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The FIRST large-scale mapping of radon concentration in soil gas and water in Romania

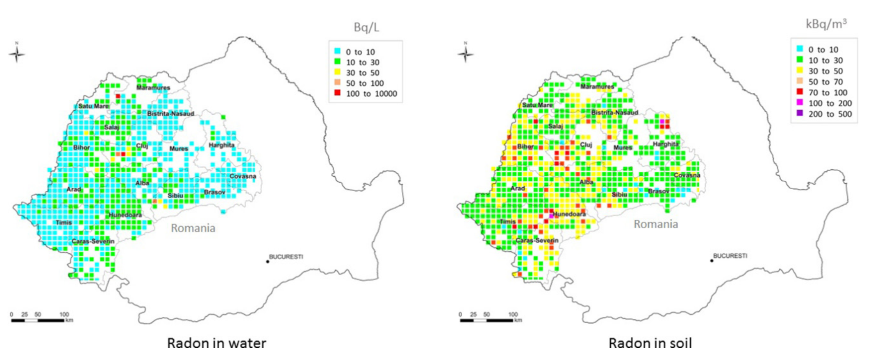
B. Burghele [a], A. Ţenter [a,*], A. Cucoş [a], T. Dicu [a], M. Moldovan [a], B. Papp [a], K. Szacsvai [a,b], T. Neda [b], L. Suciu [c], A. Lupulescu [a], C. Maloş [a], Ş. Florică [a,d], C. Baciu [a], C. Sainz [a,e]

[a] "Constantin Cosma" Radon Laboratory (LiRaCC), Faculty of Environmental Science and Engineering, "Babeş-Bolyai" University, s. no. 30, 400294 Cluj-Napoca, Romania
[b] Sapientia University, Faculty of Sciences and Arts, Calea Turzii, Street no. 4, 400193 Cluj-Napoca, Romania
[c] I.C.P.E. BISTRITA SA, Parcului street no. 7C, Bistriţa, Romania
[d] Faculty of Biology and Geology, Department of Geology, "Babeş-Bolyai" University, Mihail Kogalniceanu Street, no. 1, 400084 Cluj-Napoca, Romania
[e] Department of Medical Physics, Faculty of Medicine, University of Cantabria, c/ Herrera Oria s/n, 39011 Santander, Spain

HIGHLIGHTS

- Significant difference between the warm months and the cold months was observed.
- Low values of radon activity concentration for the sedimentary areas.
- A moderate correlation between the radon concentrations in water and soil.
- Map of radon in water indicates no significant radiological risk.

GRAPHICAL ABSTRACT



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ABSTRACT

In the framework of the last Council Directive 2013/59 (Euratom, 2014) laying down basic safety standards for protection against the dangers arising from exposure to ionizing radiation, the problem of radon was assumed in Romania at national level by responsible authorities through the design and development of a National Radon Action Plan and an adequate legislation (HG nr. 526/2018). In order to identify radon risk areas, however, it is necessary to perform systematic radon measurements in different environmental media (soil gas, water, indoor air) and to map the results. This paper presents an atlas of up-to-date radon in soil and water levels for central and western part of Romania. The radon in soil map includes data from 2564 measurements carried out on-site, using Luk3C radon detector. The Luk-VR system was used to measure radon activity concentration from 2452 samples of drinking water. The average radon activity concentration was 29.3 kBq m^{-3} for soil gas, respectively 9.8 Bq l^{-1} for water dissolved air. Mapping of radon can be a useful tool to implement radon policies at both the national and local levels, defining priority areas for further study when land-use decisions must be made.

1. Introduction

On the basis of Articles 35–36 of the Euratom Treaty, the most important objective of the EU Members States is to monitor and report

* Corresponding author.
E-mail address: ancuta.tenter@ubbcluj.ro (A. Ţenter).

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Table 1
Summary statistics of the measured radon activity concentration in soil gas and water.

