^2 (representing 1.9% of the surface of Romania), 604,556 inhabitants and includes 4 urban localities (2 municipalities – Galati and Tecuci, and 2 cities – Tirgu Bujor and Beresti) and 61 communes, totalling 181 villages [21]. The capital, Galati town, is an industrialized center located in the south of the county (Fig. 1), on the banks of Danube River, being the biggest port on the maritime Danube [20].

Gross alpha and gross beta activity concentrations were determined for a number of 136 drinking water samples, labeled DW1-DW136, in order to evaluate the accordance between the radiological parameters and the radioactive parameters of potability.

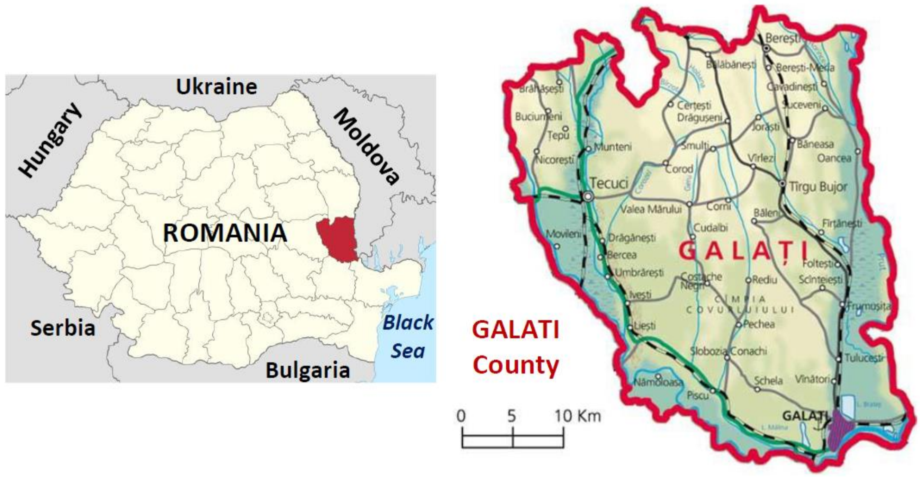

Fig. 1 – The sampling area in Galati County, Romania.

The drinking water samples were collected from all the towns and villages of Galati. From all the 136 samples, 4 samples (labeled DW1-DW4) have as source the surface water (the Danube River) which supplies Galati town, 7 samples (DW5-DW11) have as source the captured springs, and the others (DW12-DW136) have as source drilled wells.

2.2. METHOD

The most accepted protocol used as the first step of radiological characterization of drinking waters is gross alpha and gross beta activity measurement [10, 18, 19], called "thick source method", which includes the standards ISO 9696:2007 [15] and ISO 9697:2008 [16], respectively, for non-saline water.

The water samples were collected in plastic containers and were acidified with concentrated HNO_3. The acidification of the water samples reduces, by adsorption, the loss of the reactive substances from the water. Initially, it was determined the total dissolved solids (TDS) content (mg L^{-1}) in order to estimate the quantity of water necessary to be evaporated in order to obtain sufficient residue for being used in the radiometric measurements. The next step was the evaporation of the samples on a hot ceramic plate heated to obtain approximately 50 mL of concentrated sample, followed by its conversion to sulphate by mixing with 1 mL of concentrated H_2SO_4 and calcination at 350 ºC for an hour. The obtained residue was transferred to a stainless steel planchet. Each planchet was measured 10 times for 100 min for gross alpha/beta activity at Ionizing Radiation Laboratory, Department of Public Health Galati, using a low-background MPC 2000-DP (Protean Instruments Corporation) counting system with a ZnS dual phosphor detector (zinc sulphide and plastic) calibrated at Horia Hulubei National Institute of R&D in Physics and Nuclear Engineering (IFIN-HH), Magurele.

