

RESEARCH ON RADON (^{222}Rn) CONTENT IN DRINKING-WATER SAMPLES COLLECTED FROM SIBIU COUNTY

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Abstract

The isotope radon (^{222}Rn), an inert water-soluble gas that may contaminate water, represents a potential risk for human health, including cancer. The present study reports the evaluation of the ^{222}Rn concentration in drinking-water samples collected in 2020 from 10 sources located in Sibiu County, as measured using the pulse ionization chamber. Values of ^{222}Rn varied from 0.0549 to 37.4770 Bq/l, with an average of 4.586 Bq/l. These values were below the maximum level of 100 Bq/l recommended by WHO and EU Directive/Euratom. With the exception of the sample from groundwater, the others showed ^{222}Rn values below the maximum level of 11.1 Bq/l, as recommended by US EPA. In the case of impermeable soils, this radionuclide floats inside the cracks until it reaches the atmospheric air, so it is harmful to human health both in water and in the air. Evaluation of radon in water, in particular from groundwater sources becomes essential for the management of remedial solutions.

Keywords: drinking-water, pulse ionization detector, radon.

1. INTRODUCTION

Several radionuclides and their decay products have been of environmental concern, as such pollutants generated from natural or anthropogenic sources may contaminate water (Qin-Hong et al., 2010). Among radionuclides, radon, ^{222}Rn , represents a radioactive gas originating from the disintegration of radium, the latter being the decay product of uranium, ^{238}U . The half-life of ^{222}Rn is 3.82 days (Alirezazadeh et al., 2005). This element has been identified as the main source of natural radioactivity with short-term products of ^{238}U disintegration, including ^{214}Po , ^{218}Po , ^{214}Bi and ^{214}Pb (Alirezazadeh et al., 2005; Sathish et al., 2009; Richon et al., 2010; Binesh et al., 2012). Research studies reported evidence of carcinogenicity of the ^{222}Rn radioactive gas (Rafique et al., 2012), being responsible of the internal exposure in humans, according to the report of the International Commission on Radiation Protection (ICRP) (Negarestani et al., 2011). The effect of the inhaled radon on human health is lung

cancer, while that of the ingested radon through the digestive tract is gastric cancer (Alirezazadeh et al., 2005; Binesh et al., 2012; Rafique et al., 2012).

Radon is a dangerous gas contaminating soil, rocks and water, sources which may become a risk for human health. Of great concern is the presence of ^{222}Rn in water, which conducted to an increase of studies directed toward the evaluation of its level in different water sources, over the past two decades (Abdallah et al., 2007). The current methods applied for ^{222}Rn analysis include several techniques, based on (a) the gamma ray spectrometry (the so-called radiometric method); (b) the emanometry methodology; (c) liquid scintillation counting (LSC analysis) (ASRO-2020).

The aim of the present paper was to investigate the radon concentration in water sampled from different areas of the Sibiu County, Romania, in order to identify the nature of potential exposure to radon from water intended for human consumption, originating in different types of water sources, both from the surface and from the underground.

2. MATERIALS AND METHODS

Sampling procedure

A number of 10 water samples were collected, starting on February and ending on November 2020, from water supplies, as follows: Sibiu (98% surface water and 2% groundwater, originating from the Cibin river), Alma, Avrig (surface water originating the Avrig river), Dumbrăveni, Mediaș (surface water originating the Târnava Mare river), Cîsnădie and Sadu (surface water originating the Sadu river), Păltiniș (water from captured/ mixed springs), Săcel and Sibiul (water from ground wells), all zones being located in the Sibiu county.

Samples were collected in glass vessels and sealed until analysis. The maximum shelf life of samples collected in a glass container is one day, as recommended by SR ISO 5667-3. An aliquot of water was injected into the degassing vessel. Analysis was done immediately after removing the plug in order to minimize the escape of radon from the sample into the surrounding air.