Measurement type	No.	Min.	Max.	AM	SD	GM	GSD
Rn in soil (kBq m^{-3})	2564	0.2	179.0	29.3	17.4	24.5	1.9
Rn in water (Bq l^{-1})	2452	0.3	352.0	9.8	16.9	6.2	2.7

AM – arithmetic mean; SD – standard deviation; GM – geometric mean; GSD – geometric standard deviation.

environmental radioactivity (Euratom, 2010). It has been acknowledged that natural radioactivity is the main source of human exposure to ionizing radiation (Euroatom, 2014; HG nr. 526/2018). Radon (^{222}Rn), found ubiquitous in soil, rocks and water represents the main source of indoor radon. With a significant contribution to the contamination of indoor air in Romania (Todea et al., 2013) and worldwide, radon is listed by the World Health Organization (WHO, 2009) as the second leading cause of lung cancer after cigarette smoking. Areal variation of radon levels in houses primarily depends on the geological features of the investigated area (Ciotoli et al., 2017), secondly on the environmental parameters and ultimately on the building characteristics and occupational patterns (Matei, 2012). Through the latter can be mentioned the radon from tap water utilized by the household. Since radon is soluble in water, its degassing is added to the indoor exposure (Kendall and Smith, 2002; Todorovic et al., 2012). Special attention is needed when groundwater is used for supplying drinking water, as radon can become a risk factor for users if the radon concentration in the aquifer is high (NRC, 1999). Most of the cancer risks from radon in drinking water arise from the transfer of radon into indoor air, and the exposure through inhalation (WHO, 2009). A significant part of the Romanian population, from both rural and urban areas is still using raw water for drinking and household. In the present study, 73% of the analyzed water samples were collected from wells and springs, as those were pointed out as household water sources. In 2015 the Romanian government has adopted Law no. 301, implementing Directive 2013/51/Euratom (Euroatom, 2013), establishing a radon reference level of 100 Bq l^{-1} for drinking waters.

The measurements of the radon concentration in soil, has often been used as an indicator and a predictive method to evaluate the elevated indoor radon concentrations of on area (Åkerblom, 1987; Demoury et al., 2013; Cosma et al., 2013; Borgoni et al., 2014; Al-Khateeb et al., 2017; Chen and Ford, 2017; Timkova et al., 2017). The distribution of radon gas in soil can be strongly influenced by local parameters such as chemical and mineralogical composition, physical properties, climatic and geological factors. Another approach in identifying radon risk areas is to assess the geogenic radon potential (GRP), a quantity directly connected to the local geology. High GRP indicates a high probability of radon entering indoors due to geogenic reasons, such as radium content of the bedrock and rock permeability (Ciotoli et al., 2017).

The aim of this work is to assess whether risk areas could be identified based on screening measurements of radon concentration in soil gas and drinking water which will ultimately help prioritize indoor radon surveys.

2. Materials and methods

2.1. Design of survey

The investigated territory covers 16 counties, with an area of 99,837 km^2, in central and western Romania, representing ~42% of the Romanian territory. Several measurements campaigns were carried out between the years 2012 and 2018 in inhabited 10 × 10 km grid cells. The sampling points were selected near dwellings and densely populated areas. The median of the number of measurements per cell, for both radon measurements in soil and water samples was 3, with a