The solid residue required for analysis was calculated as function of the self-absorbtion curve for this installation. From our observations using the counting modes *only alpha* and *only beta* of low-background gross alpha/beta activity counter MPC-2000-DP, the ratio of residue weights for alpha measurements and beta measurements is 1:10. The self-absorbtion calibration curve has been obtained with the aid of a ^{241}Am standard solution (728 ± 22 Bq g^{-1}, IFIN-HH) and KCl salt (Merck). Therefore, the results obtained (mBq L^{-1}) are gross alpha activity ^{241}Am equivalent and gross beta activity ^{40}K equivalent. The minimum detectable activities, determined by us for the used installation are $LDA_\alpha = 6.00$ mBq for gross alpha measurements and $LDA_\beta = 25.00$ mBq for gross beta measurements.

3. RESULTS AND DISCUSSION

3.1. ACTIVITY CONCENTRATIONS

The results determined in this work for all the 136 drinking water samples are presented in Fig. 2 (gross alpha activity, Λ_α, in mBq L^{-1}) and Fig. 3 (gross beta activity, Λ_β, in mBq L^{-1}) and the statistical data for all the categories of water sources investigated by us are given in Table 1, together with TDS values and the synthetic national reports for the 1408 drinking water sources used in Romania in 2014 [22]. The complete datasets are available on request from the authors.

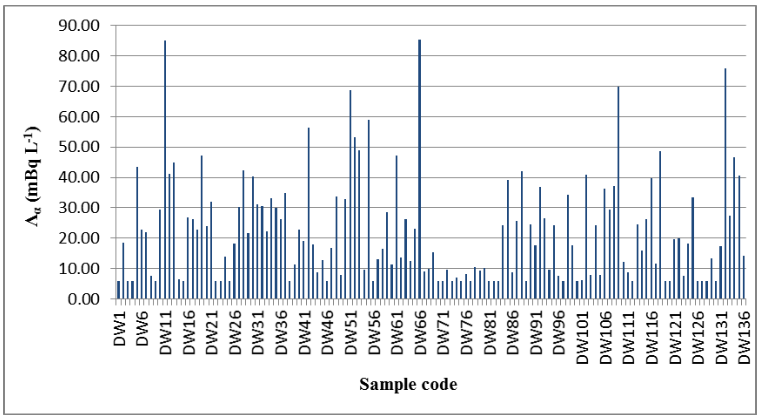

Fig. 2 –Alpha gross activity in drinking water from Galati, Romania (2014).

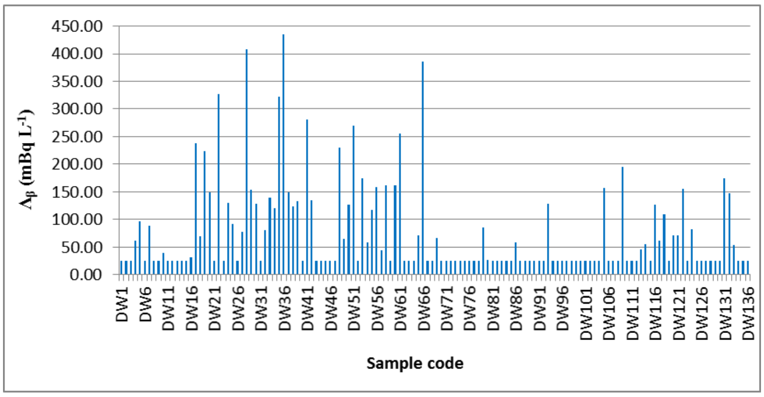

Fig. 3 – Gross beta activity in drinking water from Galati, Romania (2014).

The values of gross alpha activity (mBq L^{-1}) ranged from < 6.00 mBq L^{-1} to 85.24 mBq L^{-1} with an average of 22.18 ± 6.65 mBq L^{-1}, compared to the average 21.30 mBq L^{-1} obtained in Romania for 2014 [22] (Table 1). In 29 from the 136 analyzed samples, overall alpha gross activity is lower than LDA$_\alpha$ of 6.00 mBq. The maximum alpha gross activity of 85.24 mBq L^{-1} was determined in samples collected from Scinteiesti commune, Fantanele village (DW66), having as source the drilled wells.

