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Radiological Risk Assessment for Karstic Springs Used as Drinking Water in Rural Romania

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Abstract: Seasonal variations of the radon and radium activity concentrations in karstic water sources originating in karstic formations were investigated as part of a premiere systematic survey conducted in Romania. A database including a total of 228 drinking water samples collected from 30 distinct water sources adjacent to rural communities was compiled. The radon and radium activity concentrations for all seasons, assessed based on solid scintillation, ranged from 2.1 to 19.7 Bq/L and from 0.6 to 3.0 Bq/L, respectively. Overall, the detected radon and radium contents did not exceed the radioprotection standards recommended by national and European legislation. However, in at least one season, the measured values for 31% of the samples exceeded the 11.1 Bq/L maximum contaminant level for radon in drinking water recommended by the Environmental Protection Agency of the United States. The associated radiological risk, reported in terms of annual effective dose, was calculated to be between 9.8×10^{-6} and 6.0×10^{-5} mSv/y for radon and between 5.9×10^{-5} and 2.7×10^{-4} mSv/y for radium, which are considerably below the WHO (World Health Organization) guidelines at a value of 0.1 mSv/y.

Keywords: radon; radium; risk assessment; drinking water; karstic springs

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1. Introduction

Radium (^{226}Ra) occurs in virtually all types of rocks and soils, with its concentration being directly correlated with geological specificity. Radon (^{222}Rn) is a chemically inert and radioactive gas produced by the direct decay of ^{226}Ra. Once produced, radon infiltrates the interstitial fluids [1], where it can easily dissolve into groundwater [2]. ^{226}Ra and ^{222}Rn are two of the most common naturally occurring radionuclides in groundwater. The concentration of dissolved radon is dependent not only on the radium content of rocks but also on local factors such as groundwater chemistry, the presence of shear zones, soil porosity or the residence time of water in a specific aquifer [3–5]. Moreover, the flow of groundwater through soil and rocks containing radon and radium can lead to a concentration of radon activity in addition to that due to the original aquifer [6]. Once the groundwater reaches the surface, the radon content is almost entirely depleted [7,8] due to its radioactive decay and evasion to the atmosphere. Radon is both highly useful to humans and a carcinogen. Its presence in rocks, soil and water has greatly facilitated interdisciplinary research, such as studies on traceability in water discharge [9], fault zone detection [10], earthquake prediction by means of soil-radon [11] and groundwater-radon [12], fire detection [13] and urban pollution characterization [14]. On the other hand, its presence in living spaces and drinking water represents a major health hazard to both humans and non-human biota [15–18].

Large- and small-scale surveys conducted in Romania [19,20] pointed out that drinking water is often contaminated with radon, regardless of its origin. Water-soluble radon could

increase indoor radon activity, which is known to cause lung cancer [16] and has also been linked with induced stomach cancer [21]. Additionally, contamination of the public drinking water supply by chemical and biological pollutants occurs often in Romania according to scientific [22–24] and public records (National Institute of Public Health). One specific example is represented by the spring waters in karstic regions which originate from groundwater. These particular types of waters are known to be polluted by human activities on the surface and by bats that inhabit the water crosses in caves, both of which add to the problems caused by the inherent lack of mechanical filtering of karst waters. Groundwater flowing through karst is vulnerable to contamination due to concentrated flow with reduced transit time and little self-purification within the system. By studying karstic sources of water, Moldovan et al. [25] hypothesized that, in karstic systems with diffuse input (caves, inlets, ponors, etc.), rainfall enters underground and reaches the surface through springs within days to months, gathering potential contaminants from the surface, especially in rural areas with agricultural fields. Radon concentrations in water represent an important dosimetry issue [26]. The global average dose from the inhalation of radon from all sources is approximately 1 mSv/y [27], which is slightly lower than half the total natural radiation exposure of 2.4 mSv/y [28].

Although the spatiotemporal variability of dissolved radon has previously been investigated, the focus of studies has mostly been on groundwater discharge [8,29,30] while dose assessment based on aquifer rock type has attracted less interest. In this study, we aim to evaluate the seasonal and spatial variability of ^{222}Rn and ^{226}Ra isotopes in raw drinking water from karstic springs and their associated health risk added to an already existing biological risk.

2. Materials and Methods

A total of 30 springs of karstic origin, located throughout the Romanian territory (Figure 1), were selected for this study. All of the selected springs are used for domestic and agricultural purposes by their local rural communities.