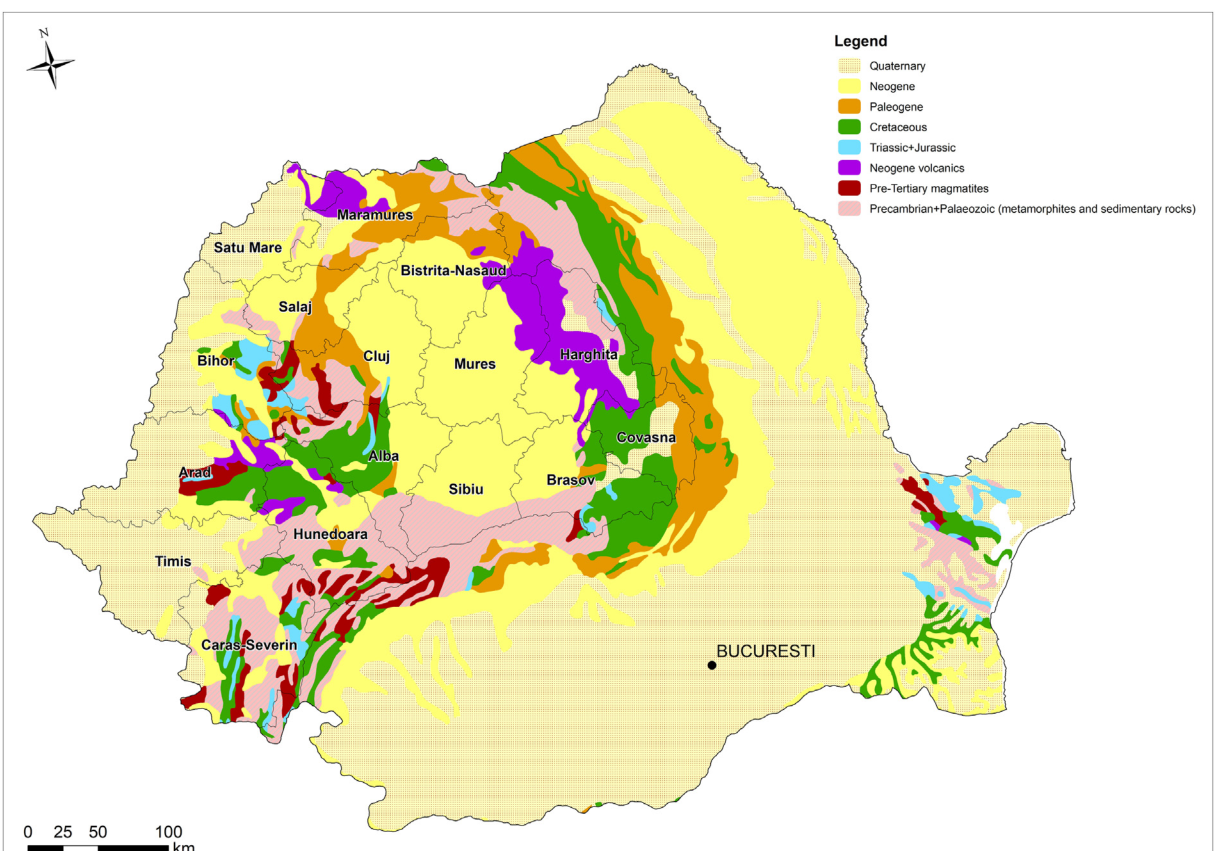

Fig. 1. Simplified geology of the study area (geological background after Institute of Geology and Geophysics, 1964–1968).

minimum/maximum of 1/17. Water samples were collected on availability. The measurements were carried out between 2012 and 2018, in all months, except those with frost. Measurements were performed exclusively during the day.

2.2. Geological features

The geological structure of the mapped area corresponds to a variety of sedimentary, magmatic and metamorphic units. The most important structural feature, the Carpathian Mountains range is represented in the study area by parts of the Eastern and Southern Carpathians and by the whole Apuseni Mountains. The latter separate two large sedimentary basins, the Transylvanian Basin in central Romania, and the Pannonian Basin in the west, both filled with Tertiary, mainly detrital deposits (Mutihac, 1990). The Quaternary alluvial deposits along the rivers partly cover the older formations. The types of soil occurring in the area are also very diverse, according to the substratum and to the specific pedogenetical conditions (Florea et al., 1971).

2.3. Radon in soil measurements

According to the JRC recommendations, a grid with 10 × 10 km cells was superposed on the above-mentioned study area (Tollefsen et al., 2014). Soil gas radon concentration was determined by measuring the radioactivity of soil gas samples extracted from a depth of 80 cm below the ground surface using a metal pipe probe (Neznal et al., 1996). The soil air sample was collected using a Janet syringe (150 ml) connected through a rubber hose to the upper end of the probe. The activity of the sampled air was measured in situ using the Luk3C radon detector (J.Plch, Czech Republic). Background control measurements of the scintillation cells were performed before each sampling. A graphical representation of the radon measurement protocol in soil, implemented according to the Nenzal method in the LiRaCC laboratory has been published by Cosma et al., 2013 (Cosma et al., 2013). The minimum detectable activity was calculated at 0.2 kBq m^{-3} for soil measurements. The quality assurance has been achieved by participating in frequent intercomparison exercises with regard to radon measurements in soil, several reports being publicly available (Papp et al., 2012; Burghele and Moldovan, 2013; Papp et al., 2013). An overview of relative deviation shows values within ±30%, the admissible value for the test criterion.

