Carpathian Journal of Earth and Environmental Sciences, August 2016, Vol. 11, No 2, p. 575 - 582

NATURAL RADIOACTIVITY OF WATER SOURCES RELATED TO GEOLOGY AND RADIATION EXPOSURE IN ALBA COUNTY

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Abstract: Alba County is defined by a complex geological structure, having pegmatite and gnais formations that can be associated to large radon concentrations. Radon and radium concentrations (^{226}Ra and ^{222}Rn) were measured in various water sources: springs, wells, rivers and public water supply network. The water samples were collected from six places in the county of Alba (Romania). The method used to find radon and radium values were the scintillation method, based on Lucas Cells. The radon concentrations found varied between 11.51 and 22 Bq/L for the well water, from 1.6 to 2.13 Bq/L for the surface water, between 19.34 and 24 Bq/L for spring water, and from 0.9 to 1.66 Bq/L in the case of public water network source, respectively. The values of the measurements performed during this study indicate that the local population is not exposed to additional natural irradiation.

Key words: radon, radium, Lucas Cells, dose, water.

1. INTRODUCTION

The most indispensable for life and the wide spread matter on the planet, water, and must be free of chemical, microbiological and radiological pollutants.

The European Commission Drinking Water Directive (98/83/EC) does not give maximum activity concentrations for the individual nuclides except tritium. It sets a maximum effective dose of 0.1 mSv/y from ingestion of public water supply. The radon and its daughters are exccepted from the calculation of this maximum effective dose. Commission Recommendation 2001/928/Euratom proposes maximum concentration values for radon (100 Bq/L) and its long lived daughters (^{210}Po: 0.1 Bq/L and ^{210}Pb: 0.2 Bq/L). For water supplied with radon concentrations above 100 Bq/L, Member States should set a reference level for the radon to be used considering also whether remedial action is needed to protect human health. A level higher than 100 Bq/L may be adopted if national surveys show that this is necessary for implementing a practical radon programme. For concentrations in excess of 1 000 Bq/L, remedial action is deemed to be justified on radiological protection grounds.

Human body is exposed to radon by inhaling (Cosma et al., 2009; Cucos-Dinu et al., 2012; Sainz et al., 2009; Cosma et al., 2014) and ingestion. Radon is readily released from surface water; consequently, groundwater contains potentially much higher concentrations of radon than surface water.

It should be noted that radon activity concentrations in surface waters is low, usually below 1 Bq/L (EC, 2001; Ryan et al., 2003). Concentrations in ground water vary from 1 to 50 Bq/L for rock aquifers in sedimentary rocks, to 10 to 300 Bq/L for wells dug in soil, and to 100 Bq/L to 50 000 Bq/L in crystalline rocks. The highest concentrations are usually associated with high uranium concentrations in the bedrock. A characteristic of radon concentrations in rock aquifers is their variability; within a region with fairly uniform rock types, some wells exhibit concentrations far above the average for that region (EU, 2001).