Quantification of radon, ^{222}Rn

The AlphaGuard type DF 2000 system and the AquaKIT were used. Before the sample measurement, a cycle of measuring the existing air was allowed in the installation, with all the containers and tubes connected. The predefined measurement cycles into the device are of 1 minute and 10 minutes, respectively. The same kind of cycle was used, as for measuring the sample. The radon background in the empty installation was measured, followed by the sample measurement. The determination of radon concentration in water samples is based on the indication of the radon monitor. This measurement value is not yet the radon concentration in the measured sample, because the removed radon has been diluted. To quantify the dilution effect, the exact internal volume in the measurement setting (V_{system}) was determined. The amount of radon remaining in the sample was determined by using the distribution coefficient k , as shown in Figure 1.

The diffusion coefficient k indicates the radon concentration ratio in aqueous phase and the existing air volume, which is temperature-dependent. As temperature drops down, the amount of water-soluble radon increases. The diffusion coefficient k increases as the temperature decreases. The influence of the radon diffusion coefficient k is low in the temperature range 10-30° C. This is suitable as long as the samples are measured under normal conditions and leads to an uncertainty of less than 2%, being

therefore negligible for the error resulting from the sample processing. Temperature deviations of $\pm 10^\circ$ C would result in a measurement error of up to 7% if this factor is not used.

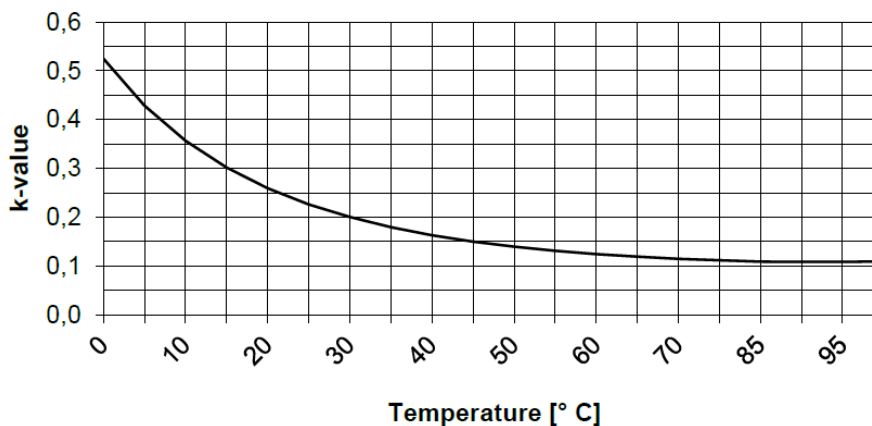


Figure 1. The temperature dependence of the diffusion coefficient k , for the water / air phase transition

The equation for the calculation of the radon concentration in the measured water sample is as follows:

$$C_{water} = \frac{C_{air} \times \left(\frac{V_{system} - V_{sample}}{V_{sample}} + k \right) - C_0 \times \left(\frac{V_{system} - V_{sample}}{V_{sample}} \right)}{1000}$$

where

C_{water} = radon concentration in water (Bq/l)

C_{air} = radon concentration in the air resulting from sparging water (Bq/l)

C_0 = background radon concentration (Bq/l)

V_{system} = internal volume of the measuring facility (ml)

V_{sample} = volume of the analyzed sample (ml)

k = diffusion coefficient

The results of ^{222}Rn concentration (C_{Rn}) are expressed in Becquerels per liter (Bq/l).

3. RESULTS AND DISCUSSIONS

The present study reports the evaluation of the concentration of ^{222}Rn in 10 samples of drinking-water collected during the months of February-November 2020 from different regional points of Sibiu County, Romania.

The results are presented in Table 1.