2.4. Radon in water measurements

From the same grid cells selected for radon in soil measurements, drinking water samples were collected from various sources, such as springs, public network and public/private wells.

Radon measurements in water were carried out using the Luk-VR system, which involves connecting a VR-scrubber to the above-mentioned radon detector (Luk3C). This method requires mixing of the dissolved radon from the water sample with the air above the water in the volume of the glass vessel. Following this procedure, the sample of air was transferred to the Luk3C, and measured by the Lucas cell method. A graphical representation of the radon measurement protocol in water, implemented in the LiRaCC laboratory has been published by Cosma et al. (2008). Drinking water samples (drinking water) originating from different wells, springs and public network were collected between 2012 and 2018 when the air temperature was above 12 °C in spring and above 16 °C in summer. At the time of sampling, the temperature of spring water was between 10 °C and 15 °C, respectively 8 °C and 18 °C for well water. Precautions were taken to avoid sampling following rainfalls. All water samples (tap, spring and well) were carefully collected in polyethylene bottles fully filled (without air bubbles) and tightly sealed in order to prevent loss of radon. The samples were brought to the laboratory and processed within 24 h.

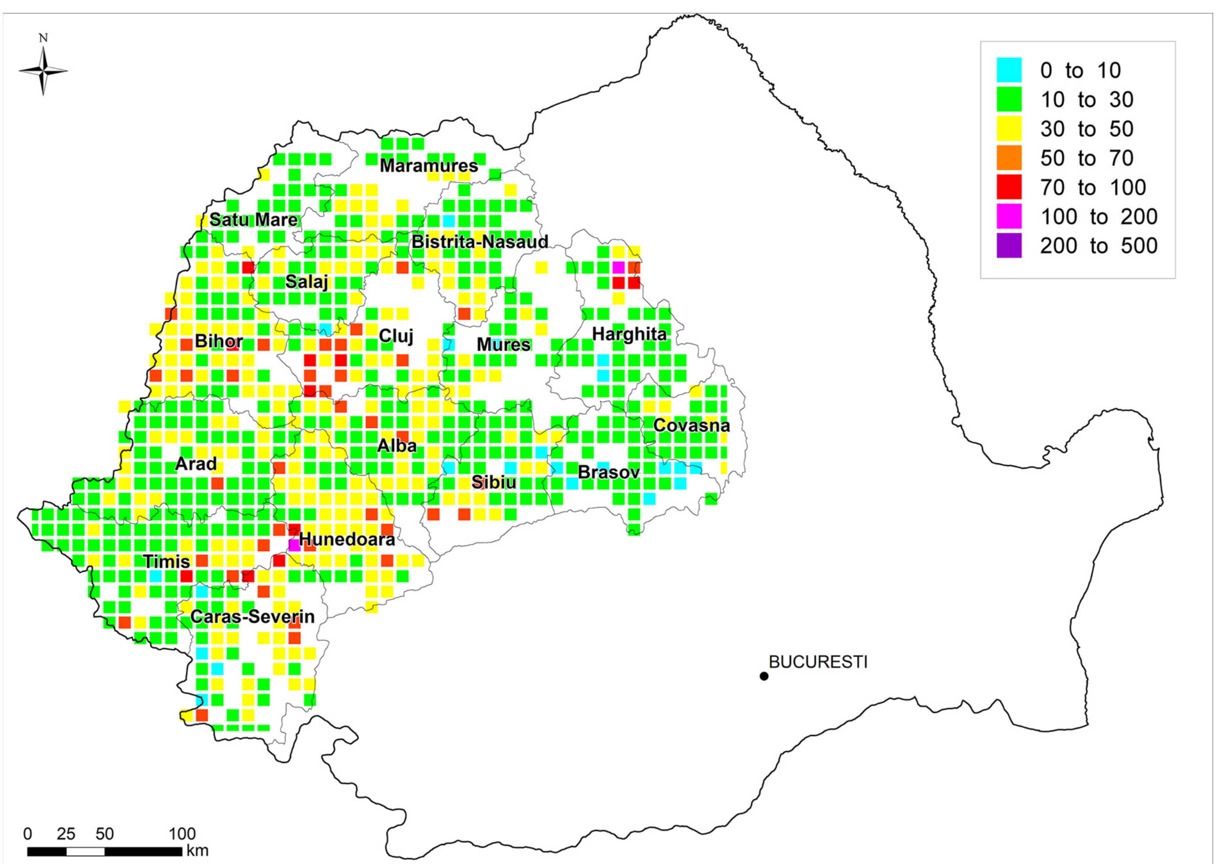

Fig. 2. Arithmetic means (AM) over 10 km × 10 km cells of radon activity concentration (kBq m^{-3}) in soil gas samples.