The values of gross beta activity (mBq L^{-1}) determined for all the investigated samples ranged from < 25.00 to 434.85 mBq L^{-1} with an average of 75.79 ± 19.69 mBq L^{-1}, compared to 100.3 mBq L^{-1} which is the average (Table 1) obtained in Romania for 2014 [22].

In 76 from the 136 analyzed samples, overall beta gross activity is less than LDA$_\beta$ of 25.00 mBq. The highest value of beta gross activity of 434.85 mBq L^{-1}

was determined in samples collected from Slobozia Conachi (DW36) (drilled wells).

In the graphics drawn in Figs. 2 and 3, in the case of the samples for which Λ_α < LDA_α and Λ_β < LDA_β, we considered the values of minimum detectable activities of 6.00 mBq L^{-1} for alpha gross activity and 25.00 mBq L^{-1} for gross beta activity.

Table 1

Statistical values obtained in this work for TDS and gross alpha/beta activity of drinking water, compared with data from the national report in 2014

Source	Sample code	Level	Λ_α (mBq L^{-1})	Λ_β (mBq L^{-1})	TDS (mg L^{-1})
All sources* Galati	DW1-DW136	minimum	<6.00±1.80	<25.00±6.25	236.00
		average	22.18±6.65	75.80±18.95	534.09
		maximum	85.24±25.57	434.85±108.71	1416.00
Surface water Galati	DW1-DW4	minimum	<6.00±1.80	<25.00±6.25	260.00
		average	9.13±2.74	34.34±8.59	346.00
		maximum	18.50±5.55	62.38±15.59	388.00
Captured springs Galati	DW5-DW11	minimum	<6.00±1.80	<25.00±6.25	328.00
		average	30.88±9.26	46.41±11.60	782.86
		maximum	85.00±25.50	96.53±24.13	1416.00
Drilled wells Galati	DW12-DW136	minimum	<6.00±1.80	<25.00±6.25	236.00
		average	22.11±6.63	78.77±19.69	526.18
		maximum	85.24±25.57	434.85±108.71	1264.00
All sources* Romania	Romania**	minimum	1	1	–
		average	21.3	100.3	–
		maximum	100	880	–

* Surface water, captured springs, drilled wells;
** Data from national report (synthesis of 1408 results obtained in Romania, 2014), ref. [22].

From Figs. 2 and 3 it can be observed a large span of the values obtained for different waters, and a site-specificity of the activity concentrations, probably due to the wide deviations of natural radionuclide composition and abundence of the minerals in the earth's crust [23]. For 84.6% of the analyzed samples, the gross alpha activity is lower than the gross beta activity, trend also reported by other authors [7, 11, 24].

Our results (average, minimum, maximum values of gross alpha/beta activity) are comparable with those obtained for various regions or/and countries in similar studies (drinking water, tap water, mineral water, spring water), as resulting from Table 2.

Table 2

Comparison of gross alpha and beta activity values obtained in this work for drinking water (in mBq L^{-1}) with literature values

Region/Country	Λ_α (mBq L^{-1})		Λ_β (mBq L^{-1})		Reference
	Average	Range*	Average	Range*	
Galati/Romania	22.18	<6.00–85.24	75.80	<25.00–434.85	Present work
Bucovina/Romania	12.13	0.40–45.40	11.34	1.51–47.45	[2]
Balaton/Hungary	189	35–1749	209	33–2015	[8]
Serbia	–	1–13	–	41–173	[24]
Italy	–	<8–186	–	<48–150	[6]
The Marche region/Italy	–	18.18–128.18	–	<41.57–258.59	[18]
Orwian/Nigeria	–	6.4–18.2	–	46–126	[13]
Sao Paulo and Minas Gerais States/Brazil	–	1-428	–	120–860	[14]
Batman/Turkey	38.1	10.8–73.4	79.6	3–347	[11]
Adana/Turkey	9.60	0.32–22.9	86	24–290.7	[25]
Hail/ Saudi Arabia	215	17–541	260	480–516	[26]
Saratonga/USA	–	<40–310	–	110–189	[27]

* min-max interval.