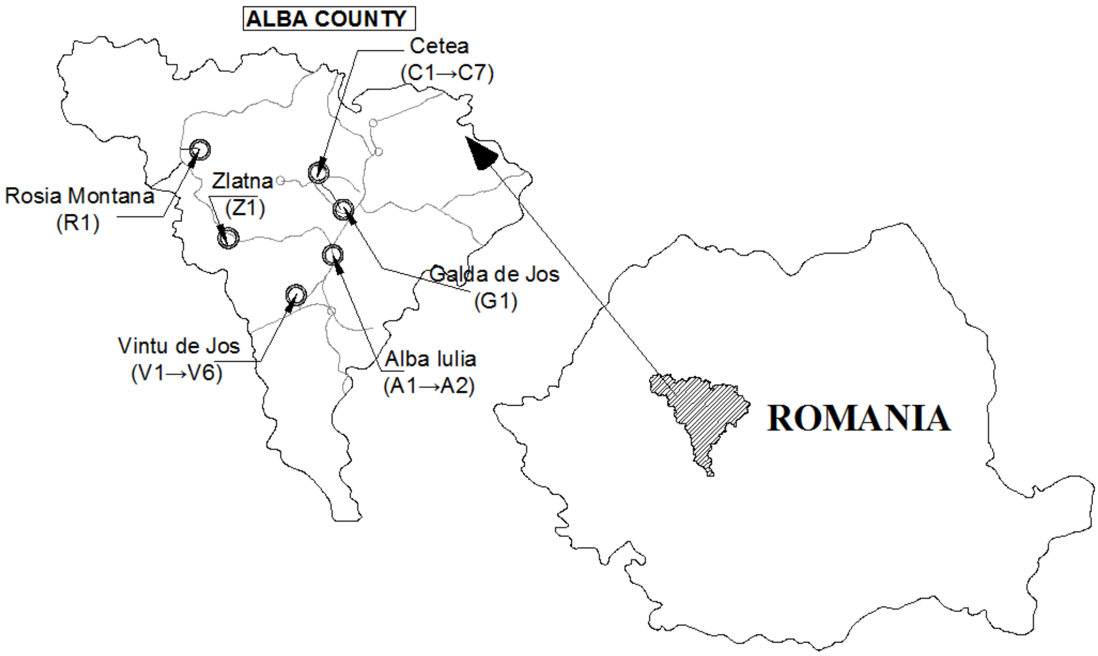

Figure 1. Map with locations of water sampling

During the present research, uranium radionuclides were measured, namely radium and radon, in samples collected from various areas in the Alba County (Fig. 1). The radium and radon concentration was measured in waters originating in various sources, such as: surface water, spring water, well water, and public supply water.

2. GEOLOGICAL BACKGROUND

In studied areas are exposed several formations belonging from a structural viewpoint to Apuseni Mts. and Transylvanian Basin.

Near by the contact between Apuseni Mts. and Transylvanian Basin are placed Galda de Jos and Vintu de Jos areas. Stratigraphic sequence consists of crystalline rocks basement and sedimentary deposits belong to the post-Laramide sedimentary cover which marks the beginning of the Uppermost Cretaceous-Paleogene Transylvanian Basin evolution (Balintoni et al., 1998). The sedimentary sequence begins with a Cretaceous marin facies succession with sandstone and silt (flysch), followed by clastic coast environment sediments (yellowish silty sandstone with polygenetic conglomerate and silts interbedded). The uppermost part of Cretaceous consists of red clastic continental facies represented by cross bedded sand and conglomerates (fluvial system environment). The entire succession after that (Uppermost Eocene, Oligocene and Lower Miocene) is represented by alternating marine brackish and lacustrine sediments (limestone, sandstone, quartz sand) with continental redbeds dominated by silt and fine sand, with a lower proportion of the channel filling rough clasts in relation to first red clastic series. The marine Lower Badenian succession covers this pile of continental deposits (Codrea & Dica, 2005). At the upper part of sedimentary sequence are disposed Pannonian deposits, consisting of marls and sands, with several levels of volcanic tuffs. On the surface, the Pannonian layer is covered by the most recent Quaternary (Pleistocene and Holocen) age alluvial deposits.

For Vintu de Jos area we have to mention that, Pian Creek, tributary to Mures River, from where the water samples were taken, drains from the north-eastern part of Sureanu Mts. The basement of the region structurally belongs to the Getic domain and consists of metamorphic rocks (meso- and epimetamorphic schists, migmatites, gneisses, paragneisses, quartzites, micaschists, amphibolites, quartzitic schists, quartzitic-sericitic schists). The magmatic formations consist of granites, quartziferous porphyries and pegmatites also occur. The sedimentary formations were transgressively overlaid on the crystalline basement and they belong to the Upper Cretaceous, the Neogene and the Quaternary (Bedelean & Bedelean, 2001).

