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NATURAL RADIOACTIVITY IN DRINKING WATER FROM GALATI AND VRANCEA AREAS, ROMANIA*

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Abstract. *Pollution and contamination of drinking water grow rapidly due to industrial growth and urbanization. They potentially cause severe problems to health, so the water quality management addresses both national and international action to assess and prevent the associated hazard. The quality of drinking water must be strictly controlled. The aim of the study was to determine the concentration of ^{210}Po, ^{210}Pb, ^{226}Ra in drinking water. The occurrence of ^{210}Po and ^{210}Pb has been determined by spontaneous deposition onto a nickel disc and the gross alpha activity has been measured. ^{226}Ra has been measured after 30 days of storage in order to be sure that ^{222}Rn in water samples reached secular equilibrium. Water samples were collected from a total of 17 sites, which serve 556,125 persons, in Galati and Vrancea areas of the East part of Romania. Samples were collected during March and April of 2015. The monitoring of drinking water samples indicated the presence of ^{210}Po, ^{210}Pb, ^{226}Ra at a concentration ranging from 1.9 to 12.5 mBq L^{-1}, 3.2 and 15.9 mBq L^{-1}, 8.0 and 30.0 mBq L^{-1}, respectively. These values were compared with the maximum permitted level, according to national legislation. The average annual committed effective dose from the intake of water ranged between $4.15x10^{-3}$ and $18.80x10^{-3}$ mSv y^{-1}, for adults, which is lower than the recommended reference value of 0.1 mSv in water for human consumption according to the current EU legislation.*

Key words: *Water, polonium, radium, lead, equivalent effective dose, Galati, Vrancea*

1. INTRODUCTION

Drinking water is the most important water for humankind. The consumed water participates in all cellular processes, but simultaneously it is a way for the ingestion of radionuclides and can pose a number of health hazards. UNSCEAR estimates that the natural sources have a contribution of 2.4 mSv y^{-1} to the effective dose, a value which includes the contribution of 0.3 mSv y^{-1} due to the usage of aliments and water [1].

The quantification of the radiological quality of drinking water is made through the calculation of the effective dose equivalent due to its ingestion, after determining the concentration of radionuclides occurring in water.

The World Health Organization (WHO) and the European Directive 2013/51 recommended a value of 0.1 mSv for the effective dose equivalent for one person per year, as a safe level [2], [3]. The natural radionuclides from drinking water come from three naturally radioactive series: thorium series, actinium series and uranium series. In the case of natural radionuclides of uranium and thorium series, the effective dose conversion factor by ingestion for adults grows in the following order:

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

Polonium is the most radiotoxic alpha emitter, due to the relatively high energy (5.3 MeV), and it is concentrated in soft tissues, such as muscles and liver, or produces sclerotic changes in blood vessels [4]. Lead tends to accumulate in bone but is also distributed to the brain, liver and kidney. ^{210}Po and ^{210}Pb might be present inside the body in the absence of direct intake from decay of ^{226}Ra [1]. The radium's metabolism is similar to the metabolism of calcium and a considerable amount is stored in the bones [5].

The guidance levels for ^{210}Po and ^{210}Pb are of 0.1 Bq L^{-1} and for ^{226}Ra is 1 Bq L^{-1}, according to the "Guidelines for drinking water quality-third edition"-WHO 2008 [6]. The derived concentrations for radioactivity in water intended for human consumption for ^{226}Ra, ^{210}Po, and ^{210}Pb are 0.5 Bq L^{-1}, 0.1 Bq L^{-1}, and 0.2 Bq L^{-1}, respectively [3].

The main goal of this study was to determine the gross alpha activity, gross beta activity and specific activities of ^{210}Po, ^{210}Pb and ^{226}Ra in drinking water from Galati and Vrancea areas of the Eastern part of Romania and to assess the effective dose equivalent due to the ingestion of radionuclides from drinking water.

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2. EXPERIMENTAL

2.1. Studied area and sampling

The analysis of drinking water was carried out in Galati and Vrancea counties, two areas located in the Eastern part of Romania, representing 4.3% of the surface of the country (Fig.1). The Galati County is situated between 45°25′ and 46°10′ North latitude and 27°20′ and 28°10′ East longitude [7] and Vrancea County between 45°23′ and 46°11′ North latitude and 26°23′ and 27°32′ East longitude [8].