The highest gross alpha and beta activity concentrations were found in samples collected from drilled wells. In any case, the maximum admitted concentrations in national legislation [28], of 100 mBq L^{-1} for alpha gross activity and 1000 mBq L^{-1} of beta gross activity, were not exceeded. There is a poor correlation between gross alpha and beta activities in the investigated waters, suggesting that different radionuclides might be responsible for the water contamination [13]. No correlation was found between Λ_α or Λ_β and TDS, fact reported in other studies [2, 14], indicating that radionuclides release from the reservoir rock to the liquid phase is dependent on a large number of parameters, being difficult to properly evaluate [14].

3.2. THE ANNUAL EQUIVALENT EFFECTIVE DOSE DUE TO WATER INGESTION

The specific activities of the natural radionuclides from drinking water are mainly due to the presence of naturally occuring radionuclides of both the uranium and thorium decay series [6, 9, 14, 24]. In order to calculate the annual internal dose, the values of effective dose conversion factor by ingestion, CF (in Sv Bq^{-1}), given in ref. [29] were used, presented in Table 3. For the most important natural radionuclides from uranium and thorium series, the effective dose conversion by ingestion for adults grows in the following order:

$^{238}U < ^{235}U < ^{234}U < ^{224}Ra < ^{226}Ra < ^{210}Pb < ^{228}Ra < ^{210}Po$.

Therefore, in order to calculate the annual internal dose, we considered that alpha gross activity is due to ^{226}Ra and ^{210}Po and beta gross activity due to ^{228}Ra and ^{210}Pb [7, 11, 24], which are emitters with the highest effective conversion factors CF.

The annual effective dose equivalent D_{ef} (Sv y^{-1}) associated with radiation exposure through the ingestion of water was estimated to assess the health risk for adults using the equation (1):

$$D_{ef} = \Lambda_{\alpha/\beta} \text{ x } IR_w \times CF \,, \quad (1)$$

where $\Lambda_{\alpha/\beta}$ is gross alpha/beta activity (Bq L^{-1}); IR_w – intake of water for one person in one year (L y^{-1}); CF – age-dependent effective dose conversion factor (Sv Bq^{-1}). In this work, the annual consumption rate (IR_w) was estimated at the value of 730 L, according to WHO [17] for an adult person (age >17 years).

Table 3

Effective dose conversion factors by ingestion, CF (Sv Bq^{-1}), for the most important natural radionuclides [29]

Radionuclide	Half life	Adults CF (Sv Bq^{-1})
^{238}U	4.47E+09 y	4.5E-08
^{235}U	7.04E+08 y	4.7E-08
^{234}U	244000 y	4.9E-08
^{224}Ra	3.66 d	6.5E-08
^{226}Ra	1600 y	2.8E-07
^{210}Pb	22.3 y	6.9E-07
^{228}Ra	5.75 y	6.9E-07
^{210}Po	138 d	1.2E-06
^{40}K	1.28E+09 y	6.2E-09

The calculated annual effective doses (mSv y^{-1}) for adult population in Galati region in 2014 due to intake of ^{210}Po, ^{226}Ra, ^{210}Pb and ^{228}Ra radionuclides from water are represented in Figs. 4–6 and the statistical values (μSv y^{-1}) for different categories of water sources are given in Table 4. Due to the fact that the CFs for ^{210}Pb and ^{228}Ra are equal, it results that the annual effective doses D_{ef} for both radionuclides are the same and were represented in a single figure (Fig. 6) and one column in Table 4. From Table 4 it can be seen that the annual effective doses due to alpha emitters in drinking waters from all sources range between 5.26–74.67 μSv y^{-1} for ^{210}Po (average 19.43 μSv y^{-1}) and 1.23–17.42 μSv y^{-1} for

^{226}Ra (average 4.53 μSv y^{-1}). In the case of the beta emitters $^{210}Pb/^{228}Ra$, the annual effective doses range from 12.59–219.03 μSv y^{-1}, with an average of 38.18 μSv y^{-1}.