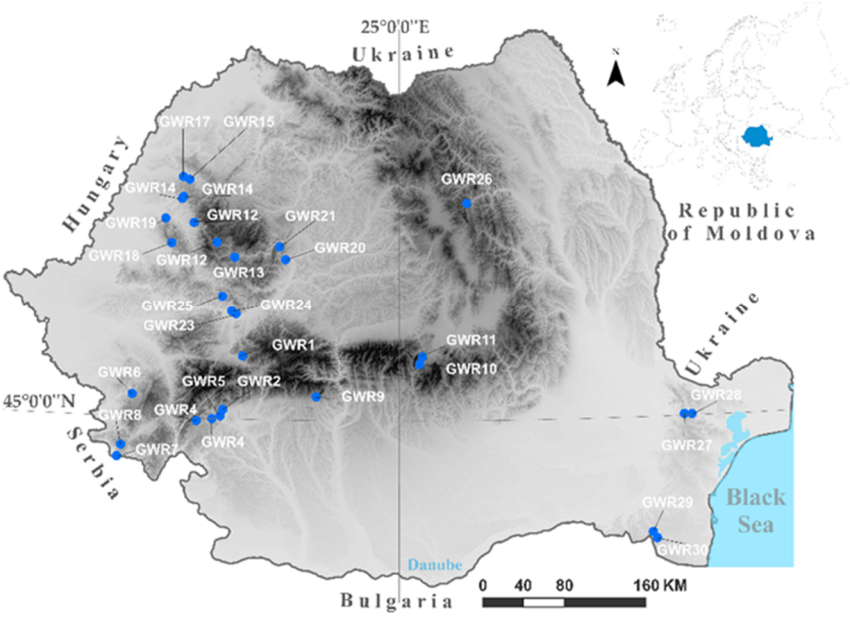

Figure 1. The sampled karstic springs in Romania (blue dots) and the position of the country in Europe (upper right).

2.1. Concentration of ^{222}Rn and ^{226}Ra

Spring water sampling was performed at 30 publicly accessed sources spread throughout Romania (Figure 1), hereafter delineated as sampling points GWR1 (GroundWater Radon) to GWR30. Due to a persistent drought, one of the springs (GWR4) could not be in

any of the field campaigns and, therefore, was excluded from our analysis. During the collection of autumn samples, GWR9 and GWR13 were also dry. Therefore, a total of 29 water samples were collected quarterly between October 2019 and August 2020. Glass bottles measuring 500 mL were filled from below the water flow and allowed to overflow vertically to avoid trapping air bubbles. The samples were then transported to the laboratory for analysis; the transport of samples did not require special conditions. The activity of ^{222}Rn was determined using the LUK VR installation (Jiri Plch, Prague, Czech Republic) shortly after sampling to avoid significant decay loss. The radon concentration was determined after the water samples reached room temperature, following the accredited protocol from LiRaCC [7]. At a room temperature of 20 °C, the solubility coefficient of radon in water is 0.254. The LUK VR installation consists of a glass vessel (scrubber) in which the radon in water is brought into equilibrium with the radon in the air above the water by vigorous stirring and the LUK 2P device. Air containing radon is transferred into a scintillation chamber (Lucas cell). The scintillation chamber is located inside the LUK 2P device in front of a photomultiplier. (Jiri Plch, Prague, Czech Republic) Scintillations produced by the active deposit of radon introduced into the chamber are detected and counted after establishing the radioactive equilibrium between radon and its active deposit, using the photomultiplier.

The preparation of samples for the measurement and determination of the radon concentration from water is described below:

Before the start of each measurement, the water samples were brought to room temperature. The sample (measuring 300 cm^3) was introduced into the scrubber, and then, the scrubber was closed and shaken very well for 1 min. It was then expected that equilibrium between the radon dissolved in the water sample and the radon in the air above it had been established. The air occupying the upper part of the scrubber (which has now become mixed with radon) was then transferred for measurement by the Lucas cell. The transfer process was conducted by using a syringe to add 150 cm^3 of distilled, radon-free water to the water sample in the scrubber, so that the level of water inside the scrubber increases and a similar volume of air is pushed out through the valve (which is open during this process) to fill the Lucas cell (145 cm^3). The Lucas cell was previously evacuated in order to be filled with the air now being forced in. A radon progeny filter was introduced between the scrubber and the Lucas cell. The Lucas cell was then connected to a photomultiplier tube for readout. The radon concentration in the water sample, C_{Rn} (Bq/L), and the number of collected counts, N (counts/s), are related as follows:

$$C_{Rn} = 7.6 \times N \text{ (count s}^{-1}\text{)}$$

Taking into account errors due to radon measurement and statistics, the minimum detection limit (MDL) was estimated to be 0.2 Bq/L. This value for the MDL is below the lowest expected radon concentration levels that are found in surface water (normally ~1 Bq/L). The statistical errors associated with the radon analysis were 5–6%, as explained by Cosma [7].