The lowest ^{222}Rn concentration (0.0549 Bq/l) was registered in the surface drinking-water collected from the Cisnădie city, while the highest one (37.4770 Bq/l) was recorded in the drinking-water sample from Păltiniș, which is a mountain resort situated at 1440 m at 32 km North of the Sibiu city. The Păltiniș water sample originated from the captured (mixed) springs, transported through supply pipes so

that it could then be filtered and disinfected. The groundwater in the mountainous region belongs mostly to the subregion corresponding to the crystalline schists. In the foothill region, significant quantities of groundwater are confined to greater depths in the piedmont deposits. Deep waters are present in the foothill region, where friable sedimentary deposits prevail (Raport de mediu, 2015). The sample of Păltiniș water was collected in November 2020 from the Cindrel Hotel network. The investigated level of the Păltiniș sample showed higher values than the other samples also due to the fact that it is a groundwater.

Table 1. Radon, ^{222}Rn , concentration in drinking-water samples collected from different sources of the Sibiu County, Romania (mean values \pm standard deviation)

Sample no.	Location	Radon (Bq/l)
1	Sibiu	0.1410 \pm 0.0221
2	Alma	0.4251 \pm 0.0547
3	Avrig	0.0890 \pm 0.0152
4	Dumbrăveni	1.1831 \pm 0.1385
5	Mediaș	0.1295 \pm 0.0201
6	Păltiniș	37.4770 \pm 2.5767
7	Săcel	3.9644 \pm 0.4076
8	Sibiul	2.3131 \pm 0.2168
9	Cisnădie	0.0549 \pm 0.0070
10	Sadu	0.0923 \pm 0.0175

The frequency distribution of ^{222}Rn concentration in drinking-water collected from the 10 areas of the Sibiu County is presented in Figure 2. The mean value of ^{222}Rn concentration in drinking-water within Sibiu County for the year 2020 was found to be 4.5860 Bq/l.

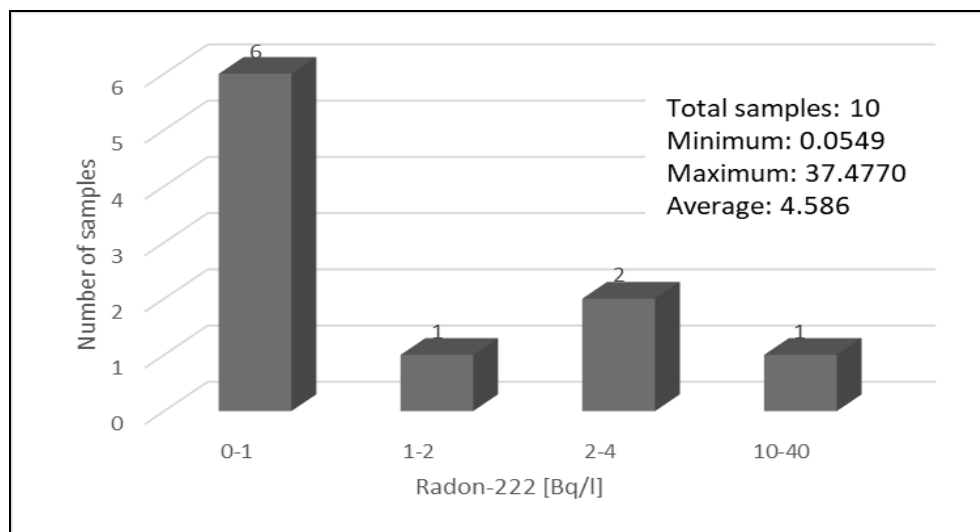


Figure 2. Frequency distribution of ^{222}Rn concentration in drinking-water in the Sibiu County

The EU Council Directive 2013/51/Euratom laying down requirements for the protection of the public health with regard to radioactive substances in water intended for human consumption set the parametric value of water for human consumption at 100 Bq/l (Council Directive, 2013). The WHO recommendation for maximum level of ^{222}Rn in drinking-water is 100 Bq/l (WHO, 2008) as well, while the US EPA established the maximum level at 11.1 Bq/l .