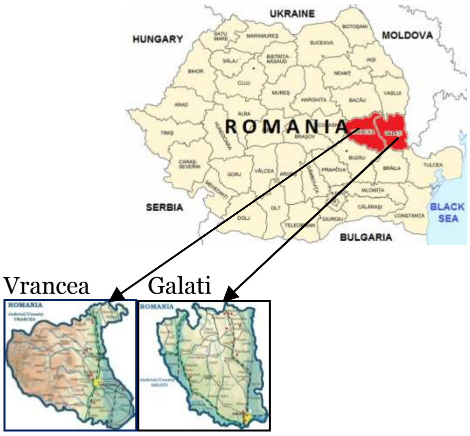

Figure 1. The sampling area in Galati and Vrancea counties, Romania (2015)

Galati County includes four urban localities (two municipalities- Galati and Tecuci, and two cities- Tg. Bujor and Beresti) and 61 communes, totaling 181 villages. The urban water supplying the Galati municipality is provided by two waterworks which use two water sources (labeled as DW1 and DW2). For the Tecuci municipality, the two other cities and six communes, the drinking water samples were collected from sources labeled DW3 -DW11.

Vrancea County includes five urban localities (two municipalities - Focsani and Adjud, and three cities - Marasesti, Odobesti and Panciu) and 68 communes, totaling 331 villages. From all five urban localities and one commune of the Vrancea County, drinking water samples labeled DW12-DW17 were collected and analyzed.

From all the 17 samples, one sample has surface water (the Danube River) as a source, three samples have a captured spring as a source, and the others have drilled wells as a source.

2.2. Measurement of the gross alpha/beta activity

The gross alpha activity and gross beta activity measurements represent the general screening of the radiological quality of drinking water; in the case that the measured values of radioactivity do not exceed the screening value adopted by World Health Organization [2], the annual effective doses are bellow the recommended reference level of 0.1 mSv y^{-1}, and no further radiological investigation for specific radionuclides is required [3].

To obtain a sufficient amount of residues for radiometric measurements, the amount of evaporated water and the total dissolved solid substance (mg L^{-1}) was determined.

The alpha/beta activity was determined in accordance with ISO 9696 and ISO 9697, respectively [9], [10]. The sample stabilized by acidification is evaporated almost to dryness, converted to the sulphate form and ashed at 350°C. A part of the obtained sample residue is transferred to a stainless steel planchet. The activity of the stainless steel planchet with residue was measured. Gross alpha/beta activity of the sample was measured using low-background MPC-2000-DP system (Protean Instruments Corporation) [11], calibrated at the National R&D Institute for Physics and Nuclear Engineering IFIN-HH, Bucharest. The counter MPC-2000-DP is equipped with a dual phosphor scintillator type detector. The samples were simultaneously measured in alpha and beta mode. The weight of the sample transferred to the stainless planchet was approximately 90 mg. This value was established according to the self-absorption curve. The calibration of MPC-2000-DP system was made by using two standard sources: ^{241}Am (serial no: 2830, LMRI France) and ^{90}Sr/Y (serial no:9891, LMRI France). The measurements were performed during a period of 30 minutes, ten times per sample.

The gross alpha or is beta activity calculated using equation (1):

$$A_{\alpha/\beta} = \frac{(R_{\alpha/\beta} - R_0) \times TDS}{m \times \varepsilon} \quad \text{Bq L}^{-1} \qquad (1)$$

where $A_{\alpha/\beta}$ is the gross alpha/beta activity of the drinking water sample, $R_{\alpha/\beta}$ is the rate of alpha/beta measurement for the sample (counts s^{-1}), R_0 is the rate of alpha/beta measurement for background (counts s^{-1}), TDS is the concentration of the total dissolved solids of the sample (g L^{-1}), m is the weight of residue transferred to a stainless steel planchet for measurements (g) and ε is the efficiency of the detector.

2.3. Determination of ^{210}Po and ^{210}Pb concentration

The concentration of ^{210}Po and ^{210}Pb was determined in accordance with STAS 1244-86 [12], [13].