The efficiency of detection of the Lucas cell was determined using a control source manufactured and certified by the State Metrological Institute of the Czech Republic. A detailed description of the working protocol has been provided by Cosma [7]. Quality assurance has been achieved by participating in intercomparison exercises and radon-in-water proficiency tests, in both intercomparison exercises laboratory LiRaCC obtained satisfactory results [31].

^{226}Ra was measured based on its secular equilibrium with ^{222}Rn, which was achieved after the samples had been stored for 30 days in sealed columns, i.e., the radium concentration in the water was equal to the radon concentration 30 days after the sample had been collected. The radium in the water was measured using the LUK VR installation (described above) and following the same measuring protocol as in the radon determinations [32]. Considering all types of possible errors (statistic, measurements, etc.) usually sum up to

about 25%, the detection limit for the radium in water measurements was assessed to be 0.05 Bq/L [33].

The physicochemical parameters of spring discharge, water temperature and conductivity were measured in situ during each sampling (Table 1). The pH and electrical conductivity (EC) were measured in situ using a portable multiparameter monitoring device with built-in temperature correction (Multi 340i, WTW, Wertheim, Germany), and the flow rate was measured by using a FP111 Flow Probe (Global Water, Phoenix, AZ, USA).

Table 1. Temporal variability of the physicochemical parameters in the studied karstic springs.

Spring	Temperature (°C)				Electrical Conductivity (µS/cm)				Flow Rate (L/min)			
	Oct	Jan	May	Aug	Oct	Jan	May	Aug	Oct	Jan	May	Aug
GWR1	15.2	5.2	12.2	15.6	380	370	420	370	20	15	15	30
GWR2	13.5	10.2	11.1	14.1	500	460	390	450	480	1890	600	1800
GWR3	12.4	9.2	11.9	13.9	280	450	390	370	50	66	60	42.8
GWR5	9	8	9.4	11.3	420	420	440	440	3600	32,400	75,000	13,860
GWR6	12.6	11.6	12.6	13.4	710	730	710	690	25	60	36	60
GWR7	15.4	7	14.4	21.6	600	530	580	660	0.5	0.03	0.8	0.25
GWR8	11.5	8.5	10.3	13.1	640	560	500	590	30	120	90	20
GWR9	-	0	8.5	8.8	-	0	300	290	-	0	1.3	12
GWR10	10.3	0	7.8	11.8	440	0	420	460	0.5	0	2	3.75
GWR11	8.6	6.8	8.4	9.1	470	460	420	480	27.3	25	30	60
GWR12	11.7	9.5	11	11.7	390	250	290	380	2	7.5	60	60
GWR13	-	6.7	9.2	10.7	-	500	420	440	-	15	15	24
GWR14	13.5	10.2	12.5	13.5	460	440	520	480	5	2.7	6	10
GWR15	11.1	10.1	11.1	12	630	650	630	630	24	10	30	50
GWR16	10	8.8	10.3	11.5	510	480	500	560	7.5	6.6	15	15
GWR17	11.4	0	10.7	11.8	670	0	660	680	2.8	0	5	7
GWR18	29.6	29.1	29.2	30.3	250	250	250	250	300	300	600	150
GWR19	13.8	11.9	14.6	15.7	570	570	630	570	6.8	6	7.5	7.5
GWR20	13.3	10.9	12.8	15.1	1050	1030	1040	1080	10	10	10	15
GWR21	11.7	8.3	12	14	540	570	540	640	100	35	30	200
GWR22	10.9	8.5	10.3	11.1	330	320	360	330	10	6.6	7.5	10
GWR23	18.2	16.3	18.9	20.3	2160	2170	2230	2210	60	80	40	28
GWR24	20.7	20.2	21.9	24.2	1820	1790	1840	1830	100	60	90	100
GWR25	10	9.6	12.5	14.2	540	560	560	550	4	2.5	2	7.5
GWR26	9.2	0	10.5	9.4	540	0	520	590	2.7	0	3	12
GWR27	14	12.9	15.8	15.5	1270	1280	1280	1240	120	42	45	90
GWR28	14.2	14.9	15.3	15.5	1210	1210	1220	1210	200	120	200	200
GWR29	13.3	4.7	17.6	20.4	1470	1540	1490	1470	2.3	21	4	3.33
GWR30	13.5	10.5	13.2	15.5	1410	1390	1420	1390	12.5	6	12	10