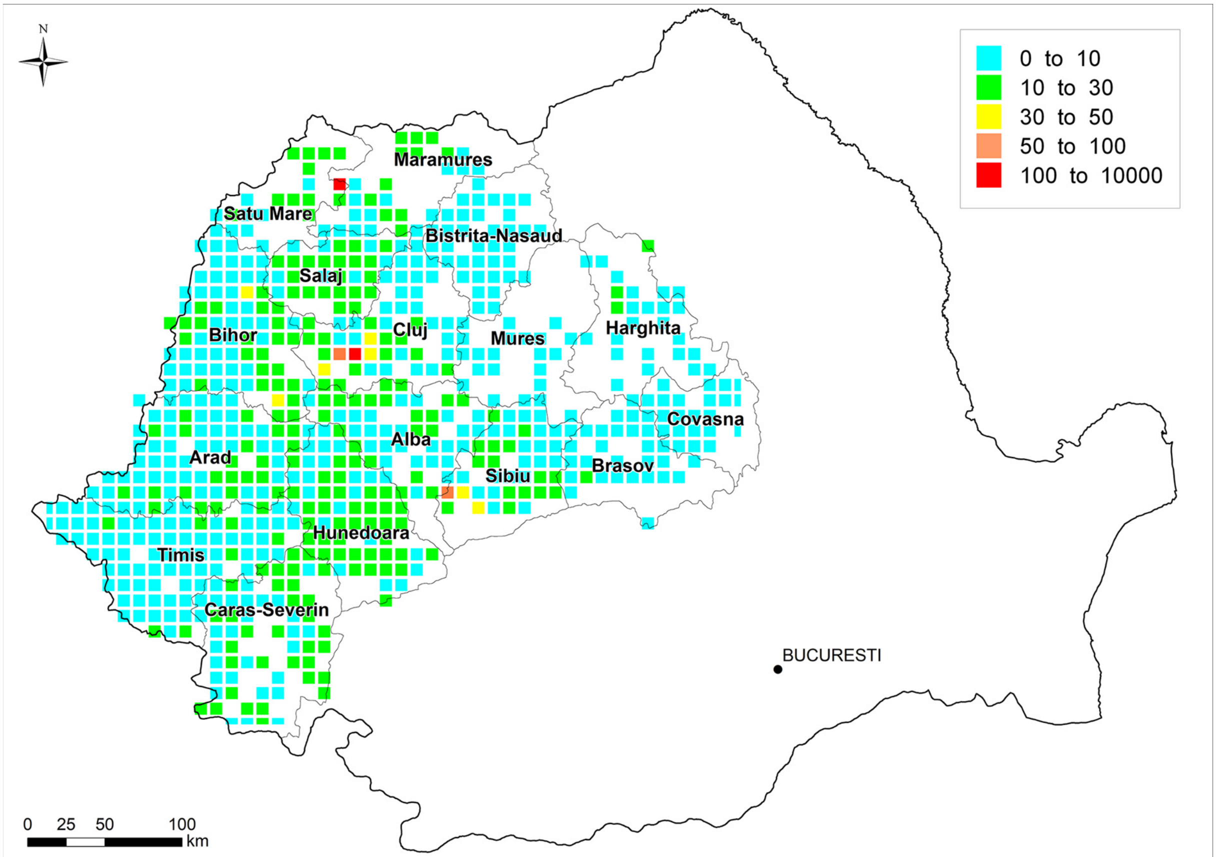

Fig. 3. – Arithmetic means (AM) over 10 km × 10 km cells of radon activity concentration (Bq l^{-1}) in water samples.

The minimum detectable activity was calculated at 0.3 Bq l^{-1} for radon activity in water samples.

Quality assurance has been achieved by participating in intercomparison exercises and a radon-in-water proficiency test (no report is publicly available), the latest test showing a percentage difference of −16 from the reference.