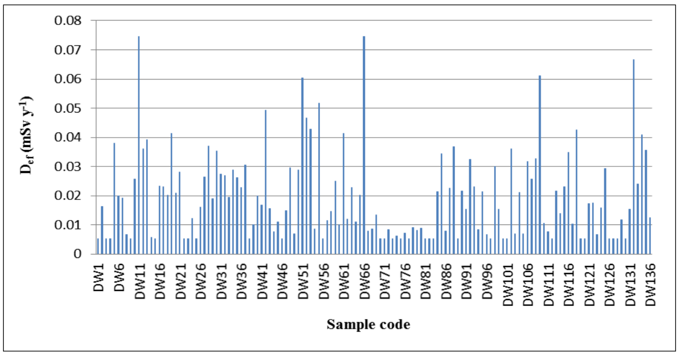

Fig. 4 – The annual effective dose (mSv y^{-1}) due to intake of ^{210}Po from drinking water (Galati, Romania, 2014).

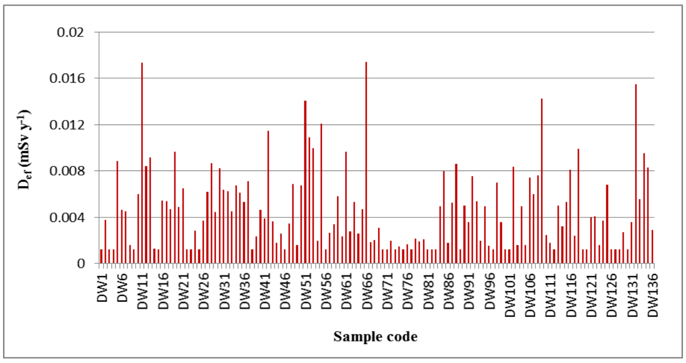

Fig. 5 – The annual effective dose (mSv y^{-1}) due to intake of ^{226}Ra from drinking water (Galati, Romania, 2014).

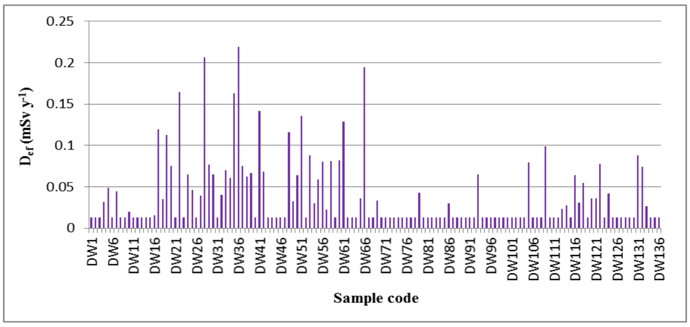

Fig. 6 – The annual effective dose (mSv y^{-1}) due to intake of $^{210}Pb/^{228}Ra$ from drinking water (Galati, Romania, 2014).

Table 4

Statistical values obtained in this work for the annual effective doses (μSv y^{-1}) of various natural radionuclides in water samples from Galati, Romania, in 2014

Source	Sample code	Level	D_{ef} (μSv y^{-1}) (adults)		
			^{210}Po	^{226}Ra	^{210}Pb/^{228}Ra*
All sources	DW1-DW136	minimum	5.26	1.23	12.59
		average	19.43	4.53	38.18
		maximum	74.67	17.42	219.03
Surface water	DW1-DW6	minimum	5.26	1.23	12.59
		average	7.99	1.87	17.30
		maximum	16.21	3.78	31.42
Captured springs	DW7-DW11	minimum	5.26	1.23	12.59
		average	27.05	6.31	23.38
		maximum	74.46	17.37	48.62
Drilled wells	DW11-D136	minimum	5.26	1.23	12.59
		average	19.37	4.52	39.67
		maximum	74.67	17.42	219.03

* CFs for ^{210}Pb and ^{228}Ra are equal and the annual effective doses for both radionuclides are the same.

In this study the results for the annual effective dose due to intake of alpha emitters ^{210}Po (Fig. 4) and ^{226}Ra (Fig. 5) from drinking water indicate values below the WHO recommended reference level of 0.1 mSv y^{-1} for one adult person [17].