2.2. Annual Effective Dose Due to Radon and Radium

The annual effective ingested doses were estimated according to the parameters introduced by the United Nations Scientific Committee on the Effects of Atomic Radiation [28] and by the International Atomic Energy Agency [34] and were calculated on the basis of the mean activity concentration using the following relation:

$$D_{water} = F \times C_w \times T \times A_w \times 10^3$$

- D_{water} (mSv/y) represents the annual effective dose of radon or radium ingestion;
- F is the committed effective dose per unit in water intake for adults, taken as 10^{-8} Sv/Bq for radon [28] and as 2.8×10^{-7} Sv/Bq for radium [34];
- C_w is the water consumption rate, taken to be 2 L/day;
- T is the duration of consumption (365 days);
- A_w is the activity of radon or radium in water (Bq/L).

2.3. Statistical Analysis

Statistical analysis of the data was performed using the OriginPro 2019b software, and a Principal Component Analysis (PCA) was performed using XLSTAT Addinsoft 2021. PCA was used for the analysis of the physicochemical and radon–radium variables in the studied springs. The statistical distribution of the data was evaluated using the Shapiro-Wilk test. For the comparison of the repeated measures, an ANOVA test with Tukey's post hoc analysis was used. In order to evaluate the intensity of the relationship between the examined variables, the Pearson correlation coefficient was calculated. The significance level α was set at 0.05.

3. Results and Discussions

3.1. Radon and Radium Concentrations

European and Romanian regulations set the admissible activity of radon in water at 100 Bq/L [35]. The US Environmental Protection Agency (EPA) [26] proposes a standard of 300 pCi/L (equivalent to 11.1 Bq/L) for radon contamination in drinking water from public water supplies.

In this study, the radon activity concentration in all seasons ranged from 2.1 to 19.7 Bq/L for the 29 active water sources (Table 2). The range of radon activity concentration is caused by specific particularities in groundwater dynamics due to the different permeability properties of the karst or other rocks crossed by the water.

Table 2. Temporal variability of the radon activity concentration in the studied karstic springs.

Sample	Radon Concentration (Bq/L)				
	Autumn	Winter	Spring	Summer	Annual Average
GWR1	2.1	2.3	4.2	2.2	2.7
GWR2	3.1	4.0	4.6	4.0	3.9
GWR3	2.4	5.1	6.6	5.1	4.8
GWR5	6.2	5.2	8.9	7.1	6.8
GWR6	4.1	4.5	4.3	4.8	4.4
GWR7	3.3	6.7	4.8	4.4	4.8
GWR8	7.2	7.1	7.1	8.5	7.5
GWR9	dried	5.2	8.1	7.8	7.1
GWR10	4.1	5.2	7.2	5.2	5.4
GWR11	6.2	4.0	7.6	5.7	5.9
GWR12	4.1	4.8	8.2	5.6	5.6
GWR13	dried	7.4	11.0	9.8	9.4
GWR14	10.9	11.6	11.0	9.5	10.8
GWR15	5.4	8.6	7.6	6.4	7.0
GWR16	11.5	10.8	13.6	12.6	12.1
GWR17	19.6	11.3	15.1	19.7	16.4
GWR18	17.0	10.8	17.3	19.0	16
GWR19	10.9	11.6	8.4	9.2	10.0
GWR20	16.1	13.6	17.5	17.5	16.2
GWR21	11.4	10.6	12.6	12.6	11.8
GWR22	5.0	4.0	6.5	4.9	5.1
GWR23	12.3	10.8	10.8	14.2	12.0
GWR24	6.9	9.6	6.1	7.8	7.6
GWR25	4.8	8.5	7.2	6.1	6.7
GWR26	2.7	3.7	6.1	4.0	4.1
GWR27	3.9	3.5	5.1	4.2	4.2
GWR28	8.1	9.7	11.6	6.4	9.0
GWR29	3.0	3.9	4.9	3.3	3.8
GWR30	5.5	6.2	6.4	5.2	5.8

The measured values are not significantly high and do not exceed the radioprotection standards recommended by national and European guidelines [35]. Radon data did, however, exceed the EPA limit (11.1 Bq/L) for at least one season in the case of nine water

sources (31% from total number of samples), while samples from GWR17 and GWR20 exceeded the limit in all four investigated seasons.