Cetea area is placed in the Trascau Mts., geologically, in this area are exposed Mesozoic deposits consisting of Upper Jurassic Stramberk Limestone and alpine ophiolithes. Cretaceous sediments (conglomerates, sandstones, marls) are surrounded by the Jurassic rocks (Telbisz et al., 2014). These formations lay as tectonic nappes on the Proterozoic-Paleozoic crystalline basement of Apuseni Mts. In limestone deposits, the karstification processes were carried on by the percolation water from the surface and a lot of caves and other karstic forms were created.

In the Zlatna area, the main feature of the local geology is the presence of Lower Cretaceous limestones and conglomerates, above an orphiolitic complex from the Lower Jurassic (Pope et al., 2004).

The basement of Roșia Montană consists of pre-Mesozoic metamorphic rocks covered by Cretaceous and Tertiary sedimentary deposits. The Cretaceous deposits, predominantly in a flysch facies, are covering most of the area. The whole pile of rocks has been intruded by Tertiary magmatites that occur as volcanic and sub-volcanic bodies, placed along three parallel north-west trending lineaments (Lazăr et al., 2014).

3. MATERIAL AND METHODS

3.1. Measurement sites

Water samples were collected from five areas of the Alba County (Alba Iulia, Vintu de Jos, Galda de Jos, Zlatna, Cetea and Rosia Montana), from different waters sources (wells, springs, rivers and public supplies) (Fig. 1). The coordinates of entire sampling area range between 46 degrees 18 minutes and 45 degrees 59 minutes North latitude, respectively 23 degrees 7 minutes and 23 degrees 34 minutes East latitude. Water sampling was done from September 2011 to November 2011.

3.2. Water radon and radium concentration measuring procedure

The water samples were collected in glass bottle of 0.5l, fully filled and tightly sealed and transported to the laboratory for measurement purposes. The time interval between sampling and measurement was of maximum 48 hours and in measuring the radon concentration the half time was considered and corrections were made accordingly.

The radon in water was found with the help of the LUK-VR system (Fig. 2) which includes the LUK 3A device and the scrubber for radon extraction from water.

Samples were carefully introduced in the scrubber (Plch, 2012), a glass container provided with two outlets at the top. The first outlet is a glass tube going up to the bottom of the scrubber, while the second outlet leads to a rubber tube connected to a stopcock. After the water sample was introduced in the scrubber, it was then stirred for about 1 minute so that the radon in water is brought into balance with the radon in the air at the top of the scrubber.

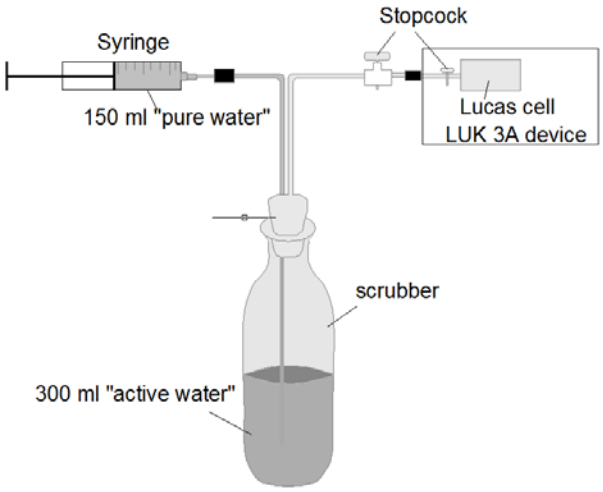

Figure 2. LUK-VR system

Before the sample is introduced, the Lucas Cell is emptied with a hand pump. The first outlet is connected to the Janet syringe, filled with 150 ml distilled water; the second outlet is connected to the LUK 3C device. The air containing radon is transferred to the Lucas Cell inside LUK 3A by successively turning on the tap of the LUK 3A device and of the one on the rubber tube. When air is transferred in the cell, water is also moved from the Janet syringe into the scrubber. This amount of water forces the active air to enter the Lucas Cell without suffering any dilution (Moldovan et al., 2013). Then, air is moved to Lucas cell, the tap of the LUK 3A device is turned off and measurement starts. The measuring method is detailed by Cosma et al., 2008 and Encian et al., 2013. Radon concentration is calculated with the equation given by Plch, 2012:

$$A(Bq / L) = 7.6 \times N(c / s) \quad (1)$$

where: A is the concentration of radon from the water (Bq/L) and N is the number of impulses per 100 seconds.