A quantity of 4 L of each water sample was acidified up to pH=2. Then, the sample was mineralized as follows: evaporation of the sample to near dryness; addition of 5 mL HNO3 (density of 1.40 g L^{-1}) and 5 mL H2O2 (30%), followed by evaporation – this step was repeated 4-5 times until white residue was obtained (up to getting a white residue). The white residue was dissolved in 15 mL HCl (density of 1.19 g L^{-1}) and then was evaporated to near dryness– this step was repeated twice. In this way, a white mineralized residue developed, which was transferred in a beaker with 15 mL HCl (density of 1.19 g/L) and 15 mL of distilled water. The solution was diluted to 100 mL with distilled water. Then, NaOH (30%) was used to neutralize the sample in the presence of the phenolphthalein. The neutral solution was acidified with HCl (density of 1.19 g L^{-1}) up to pH of 1.5 and 200 mg of ascorbic acid was added to prevent co-plating of interfering ions. In the end, ^{210}Po in the solution was spontaneously plated for four hours on a nickel disc. Also, the system was heated at 90 °C. Gross alpha activity of the nickel disc was measured using MPC-2000-DP system, during a period of 400 minutes. The specific activity of ^{210}Po is calculated as:

$$A_{210Po} = \frac{(R_{sample} - R_0)}{V_{sample} \times \varepsilon \times \eta} \quad \text{Bq L}^{-1} \qquad (2)$$

where A_{210Po} is the activity concentration of the sample (Bq L^{-1}), R_{sample} is the rate of measurement for the sample (counts s^{-1}), R_0 is the rate of measurement for background (counts s^{-1}), V_{sample} is the volume of the sample (L) and η is the chemical recovery of radiochemical separation.

The rest of the solution was stored for three months and then the procedure of spontaneous deposition on the nickel disc was repeated. The nickel disc is gross alpha measured. ^{210}Pb activity is determined through its daughter ^{210}Po.

The new activity concentration of ^{210}Pb is calculated using the corrected formula:

$$A_{210Pb} = \frac{(R_{sample} - R_0)}{V_{sample} \times \varepsilon \times \eta \times 0.37} \quad \text{Bq L}^{-1} \qquad (3)$$

where 0.37 is a correction factor which indicates the value of the secular equilibrium stage of ^{210}Po and ^{210}Pb after three months.

Since the method used does not allow simultaneous determinations of recovery yield and the concentration of Po-210 or Pb-210, in this paper a recovery yield at the amount of 100% was considered.

Therefore the concentrations determined for ^{210}Po and ^{210}Pb may be higher.

2.4. *Determination of ^{226}Ra concentration*

Ra-226 measurement method indicated in the instructions for use of the instrument SARAD-RTM were used [14].

The water samples were stored 30 days in a sealed system before the determination of the activity of ^{226}Ra (Fig. 2). At the end of this period, enough ^{222}Rn was built up from ^{226}Ra water samples.

After this period, the bubbling flask was connected to the module SARAD RTM 1688-2, without being opened, using Mohr's clamps (Fig. 3). After the connection of the bubbling flask to the measurement system, the Mohr's clamp was opened in such a way that the water did not come into contact with the air outside.

Figure 2. The sample sealed and stored for 30 days

The measurements were performed for 30 minutes and were repeated four times for each sample. Before each sample's measurement, the module, all connection tubes and the optional protection flask had to be flushed with fresh air for at least 15 minutes.

Figure 3. SARAD RTM-1688-2 measuring system for ^{226}Ra in water

The acquired measurement data are downloaded to the serial interface. The software calculates the activity concentration of ^{226}Ra (Bq L^{-1}) using the equation [14]:

$$A_{226Ra} = \frac{1}{V_{sample}} \times C_{air} \times [k \times V_{sample} + V_{air}] \quad \text{Bq L}^{-1} \qquad (4)$$

here, A_{226Ra} – the activity concentration of ^{226}Ra (Bq L-1), V_{sample} – the volume of the sample (L), C_{air} – the value indicated by the measuring system (Bq L-1), V_{air} – the volume of the air from the measuring system (L), k – the temperature correction factor.