It was also observed that six of the samples (21% of the total number of samples) collected from karstic springs used as drinking water by the local communities showed annual radon activity concentrations exceeding the EPA limit (Table 2). Fluctuation of the radon activity concentration in samples collected from the same body of water throughout the year has been reported by other studies [36–38], yet no consensus has so far been reached on the causes of such variations.

The ^{226}Ra concentrations from the 29 springs, collected in all seasons, ranged between the lowest limit of detection for the used method and 3.0 Bq/L (Table 3), with an average of 1.1 Bq/L. In this study, we determined a radium concentration above the recommended World Health Organization guideline level for drinking water (1.0 Bq/L) in nine sources (30% of the total number of samples). The radium concentration in twenty spring water samples (70% of the total number of samples) were below this value [28].

Table 3. Temporal variability of the radium activity concentration in the studied karstic springs.

Sample	Radium Concentration (Bq/L)				
	Autumn	Winter	Spring	Summer	Annual Average
GWR1	bdl	0.6	0.6	bdl *	0.6
GWR2	bdl	0.8	0.6	bdl	0.7
GWR3	0.6	1.0	0.8	0.7	0.8
GWR5	0.8	1.1	1.0	0.8	1.0
GWR6	0.6	0.7	0.6	0.8	0.7
GWR7	1.0	1.0	1.1	0.9	1.0
GWR8	1.2	0.8	1.0	1.0	0.9
GWR9	dried	0.7	0.6	0.8	0.7
GWR10	1.5	1.2	1.2	1.1	1.2
GWR11	1.1	1.3	1.1	1.2	1.2
GWR12	0.6	0.7	0.7	0.9	0.8
GWR13	dried	0.6	0.6	bdl	0.6
GWR14	1.4	1.5	1.3	1.2	1.3
GWR15	0.9	1.0	0.9	1.1	1.0
GWR16	1.1	0.9	0.9	0.7	0.8
GWR17	2.3	2	1.8	1.6	1.8
GWR18	1.8	1.5	1.4	1.3	1.4
GWR19	0.7	bdl	0.6	0.8	0.7
GWR20	2.9	3	2.5	2.4	2.6
GWR21	2.4	2.4	2.4	2.6	2.4
GWR22	0.7	0.9	0.8	0.6	0.8
GWR23	1.8	1.5	1.5	1.4	1.5
GWR24	0.6	bdl	0.7	bdl	0.7
GWR25	dry	bdl	0.6	0.6	0.6
GWR26	0.8	1.0	0.7	0.8	0.8
GWR27	0.8	0.8	0.8	0.7	0.8
GWR28	0.7	0.8	0.7	0.6	0.7
GWR29	1.3	1.2	0.8	1.1	1.0
GWR30	0.8	0.6	bdl	0.6	0.6

* Abbreviations used: bdl—below the detection limit (0.05 Bq/L).

No correlation between the radon or radium activity concentrations and the measured physicochemical parameters was found in the PCA (Figure 2). Radon and radium are separated in a different quadrant from the flow rate, while temperature and electrical conductivity are grouped together. Very few springs are defined by temperature and conductivity (GWR23), flow rate (GWR5), or radon and radium (GWR20). Most of the springs are negatively correlated with radon and radium, especially along the F1 axis.