With a view to the annual effective dose due to intake of beta emitters ^{210}Pb/^{228}Ra from drinking water (Fig. 6), for 11 samples (having as sources drilled wells) – DW17, DW19, DW22, DW28, DW35, DW36, DW41, DW48, DW51, DW61, DW66 – the calculated values indicate an exceeding of the norm of 0.1 mSv y^{-1}. The highest value of the annual effective dose due to ^{210}Pb/^{228}Ra intake from drinking water was found for sample DW36 (Slobozia Conachi), which obviously had the highest gross beta activity.

Of course, the assumption of gross alpha activity as being due to ^{210}Po and ^{226}Ra and gross beta activity due to ^{210}Pb and ^{228}Ra, which are highly radiotoxic radionuclides, is a drastic one [11, 24], the worst case. It is necesary the determination of specific activities of the ^{210}Po, ^{226}Ra, ^{210}Pb and ^{228}Ra radionuclides from potable water in order to perform a real assessment of the annual effective dose by water ingestion. It should also estimate the contribution of ^{40}K radionuclide, with a lower CF but with a major contribution in drinking water. However, as recommended by Directive 2013/51 Euroatom [30], if gross alpha activity and gross beta activity are less 0.1 Bq L^{-1} and 1 Bq L^{-1}, respectively, it can be considered that the annual effective doses are below the WHO recommended reference level of 0.1 mSv y^{-1} and it does not require radiological investigation unless it is known from other source of information that the specific radionuclides present in water may cause a dose larger than 0.1 mSv y^{-1} [30] .

Harmonization of European Directive 51/2013 [30] in Romania was made by Law 301/2015 [31] establishing health protection requirement with regarding the radioactive substances in drinking water. In the next years, according to this law, the total effective dose by water ingestion will be estimated through the determination of all the natural radionuclides whose presence has been detected in a water reservoir, especially the ones with the biggest effective dose conversion factors, with the exception of tritium, ^{40}K, radon and short-lived products from radon disintegration. This represents the subject of future studies.

4. CONCLUSIONS

The present study is the assessment of the gross alpha and gross beta activity of all drinking water sources used in 2014 in Galati County, Romania, and the radiological implications on the adult population which are using these waters for consumption. It is the first study performed in the region regarding the radioactivity of drinking water and will serve as a base in further investigations. The results will be correlated with those obtained in Lower Danube Euroregion for outdoor radiation dose rates and water (surface, underground) chemical and microbiological parameters [32, 33].

The values of gross alpha and beta activity are below the recommended values in national and European legislation – 0.1 Bq L^{-1} and 1 Bq L^{-1}, respectively. It was evaluated the annual effective dose due to the ingestion of drinking water, based on the assumption that the major contribution had the ^{210}Po, ^{226}Ra (alpha emitters) and ^{210}Pb, ^{228}Ra (beta emitters) radionuclides. Starting with a value of 193.23 mBq L^{-1} for gross beta activity, the assumption that beta gross activity was entirely due to the ^{210}Pb or ^{228}Ra radionuclide becomes too drastic, resulting in the exceeding of the anual effective dose of 100 μSv y^{-1} (in this study 11 samples out of 136 are in this situation). To correctly estimate the annual effective dose due to the beta-emitting radionuclides in the studied samples, it is necessary to determine the specific activity of ^{40}K, ^{210}Pb and ^{228}Ra radionuclides.

This study aimed to reveal that although gross beta activity from drinking water is below 1 Bq L^{-1}, the recommended annual effective dose (100 μSv y^{-1}) can be exceeded. This happens in the case that we consider the entire beta activity due to ^{210}Pb or ^{228}Ra radionuclides. It is necessary to establish the specific activities of natural radionuclides in drinking water from Romania, especially for the radionuclides with the highest effective dose conversion factors, to be able to certainly state that the annual effective dose due to ingestion of water does not exceed 100 μSv y^{-1} for beta activities values below 1 Bq L^{-1}.

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