3. Results and discussion

3.1. *Activity concentration*

The values of the activity concentrations for gross alpha, gross beta, ^{210}Po, ^{210}Pb and ^{226}Ra, together with total dissolved substances (TDS) and total population in each investigated site are presented in Table 1. No exceeding of the maximum permitted limits was registered for gross alpha activity (100 mBq L^{-1}), gross beta activity (1000 mBq L^{-1}), and activity concentration of ^{210}Po (136 mBq L^{-1}), ^{210}Pb (25 mBq L^{-1}), and ^{226}Ra (88 mBq L^{-1}) stipulated in the national legislation [15].

For the all samples, the ratio between gross alpha activity and gross beta activity is lower than 1. In the case of three samples, gross alpha activity shows values lower than 6.0 mBq L^{-1}, which is the minimum detectable activity. The highest value of gross alpha activity was found in the sample DW16, which belongs to Marasesti city, Vrancea. The highest gross beta activity was found in Gugesti, Vrancea (the sample DW17). Also, at this site, the highest value for activity concentration of ^{210}Pb was found. The highest activity concentration of ^{226}Ra and ^{210}Po was found in Galati (DW1 and DW7, respectively).

Table 1. Gross alpha, gross beta, and radionuclide activity concentration (in mBq L^{-1}) in drinking water samples from Galati and Vrancea areas, Romania, in 2015

Sample code	Source	Persons	TDS (mg L^{-1})	A_{α} (mBq L^{-1})	A_{β} (mBq L^{-1})	A_{210Po} (mBq L^{-1})	A_{210Pb} (mBq L^{-1})	A_{226Ra} (mBq L^{-1})
DW1	surface water (Danube River)	260000	564	6.0±2.4	412.3±123.7	2.0±0.4	3.3±0.7	30.0±9.0
DW2	drilled wells	104000	732	8.4±3.4	416.8±125.0	1.9±0.4	3.2±0.7	23.0±6.9
DW3	drilled wells	28000	504	8.7±3.5	221.6±66.5	2.0±0.4	5.3±1.2	13.0±3.9
DW4	captured spring	1905	756	39.0±11.7	219.9±66.0	12.0±2.6	8.4±1.7	27.0±8.1
DW5	drilled wells	4425	776	13.3±5.3	389.9±117.0	12.2±2.7	3.3±0.8	20.0±6.0
DW6	captured spring	669	1016	6.0±2.4	628.4±188.5	2.0±0.4	11.7±2.7	18.0±5.4
DW7	captured spring	345	736	16.8±6.7	484.8±145.4	12.5±2.7	3.8±0.9	27.0±8.1
DW8	drilled wells	750	864	6.0±2.4	564.0±169.2	2.4±0.5	7.5±1.5	20.0±6.0
DW9	drilled wells	600	968	27.7±11.1	677.3±203.2	2.5±0.6	5.3±1.1	27.0±8.1
DW10	drilled wells	771	684	27.7±11.1	313.4±94.0	2.0±0.4	3.8±0.8	23.0±6.9
DW11	drilled wells	825	968	19.6±7.8	226.6±68.0	7.8±1.7	7.7±1.5	8.0±2.4
DW12	drilled wells	18432	744	21.1±8.4	91.6±27.5	2.8±0.6	3.3±0.7	8.0±2.4
DW13	drilled wells	98646	1032	56.8±22.7	329.7±98.9	2.0±0.4	6.3±1.4	8.0±2.4
DW14	drilled wells	8566	668	36.8±14.7	122.9±36.9	1.9±0.4	3.4±0.7	15.0±4.5
DW15	drilled wells	8735	592	9.8±3.9	615.4±184.6	2.0±0.4	4.4±1.0	24.0±7.2
DW16	drilled wells	12667	692	57.1±22.9	324.1±97.2	2.0±0.4	3.2±0.0	27.0±8.1
DW17	drilled wells	6789	888	20.0±8.0	901.2±270.4	4.5±1.0	15.9±3.7	14.0±4.2
mean	All sources*	556125**	775.52	22.4±8.7	408.2±122.5	4.4±1.0	2.9±1.2	19.5±5.8
range			504-1032	<6-57.1	91.6-901.2	1.9-12.5	3.21-15.9	8-30

*surface water, captured spring, drilled wells;

**the sum of water users (Galati and Vrancea, Romania).

From Table 1 it can be seen that the average value of TDS concentration for Galati samples (DW1 - DW11) is 760 mg L^{-1} and is comparable with the average value of 797 mg L^{-1} obtained for Vrancea samples (DW12 - DW17). The observed gross alpha activity found in the first data set (expressed in average±standard deviation) is 16.3±11.1 mBq L^{-1}, while for the second data set is 33.6±20.0 mg L^{-1}. The average value for gross beta activity is 432.8±129.8 mBq L^{-1} for the first data set and 373.5±111.9 mBq L^{-1} for the second data set. The activity concentrations of ^{210}Po and ^{226}Ra are 1.5 higher in the first data set than the second data set.