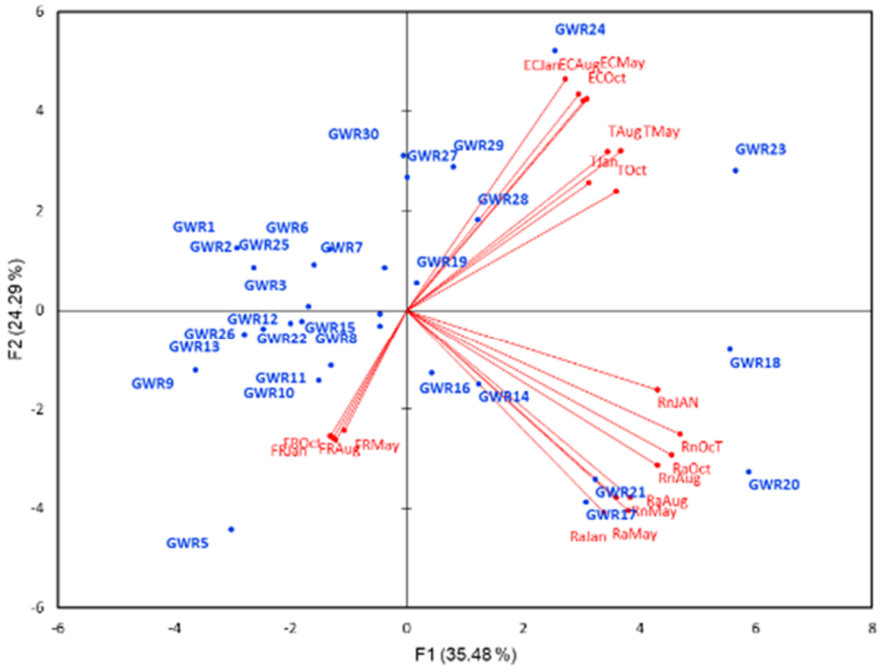

Figure 2. The PCA of temperature (T), electrical conductivity (EC), flow rate (FR), radon (Rn), and radium (Ra) in the analyzed karstic springs (GWR—GWR30, except GWR4) for four months (Oct = October 2019; Jan = January 2020; May = May 2020; Aug = August 2020).

3.2. Radon and Radium Temporal Variability

The seasonal variability of radon and radium concentrations in karstic springs, assessed based on spot samples collected during the four seasons, is presented in the box plots in Figures 3 and 4.

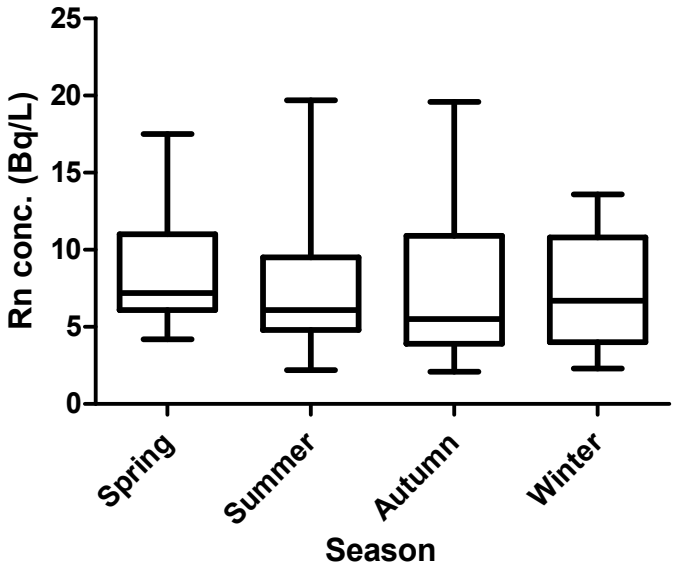

Figure 3. Seasonal (season) distribution of radon activity concentration (Bq/L) in 29 Romanian karstic springs surveyed during 2019–2020.

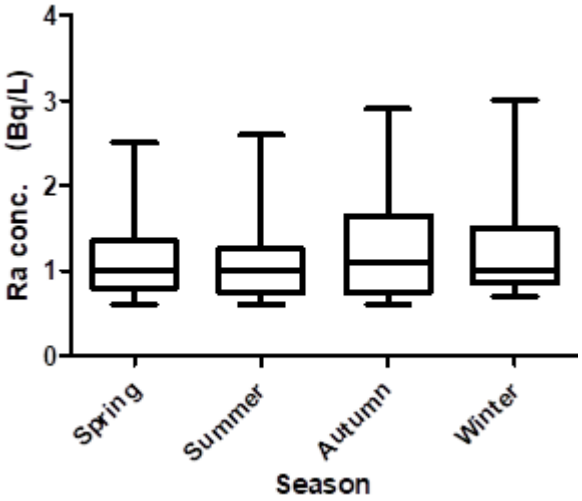

Figure 4. Seasonal (season) distribution of radium concentration (Bq/L) in karstic springs.

The annual values and seasonal measurements of the radon activity concentrations show a log-normal distribution, as confirmed by the Shapiro-Wilk test when applied to the log-transformed data. The geometric means of the radon concentrations are between 6.05 Bq/L (Autumn–October) and 7.85 Bq/L (Spring–May), while the annual mean is 7.0 Bq/L. By applying the ANOVA test for repeated measures to the log-transformed data on the radon concentrations, a significant difference was obtained ($F = 8.12$, $p < 0.001$). The Tukey post hoc test indicated a significant difference between spring and autumn ($p < 0.001$) and between spring and winter ($p < 0.01$).