In order to calculate the radium in the water, the same steps are passed through as in the case of the radon; the only difference consists in the fact that the samples are kept in the laboratory at room temperature for 30 days, an interval in which water radon activity comes in secular balance with radium activity and thus, one can state that radium concentration is equal to measured radon concentration.

4. RESULTS AND DISCUSSION

4.1. Radon and radium measurements

The number of samples under investigation for measuring radon and radium concentrations was 18, water sources being: spring, well, river and public water supply. The samples were taken from six areas in the Alba County, as follows: two samples from Alba Iulia (A1, A2), seven samples from the village of Cetea (C1...C7), six samples from the commune Vintu de Jos (V1 to V7) and one sample from Rosia Montana, Zlatna and Galda de Jos village (Fig. 1).

The average of radon concentration from the samples originating from underground/deep water (wells and springs) was 19.05 Bq/L, and 1.57 Bq/L respectively for samples from surface sources (river and public network) (Table 1). One can see that samples taken from the Vintu de Jos area (V2, V3, V4) present values over those taken from the commune Galda de Jos (C1, C2, C3, C4, C5, G1) underground/deep water (wells and springs).

Cosma et al., (2008) calculated the radon concentration in surface waters from Transylvania area, such as rivers, lakes and creeks, reaching values in the range 0.5-10 Bq/L and an average of 2.6 Bq/L (Table 2). One can also appreciate that the average found in the present research, namely 1.57 Bq/L, comes in agreement with the value obtained by Cosma et al., (2008), and is smaller than the value of the mentioned team. The value range found in the present study agrees with other studies in the world, included in table 2.

In our research, a radon concentration ranging between 19.34 and 24.00 Bq/L was obtained for spring waters, One can find out that the values reached are higher than the values of Ali et al., (2010), Baykara & Dogru (2006), Al-Bataina et al., (1997) in Pakistan, Turkey and Jordan respectively, but the maximum value (24 Bq/L) is smaller than the value found in Romania by Cosma et al., (2008), of 129.30 Bq/L, and in Venezuela by Horvath et al., (2000) respectively.

Table 1. Statistics of results

No.	Source	No. of samples		Range		Average value	
		Rn	Ra	Rn [Bq/L]	Ra [mBq/L]	Rn [Bq/L]	Ra [mBq/L]
1	well	7	-	11.51-22.83	-	16.44	-
2	spring	2	-	19.34-24.00	-	21.67	-
3	drinking water	2	-	0.90-1.66	-	1.28	-
4	river	2	5	1.6-2.13	16.3-118	1.87	43.52

Table 2. Comparison of radon concentration in various types of waters with previous measurements from other countries