3.2. Effective dose equivalent

The specific activities of the natural radionuclides from drinking water are due to the presence of naturally occurring radionuclides of both the uranium and thorium decay series. To calculate the annual internal dose, we used the values of effective dose conversion factor by ingestion (CF) shown in Table 2 for each radionuclide of interest [16]:

Table 2. Effective dose conversion factor for ^{210}Po, ^{210}Pb and ^{226}Ra

Radionuclide	T1/2	Effective dose conversion factor by ingestion, CF (Sv Bq^{-1})		
		lactation age*	children (1-17 years)	adults (>17 years)
^{226}Ra	1600 y	4.7E-06	1.5E-06	2.8E-07
^{210}Pb	22.3 y	8.4E-06	1.9E-06	6.9E-07
^{210}Po	138 d	2.6E-05	1.6E-06	1.2E-06

*lactation age-babies without breast feeding

In order to assess the health risk for population, the annual effective dose equivalent associated with radiation exposure through ingestion of the all water samples was estimated with the formula:

$$D_{ef} = \Sigma(A_Y \times IR_W \times CF) \qquad (5)$$

where Def is the annual effective dose equivalent (Sv y^{-1}), A_y – the activity concentration of radionuclide Y (Y=^{210}Po, ^{210}Pb, ^{226}Ra) (Bq L^{-1}), IRw –water intake by one person during one year (L y^{-1}), CF – effective dose conversion factor (Sv Bq^{-1}), given in Table 2 for each age category of population. The annual consumption rate was estimated at the value of 730 L for adults, 350 L for children and 250 L for lactation age, according to WHO 2011 [1]. The results obtained by us for the annual effective dose equivalent due to ingestion of ^{210}Po ^{210}Pb and ^{226}Ra radionuclides from drinking water are presented in Table 3.

Table 3. The annual effective dose equivalent due to ingestion of ^{210}Po, ^{210}Pb, and ^{226}Ra from drinking water