There are national standards regarding the water quality with respect to ^{222}Rn , as follows: (a) ISO 13164-2:2020 Water quality ^{222}Rn Part.2. Test method using gamma-ray spectrometry; (b) ISO 13164-3:2020 Water quality ^{222}Rn Part.1-3. Test method using emanometry, and (c) ISO 13164-4:2020 Water quality ^{222}Rn . Test method two - phase liquid scintillation counting (ASRO, 2020).

Table 2. Reported values of radon concentration in water samples collected from different world regions.

Type of water sample	^{222}Rn (Bq/l)	Region, country	Ref.
Drinking water	3.176-49.932	Shanono and Bagwai, Nigeria	Bello et al., 2020
Drinking water	<3	Novi Sad /Serbia	Todorovic et al., 2012
Surface water	<1	Slovenia	ISO 13164-3, 2013
Groundwaters	1-1000	La Garrotxa, Spain	ISO 13164-3, 2013
Groundwaters	3043	Mountains Sudety, Poland	Przylibski et al., 2014
Groundwaters	17-3856	Nisa, Portugal	Pereira et al., 2015
Unbottled mineral spring waters	1.4-105	South Catalonia, Spain	Fonollosa et al., 2016
Unbottled mineral spring waters	1.5-181	Euganean Thermal District, Padua, Italy	Cantaluppi et al., 2014
Fountain water	10-300	Norway	ISO 13164-3, 2013
Fountain water	4-63560	Stockholm, Sweden	Skeppstrom et al., 2006
Drinking water	2.96 ± 0.966	West Region of Iran	Pirsaheb et al., 2015
Drinking water	0.87-0.384	Bandar Abbas City, Iran	Fakhri and Oliveri et al., 2016
Tap water	0.46 ± 0.04	Minab City, Iran	Fakhri and Kargosha et al., 2016
Bottled water	0 – 901 (Bq/m ³)	Bandar Abbas City, Iran	Fakhri and Hossein et al., 2016
Groundwaters	0.168–78.509	Ijero, Ekiti State, Nigeria	Akinnagbe et al., 2018
Groundwaters	7.41 ± 2.04	Zaria, Nigeria	Garba et al., 2012
Groundwater	7.002 ± 0.33	Kaduna State, Nigeria	Kamba et al., 2016

According to the maximum levels admitted for water for human consumption, samples of drinking-water from Sibiu County showed values below those reported by the EU and WHO regulations. With regard to the US EPA recommendations, only the sample from Păltiniș mountain-resort exceeded the established values, while the others can be considered appropriate with values well below the

maximum admitted level. Studies have shown that radon concentration may decrease during processing, transport or storage of water (Gruber et al., 2009). However, evaluation of the radon concentration in different sources of water, in particular in groundwater, becomes essential for the potential detection of radon in drinking-water. In such cases, measurements for the reduction of radon concentration are required, as inhalation or ingestion of radon become harmful for human health. Table 2 indicates several values of ^{222}Rn concentration in water samples collected from different world regions.

4. CONCLUSIONS

The average concentration of radon, ^{222}Rn , determined in 10 water samples collected from different areas of the Sibiu County, in 2020, was 4.586 Bq/l, value which did not exceed the maximum level set by EPA and WHO regulations. Out of the total 10 samples, 6 samples were surface waters, and the rest were groundwaters and wells. The minimum value (0.0549 Bq/l) was found in the Cisnădie water sample, which is surface water. The water samples collected from Dumbrăveni, Săcel and Sibiel (groundwaters, from wells) contain radon in concentration of 1.1831 Bq/l, 3.9644 Bq/l, and 2.3131 Bq/l, respectively, being slightly higher than that of Cisnădie sample, but within the allowed limits. The maximum value of ^{222}Rn (37.4770 Bq/l) was found in the groundwater sample from Păltiniș area, concentration that exceeds the EPA allowed limits, but were within the WHO guideline set at 100 Bq/l. The recommendation is to use water storage tanks for aeration and radon removal from water samples showing high concentration of radon, in order to reduce the consequences of exposure to this radioactive gas. Water sources high in radon should be avoided for household use.

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