Country	**Range (Bq/L)**	**Reference**
Well water		
Saudi Arabia	0.89-35.44	Alabdula'Aly, (1999)
Turkey	1.42-53.64	Akar et al., (2012a)
Mexico	1.78-39.75	Villalba, et al., (2005)
China	6.00-127.00	Xinwei (2006)
Austria	1.46-118.70	Wallner & Steininger (2007)
Romania	0.60-112.60	Cosma et al., (2008)
Romania	11.51-22.83	Present work
Public water supply		
Turkey	0.91-12.58	Akar et al., (2012b)
Brazil	0.39-0.47	Marques, et al., (2004)
Jordan	2.50-4.70	Al-Bataina, et al., (1997)
China	8.00-18.00	Xinwei (2006)
Italy	3.90-6.90	Marques, et al., (2004)
Saudi Arabia	0.90-2.10	Tayyeb, et al., (1998)
Romania	0.90-1.66	Present work
Spring water		
Pakistan	1.60-10.20	Ali, et al., (2010)
Turkey	0.13-0.90	Baykara & Dogru (2006)
Jordan	3.30-10.70	Al-Bataina, et al., (1997)
Venezuela	0.10-576.00	Horvath, et al., (2000)
Romania	2.00-129.30	Cosma et al., (2008)
Romania	19.34-24.00	Present work
River water		
England	0.08-1.17	Al-Masri & Blackburn (1999)
Brazil	0.43-2.40	Marques et al., (2004)
Turkey	1.26	Baykara & Dogru (2006)
Romania	0.50-10.00	Cosma et al., (2008)
Romania	1.60-2.13	Present work

In public water supply, the values in the present study are close to the interval determined by Tayyeb et al., (1998) in Saudi Arabia, but much smaller than the values found in China (Xinwei 2006).

In underground water taken from wells, the interval of values 11.51-22.83 Bq/L agrees with studies performed both in Romania (Cosma et al., 2008) and in other countries in the world. However, the upper limit found in these investigations (22.83 Bq/L) is much smaller than the other studies, as one can see in table 2.

Radium concentration was found only in samples originating in surface water (river), the average for these river sourced samples being 43.52 mBq/L, with a maximum value of 118 mBq/L for the sample taken from the Zlatna town, respectively a minimum value of 16.3 mBq/L for samples collected from the Rosia Montana town and Vintu de Jos village (Table 3).

Romanian legislation does not contain standards for radon in drinking water but the value recommended by the World Health Organization (WHO, 2008) in drinking water for public water supplies is 100 Bq/L, and the maximum contaminant level (MCL=11.1 Bq/L) proposed by United States Environmental Protection Agency for radon level in drinking water (US.EPA, 1991).

Radon is a very mobile gas, it can easily move through fractures and openings in rocks and into the pore spaces in an aquifer or soil. The amount of radon in ground water is related to several factors, including the amount of uranium available in the source rocks, the location of the radium atom in the mineral grain (how close it is to the grain's surface), and the physical properties of the aquifer materials, such as porosity. Thus, it can be expected vary values of this radioactive gas in different types of waters according to the previous mentioned factors.

Table 3 resumes a relation between geology and radon, respectivelly radium concentrations from water samples presented in our study. Due to the small number of samples for Alba Iulia, Roșia Montană and Zlatna areas and the lack of radon values for water sources like rivers in these areas, a correlation between natural radioactivity and geology could not be provided. Consequently, the discussed Alba County areas from geology and radon concentration relation perspective will be: Galda de Jos, Vintu de Jos and Cetea.

The highest radon concentration defines both well and spring waters in the area of Vintu de Jos village. This can be explained by the presence of thick detrital sedimentary sequences forming the area substrate. The sediments were carried by fluvial systems with high discharge from the source areas, probably to a wide extent over the whole area of the present-day Apuseni Mountains or even farther (Codrea et al., 2010). Therefore, we could state that lithologically, there is much heterogeneity.

Table 3. Radon and radium concentrations. Acronyms A, C, R, G, V, and Z indicate the source area of the sample, namely Alba-Iulia, Cetea, Rosia Montana, Galda de Jos, Vintu de Jos and Zlatna