Sample code	Persons	Efective dose due to ingestion of ^{210}Po (μSv y^{-1})			Efective dose due to ingestion of ^{210}Pb (μSv y^{-1})			Efective dose due to ingestion of ^{226}Ra (μSv y^{-1})			Total effective dose (μSv y^{-1})		
		lactation age	children	adults	lactation age	children	adults	lactation age	children	adults	lactation age	children	adults
DW1	260000	12.68 ±2.79	1.09 ±0.2	0.17 ±0.04	6.83 ±1.57	2.16 ±0.50	1.64 ±0.38	35.25 ±10.58	2.94 ±0.88	6.30 ±1.89	54.75 ±14.93	20.56 ±1.62	8.11 ±2.30
DW2	104000	12.35 ±2.72	1.06 ±0.23	0.17 ±0.04	6.76 ±1.55	2.14 ±0.49	1.62 ±0.37	27.03 ±8.11	2.25 ±0.68	4.83 ±1.45	46.14 ±12.38	5.46 ±1.40	6.62 ±1.86
DW3	28000	12.87 ±2.83	1.11 ±0.2	0.17 ±0.04	11.08 ±2.55	3.51 ±0.81	2.66 ±0.61	15.28 ±4.58	1.27 ±0.38	2.73 ±0.82	39.22 ±9.96	5.89 ±1.43	5.56 ±1.47
DW4	1905	77.94 ±17.15	6.71 ±1.48	1.05 ±0.23	17.69 ±3.54	5.60 ±1.12	4.24 ±0.85	31.73 ±9.52	2.65 ±0.79	5.67 ±1.70	127.35 ±30.20	14.96 ±3.39	10.96 ±2.78
DW5	4425	79.06 ±17.39	6.81 ±1.50	1.07 ±0.23	7.01 ±1.61	2.22 ±0.51	1.68 ±0.39	23.50 ±7.05	1.96 ±0.59	4.20 ±1.26	109.57 ±26.06	10.99 ±2.60	6.95 ±1.88
DW6	669	13.07 ±2.87	1.13 ±0.25	0.18 ±0.04	24.65 ±5.67	7.80 ±1.79	5.91 ±1.36	21.15 ±6.35	1.76 ±0.53	3.78 ±1.13	58.86 ±14.89	10.69 ±2.57	9.87 ±2.53
DW7	345	81.12 ±17.85	6.99 ±1.54	1.09 ±0.24	7.88 ±1.81	2.49 ±0.57	1.89 ±0.43	31.73 ±9.52	2.65 ±0.79	5.67 ±1.70	120.72 ±29.17	12.13 ±2.90	8.65 ±2.38
DW8	750	15.44 ±3.40	1.33 ±0.29	0.21 ±0.05	15.72 ±3.14	4.98 ±1.00	3.77 ±0.75	23.50 ±7.05	1.96 ±0.59	4.20 ±1.26	54.66 ±13.59	8.27 ±1.88	8.18 ±2.06
DW9	600	16.54 ±3.64	1.42 ±0.31	0.22 ±0.05	11.07 ±2.32	3.51 ±0.74	2.66 ±0.56	31.73 ±9.52	2.65 ±0.79	5.67 ±1.70	59.33 ±15.48	7.58 ±1.84	8.55 ±2.31
DW10	771	12.94 ±2.85	1.11 ±0.25	0.17 ±0.04	8.04 ±1.69	2.55 ±0.53	1.93 ±0.40	27.03 ±8.11	2.25 ±0.68	4.83 ±1.45	48.00 ±12.64	5.91 ±1.46	6.93 ±1.89
DW11	825	50.73 ±11.16	4.37 ±0.96	0.68 ±0.15	16.08 ±3.22	5.09 ±1.02	3.86 ±0.77	9.40 ±2.82	0.78 ±0.24	1.68 ±0.50	76.21 ±17.20	10.25 ±2.21	6.22 ±1.43
DW12	18432	18.28 4.02	1.58 ±0.35	0.25 ±0.05	6.93 ±1.46	2.19 ±0.46	1.66 ±0.35	9.40 ±2.82	0.78 ±0.24	1.68 ±0.50	34.61 ±8.30	4.55 ±1.04	3.59 ±0.91
DW13	98646	12.68 ±2.79	1.09 ±0.24	0.17 ±0.04	13.22 ±3.04	4.19 ±0.96	3.17 ±0.73	9.40 ±2.82	0.78 ±0.24	1.68 ±0.50	35.29 ±8.65	6.06 ±1.44	5.02 ±1.27
DW14	8566	12.35 ±2.72	1.06 ±0.23	0.17 ±0.04	7.14 ±1.50	2.26 ±0.47	1.71 ±0.36	17.63 ±5.29	1.47 ±0.44	3.15 ±0.95	37.12 ±9.50	4.80 ±1.15	5.03 ±1.34
DW15	8735	12.87 ±2.83	1.11 ±0.24	0.17 ±0.04	9.20 ±2.11	2.91 ±0.67	2.21 ±0.51	28.20 ±8.46	2.35 ±0.71	5.04 ±1.51	50.27 ±13.41	6.37 ±1.62	7.42 ±2.06
DW16	12667	12.87 ±2.83	1.11 ±0.24	0.17 ±0.04	6.74 ±0.10	2.13 ±0.03	1.62 ±0.02	31.73 ±9.52	2.65 ±0.79	5.67 ±1.70	51.34 ±12.45	5.89 ±1.07	7.46 ±1.76
DW17	6789	28.93 ±6.36	2.49 ±0.55	0.39 ±0.09	25.89 ±7.67	8.20 ±2.43	6.21 ±1.84	16.45 ±4.94	1.37 ±0.41	2.94 ±0.88	78.77 ±18.96	14.44 ±3.39	11.34 ±2.81

The annual effective dose equivalent associated with radiation exposure through water intake allows assessing the health risk for adult, children and baby members of the population. The results from this study (Table 3) indicated that the annual effective dose equivalents are lower than the WHO recommended level of 100 μSv y^{-1} for children and adults, while the highest values have been found for lactation age (for the sample DW4, Dw5 and DW7 from Galati). As expected, the highest values of the effective dose equivalent correspond to the samples having the largest activity concentration of ^{210}Po. The main reason is its higher conversion factor. The annual effective dose equivalent has an average value of 63.79±15.75 μSv y^{-1} for babies, 8.26±1.94 μSv y^{-1} for children and 7.44±1.93 μSv y^{-1} for adults.