A similar statistical analysis was applied to radium measurements. Thus, a log-normal distribution was obtained for the four seasons but not for the annual computed means. The seasonal geometric means range from 1.03 Bq/L (Summer–August) to 1.15 Bq/L (Winter–January). A statistically significant difference was obtained by applying the ANOVA test for repeated measures and Tukey's post hoc analysis to the radium concentrations during the hot (summer) and cold (winter) seasons ($p = 0.02$).

As shown, the highest radium activity occurred in autumn while the highest radon activity occurred in spring. The concentration of radon is higher in spring because the precipitation during this period combined with the melting of the snow increases the flow of the springs and decreases the flow time; consequently, the radon does not have time to be released from the water. On the other hand, the concentration of radium is higher in autumn because the flow of the springs is lower and the dissolution of rocks is more efficient. In order to show a correlation between the concentrations of radon and radium, the flow of the springs should be constant over a long period of time, but this was not achieved due to meteorological conditions.

Figure 5 shows the correlations between the seasonal and annual radon and radium activity concentrations by calculating the Pearson correlation coefficient. A moderate correlation (0.65–0.75) was found between radon and radium measurements, regardless of the season in which the samples were taken.

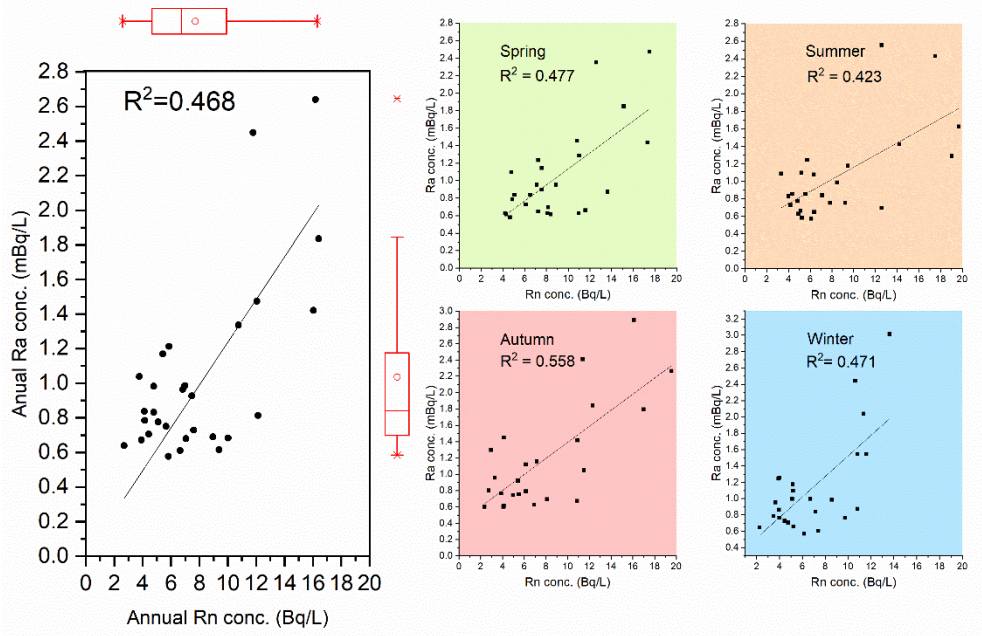

Figure 5. The correlations between the seasonal (**right**) and annual (**left**) radon and radium concentrations.

3.3. Risk Assessment

The radiological health risks of ^{222}Rn and ^{226}Ra are mainly internal, occurring through inhalation and ingestion, and lead to dangerous health issues linked with the respiratory and digestive systems. Most of the cases of cancer attributed to radon in the household water supply arise from the inhalation of radioactive by-products produced by radon that has been released from the water into the air rather than from drinking the water. Furthermore, the increased level of indoor radon that is caused by using water in the home is generally small when compared with the level of indoor radon that originated in the soil beneath the home. Inhalation occurs during the degasification of radon when water is collected and used indoors, while ingestion occurs through the consumption of water containing radon and radium [38].

The consumption of the water investigated in the present study would lead to an annual effective dose of radon [31] between 9.8×10^{-6} and 6.0×10^{-5} mSv/y (Table 4). The highest risk was measured at the spring GWR17, though the radiological risk from the GWR4 spring could not be assessed during the period studied. The associated radiological risk, reported in terms of annual effective dose calculated for exposure to radium [34] dissolved in the investigated waters, was between 5.9×10^{-5} and 2.7×10^{-4} mSv/y. In this study, the average annual effective dose due to radon and radium dissolved in water samples was less than the WHO (World Health Organization's) guideline value of 0.1 mSy/y [28].