No./sample code	Source type	^{222}Rn concentration (Bq/L)	^{226}Ra concentration (mBq/L)	Relation with geology
1/A1	public supply	1.66	-	Unheeded
2/A2	river	-	40	
3/C1	well	15	-	Relatively low radon sources rocks (dominantly limestones and ophiolithes) Proterozoic-Paleozoic crystalline basement
4/C2	well	13.26	-	
5/C3	well	11.51	-	
6/C4	well	14.54	-	
7/C5	well	16.47	-	
8/C6	river	1.60	-	
9/C7	river	-	27	
10/G1	spring	19.34	-	Fluvial sandstones and fluvial sediments (detritic sedimentary sequences) that could contain radioactive minerals
11/R1	river	-	16.30	Unheeded
12/V1	public supply	0.90	-	Metamorphic rocks of granitic composition gneisses, paragneisses, quartzites, micaschists, amphibolites, quartzitic schists, quartzitic-sericitic schists). Magmatic formations consist of granites, quartziferous porphyries and pegmatites
13/V2	well	22.83	-	
14/V3	spring	24	-	
15/V4	well	21.44	-	
16/V5	river	2.13	-	
17/V6	river	-	16.30	
18/Z1	river	-	118	Unheeded

Taking into account the above mentioned aspect, the detrital deposits have more significant radiogenic potential than limestone rocks, both widespread in Apuseni Mts. In addition, the magmatic formations consist of granites, quartziferous porphyries and pegmatites also occurring in this area, generally sources of high radon, could be the cause of relatively high value of radon in spring water.

But, in the same time, the rocks that contain significant proportions of flaky minerals, usually oriented in rock mass, such micaschists, limit the vertical movement of radon (Bican-Brișan et al., 2013) and consequently, its entering into groundwater. Thereby, in the basement of our areas with these types of rocks is created a balance in terms of generation / migration of radon.

Taking into account the above mention aspects, in waters from Vințu de Jos and Galda de Jos areas the radon concentrations are higher than in the Cetea area where prevails relatively poor radon source rocks (dominantly limestones and ophiolithes).

4.2. The effective dose

In the case of water taken from wells and springs, the effective dose received by the individuals consuming these waters was calculated. (Table 4).

The conversion factors recommended by three international organizations (UNSCEAR, 1993; NRCP, 1988; ICRP, 1993) were used. The annual effective dose taking into account Rn-222 concentration was calculated according to the following formula (Somlai et al., 2007; Todorovic et al., 2012):

$$E = K \times C \times KM \times t \qquad (2)$$

where: **E** is the effective dose from ^{222}Rn ingestion (Sv), **K** is the ingesting dose conversion factor of Rn-222 ($SvBq^{-1}$), **C** is the concentration of ^{222}Rn (Bql^{-1}), **KM** is the water consumption (2 or 1 litre/day), **t** is the duration of consumption (365 days). For the dose calculation, a consumption of one litre/day for one adult or child drinking the same water and directly from the source point was assumed (Galán et al., 2004; Todorovic et al., 2012).

The effective dose does not exceed the value 1 mSv/year (UNSCEAR, 2000), and contributes to the annual dose with only 0.107 mSv/year in children, value obtained with a conversion factor recommended by UNSCEAR report ($2x10^{-8}$ $SvBq^{-1}$) or 0.374 mSv/year, when the conversion factor used is recommended by ICRP ($0.07x10^{-6}$ $SvBq^{-1}$).

In adults, with an average water consumption of 1 litre per day, the value of the calculated dose was 0.049 mSv/year calculated with the conversion factor recommended by UNSCEAR report (10^{-8} $SvBq^{-1}$), 0.019 mSv/year with the conversion factor recommended by NRCP ($0.35x10^{-8}$ $SvBq^{-1}$) or 0.053 mSv/year with a conversion factor recommended by ICRP ($0.01x10^{-6}$ $SvBq^{-1}$).

The values of the dose coming from drinking water can be compared to studies performed by other authors. Thus, Moldovan et al., (2013) determined the dose of radon in drinking water (wells and springs) using an average of 16.45 Bq/L for the Alba County and the conversion factors recommended by three international organizations (UNSCEAR, 1993; ICRP, 1993; NRCP, 1988). The effective doses obtained vary in the interval $2.1x10^{-2}$ mSv/year and $4.2x10^{-1}$ mSv/year (Moldovan et al., 2013; Cosma et al., 2008).