4. CONCLUSIONS

This is the first detailed study of radionuclide concentration in drinking water from Galati and Vrancea areas, Romania. Also, it is the first study regarding the assessment of the annual effective dose equivalent due to the drinking water consumption in this area. This was achieved as a result of the new requirements according to the Law 301/2015. Provisions of the European Directive 51/2013 were transported into Romanian national legislation through the Law 301/2015 [17] by establishing health protection requirements regarding the radioactive substances in drinking water.

The gross alpha, gross beta, ^{210}Po, ^{210}Pb and ^{226}Ra activity concentrations of drinking water in 2015 in the most important waterworks from Galati and Vrancea areas, Romania, were measured. The activity concentration data obtained in this study are lower than the guidance level according to World Health Organization and nationals rules.

The values for the annual effective dose equivalent due to the consumption of drinking water have been found below 100 μSv y^{-1}, except for babies (without breast-feeding). These values correspond to the samples DW4, DW5, DW7 (notwithstanding that the activity concentration of the investigated radionuclides had values below maximum concentrations allowed).

This study highlights the fact that although the values of activity concentrations for the investigated radionuclides are lower than the maximum allowed concentration, the recommended annual effective dose (100 μSv y^{-1}) can be exceeded. It is necessary to monitor activity concentrations of radionuclides in drinking water (from Romania and generally), especially for the radionuclides with the highest effective dose conversion factor in order to assess the equivalent annual effective dose.

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REFERENCES

1. United Nations Scientific Committee on the Effect of Atomic Radiation (UNSCEAR) Sources and effects of ionizing radiation United Nations (2000).
2. World Health Organization (WHO). Guidelines for drinking water quality, WHO (2011).
3. Council Directive 2013/51 Euratom laying down requirements for the protection of the health of the general public with regard to radioactive substances in water intended for human consumption.
4. G.M. Stewart, S. W. Fowler and S.W. Fisher. "The bioaccumulation of U- and Th-series radionuclides in marine organisms", *Radioactivity in the Environment,* vol. 13, pp. 269-305, 2008.
5. J. Molinari and W.J. Snodgrass, "The chemistry and radiochemistry of radium and the other uranium and thorium natural decay series," *The Environmental behavior of radium,* IAEA 1, pp.1-56, 1990.
6. Guidelines for drinking-water quality-Third edition incorporating the first and second addenda, Vol. 1, Recommendation World Health Organization, 2008.
7. The Council of Galati County Retrieved from: http://www.cjgalati.ro/index.php/judeul-galai
8. The Council of Vrancea County Retrieved from: http://www.cjvrancea.ro/files/file/Strategia%20de%20dezvoltare%20a%20jud/hot177anexa.pdf
9. *Water quality - Measurement of gross alpha activity in non-saline water – Thick source method*, ISO 9697:2007
10. *Water quality - Measurement of gross beta activity in non-saline water – Thick source method*, ISO 9696:2008
11. *Operating manual technical documentation low background total alpha and beta global - alpha measuring system type Protean-PIC.* MPC-2000-DP.
12. *Water. The analysis of Po-210 concentration in water.* STAS 12444, 86.
13. *The methodological book Radiation Hygiene*, Ministry of Health, 1981
14. *Operating manual technical documentation of module SARAD* RTM-1688-2.
15. *Drinking water*. STAS 1342, 1991
16. *Norme Fundamentale de Securitate Radiologica* Retrieved from: http://www.cncan.ro/assets/NSR/nsr01.pdf
17. Law 301/2015 by establishing health protection requirements regarding the radioactive substances in drinking water, Monitorul Official, 2015.