Table 4. Radon and radium annual activity concentrations and their associated dose calculated for the investigated karstic springs.

Sample	Radon		Radium	
	Annual Conc. (Bq/L)	Dose (mSv/y) UNSCEAR	Annual Conc. (Bq/L)	Dose (mSv/y) IAEA
GWR1	2.7	9.80×10^{-6}	0.6	6.50×10^{-5}
GWR2	3.9	1.40×10^{-5}	0.7	6.90×10^{-5}
GWR3	4.8	1.70×10^{-5}	0.8	8.50×10^{-5}
GWR5	6.8	2.50×10^{-5}	1.0	9.80×10^{-5}
GWR6	4.4	1.60×10^{-5}	0.7	7.20×10^{-5}
GWR7	4.8	1.70×10^{-5}	1.0	1.00×10^{-4}
GWR8	7.5	2.70×10^{-5}	0.9	9.50×10^{-5}
GWR9	7.1	2.60×10^{-5}	0.7	6.80×10^{-5}
GWR10	5.4	2.00×10^{-5}	1.2	1.20×10^{-4}
GWR11	5.9	2.10×10^{-5}	1.2	1.20×10^{-4}
GWR12	5.6	2.10×10^{-5}	0.8	7.70×10^{-5}
GWR13	9.4	3.40×10^{-5}	0.6	6.30×10^{-5}
GWR14	10.8	3.90×10^{-5}	1.3	1.40×10^{-4}
GWR15	7.0	2.50×10^{-5}	1.0	1.00×10^{-4}
GWR16	12.1	4.40×10^{-5}	0.8	8.30×10^{-5}
GWR17	16.4	6.00×10^{-5}	1.8	1.90×10^{-4}
GWR18	16.0	5.90×10^{-5}	1.4	1.50×10^{-4}
GWR19	10.0	3.70×10^{-5}	0.7	7.00×10^{-5}
GWR20	16.2	5.90×10^{-5}	2.6	2.70×10^{-4}
GWR21	11.8	4.30×10^{-5}	2.4	2.50×10^{-4}
GWR22	5.1	1.90×10^{-5}	0.8	7.90×10^{-5}
GWR23	12.0	4.40×10^{-5}	1.5	1.50×10^{-4}
GWR24	7.6	2.80×10^{-5}	0.7	7.40×10^{-5}
GWR25	6.7	2.40×10^{-5}	0.6	6.20×10^{-5}
GWR26	4.1	1.50×10^{-5}	0.8	8.50×10^{-5}
GWR27	4.2	1.50×10^{-5}	0.8	8.00×10^{-5}
GWR28	9.0	3.30×10^{-5}	0.7	7.10×10^{-5}
GWR29	3.8	1.40×10^{-5}	1.0	1.10×10^{-4}
GWR30	5.8	2.10×10^{-5}	0.6	5.90×10^{-5}

4. Conclusions

This study presents the first attempt at investigating the activity concentrations of radon and radium in Romanian karstic springs and at assessing their potential health hazards for users. The measured values do not exceed the radioprotection standards recommended by Romanian and European guidelines. The low concentrations clearly indicate that radon concentration in spring waters primarily depends on the lithology/geology, the tectonic structures and the presence of uranium minerals in the rock.

This article aims to undertake a risk assessment for exposure to radon in drinking water and thus to establish a link between radon in the air and water in terms of health impact. The radiological health risk for population exposure to radon and radium could be considered mainly internal, occurring through inhalation and ingestion, which leads to dangerous health issues linked with the respiratory and digestive systems. Inhalation occurs through the degasification of radon when water is collected and used indoors, while ingestion occurs through the consumption of water containing radon and radium.

Overall, in the investigated regions, there are no radiological protection requirements due to exposure to radon and radium from drinking water. The estimated doses from the consumption of water based on current preliminary results are insignificant when compared with other natural sources of radiation, such as indoor radon [39].

The correlation between radon and radium levels was moderate; ^{222}Rn may have originated from differences in the geological characteristics of the underground pathways of each of the water sources. Locations where the radon concentration values exceeded

the EPA-recommended limits should be monitored for a period of time to confirm these values. Monitoring these sources, especially those showing high fluctuations in radon concentration, would be an important way to prevent the possible radiologic risk caused by water ingestion and would contribute to a better understanding of the distribution of radioactive gases in Romanian groundwater.

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