In the Southern area of Hungary, Somlai et al., (2007) appreciated that the effective dose due to public water supply consumption in adults does not reach the value 0.1mSv/year, taking into account a conversion factor of 10^{-8} $SvBq^{-1}$ and a water consumption of one litre/day (Somlai et al., 2007). In Spain, Galán et al., (2004) have determined the dose due to water consumption and their values range between $4.2x10^{-4}$ mSv/year and 3.3 mSv/year.

Table 4 The effective doses for the samples in Alba County

N0./simple code	Rn^{222} [mSv]*Effective dose				
	UNSCEAR, 1993, 2000		NRCP, 1988	ICRP, 1993	
	Children	Adults	Adults	Children	Adults
1/A1	0.012	0.006	0.002	0.042	0.006
3/C1	0.110	0.005	0.019	0.383	0.055
4/C2	0.097	0.048	0.017	0.338	0.048
5/C3	0.084	0.042	0.015	0.294	0.042
6/C4	0.106	0.053	0.019	0.371	0.053
7/C5	0.120	0.060	0.021	0.421	0.060
10/G1	0.141	0.070	0.025	0.494	0.071
12/V1	0.006	0.003	0.001	0.023	0.003
13/V2	0.167	0.084	0.029	0.583	0.083
14/V3	0.175	0.088	0.031	0.613	0.088
15/V4	0.157	0.079	0.027	0.55	0.078

*values were calculated for an average water consumption of 1 litre per day

5. CONCLUSIONS

In this paper, are presented the results for the ^{226}Ra and ^{222}Rn concentrations in the case of seven samples of well water, two samples of spring water, seven samples of river water and two samples of public supply water. Measurements were performed with the LUK-VR system, made up of the LUK 3A device and a scrubber.

The highest values were found in spring water samples, having an average of 21.67 Bq/L, and the smallest values were found in public water supply, with an average of 1.28 Bq/L, respectively. The arithmetic average of the radon concentrations in deep underground water (wells and springs) leads to the value 19.05 Bq/L, which is much higher than the value for surface waters (1.57 Bq/L). A relation between the natural radioactivity of public supplies and rivers is not taking into account.

Radon concentrations in wells and springs are different. The difference of radon concentrations found in deep waters could be explained by the different lithological content of the underground layer of water-bearing permeable rocks.

The high radon concentrations in Vintu de Jos area in springs and well water can be associated to the presence of gneisses, granites and pegmatites in the basement of area. Fluvial sandstones and fluvial sediments, widespread in stratigraphical sequence of Galda de Jos area could contain radioactive minerals. Relative low values of radon in the wells from Cetea area could be related to the relative low radon source rocks like limestones and ophiolithes that dominate the lithology of area.

The effective dose of the population exposure to radiation by consumption of water of the types mentioned, be they children or adults, was also calculated , but it did not exceed 1 mSv/an in any of the cases discussed.

As, in general, all the determined concentrations present values below the revised reference values of 100 and 146 Bq/L proposed by the European Union, and respectively The United States of America, it can be concluded that there is no reason to consider such water sources raise a problem regarding radon exposure by ingestion.

Small amounts of radon can also be released from the water supply into the air, especially if the water source is underground. As the radon moves from the water to air, it can be inhaled. Water that comes from deep, underground wells in rock may have higher levels of radon, whereas surface water (drawn from lakes or rivers) usually has very low radon levels. For the most part, water does not contribute much to overall exposure to radon.

Acknowledgments

This paper was supported by the project project PN-II-PT-PCCA-2011-3.2-1064 - Radon Map (residential, geogenic, water) for Center, West and North-West regions from Romania (RAMARO acronym)

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Received at: 16. 03. 2015
Revised at: 16. 06. 2016
Accepted for publication at: 23. 06. 2016
Published online at: 02. 07. 2016