2.5. Data analysis

Statistical analysis of the data was performed using GraphPad Prism 5.0. The statistical distribution of the data was evaluated using the Shapiro-Wilk test. For the comparison of the samples ($n > 2$), the Kruskal-Wallis non-parametric test was performed, with Dunn's post-hoc analysis. In order to evaluate the intensity of the relationship between the examined variables, the Pearson correlation coefficient was calculated on log-transformed data. The significance level α was chosen at 0.05. The maps of the geographic distribution of radon in soil and water were built in accordance with the JRC reference grid using the Classed Post Map function of Surfer 15 and completed in ArcGis 9.

3. Results and Discussions

The study area, spanning over 16 counties, was divided into 1111 polygons according to the internationally suggested 10 × 10 km grid for the European indoor radon map (Tollefsen et al., 2014). Approximately 12% of cells correspond to rocky mountain terrain, significant number of dwellings, which led to 977 cells being susceptible to radon measurements in soil and water, but also indoors (data on indoor radon is under review elsewhere). From a map-level integration perspective, 2564 measurements of radon activity in soil were distributed in 761, while the 2452 radon in water measurements led to the coverage of 694 cells within the studied area. Table 1 shows the descriptive statistics on the concentration of radon activity in soil and water samples.

The arithmetic mean for radon concentration measurements in soil was 29.3 kBq m^{-3}. The recorded values, ranging between 0.2 and 179.0 kBq m^{-3} are comparable to those reported for typical European soil (Beaubien et al., 2003; Al-Khateeb et al., 2017). The geometric mean obtained in the present study was within the range reported at small-scale level in Romania (Cucoş et al., 2017, Papp et al., 2017). The spatial distribution of the integrated arithmetic mean for each cell in terms of radon activity concentration in soil gas samples has been mapped in Fig. 2. By applying the Shapiro-Wilk test, the normal distribution of log-transformed data was not confirmed ($p < 0.01$). One possible explanation is the lack of stratification of results in accordance with environmental parameters that may affect the concentration of radon activity in the soil. It is well known that radon emitted from soil is strongly influenced by weather conditions. Lupulescu et al. (2018)

Table 2
Pearson correlation coefficient analysis for log-transformed data of radon measurements in soil vs. water.

Type of analysis	All raw data	All cells	Cells with ≥2 measurements	Cells with ≥3 measurements	Cells with ≥5 measurements	Cells with ≥6 measurements
$r_{soil - water}$	0.16	0.36	0.33	0.28	0.47	0.47
nr. data	1702	648	496	351	85	45
p	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01

obtained a good negative correlation between the concentration of radon activity in soil and the atmospheric pressure ($r = -0.83$), respectively important seasonal variations in radon activity concentration in soil. In the present study, by applying the Kruskal–Wallis nonparametric test with Dunn's post-hoc analysis, a statistically significant difference was obtained between the median of the soil measurements made in August and the rest of the months, namely June vs. January and June vs. October.

By seasonal grouping, a statistically significant difference between the warm months and the cold (spring and autumn) months was observed, the radon activity levels in the soil during the summer being significantly lower than those recorded in the other seasons. The results are explained by the fact that during the warm season the soil humidity is generally lower, which leads to an increase in the rate of diffusion of radon in the atmosphere and, implicitly, to the reduction of the concentration of radon in soil (Taipale and Winqvist, 1985).

Considering Figs. 1 and 2, low values of radon activity concentration in soil were observed for most of the sedimentary areas of the Transylvanian Depression, and in Banat region (Arad and Timiş counties), while intermediate values have been measured in the middle part of the Western Plane (Bihor County). High values can be correlated with the crystalline area of the Eastern Carpathians. Based on the available data, the occurrence of radon, is satisfactorily explained by the geological structure of the investigated area.

Water samples were mostly collected from wells (69%). Public network water was the second most utilized drinking water source in the investigated sites, consequently 27% of radon in water measurements were performed in tap water. The remaining 4% of the water samples were taken from natural springs. Fig. 3 shows the spatial distribution of the integrated arithmetic mean for each cell in terms of radon activity concentration in water samples. The arithmetic mean of radon activity concentration in all water samples was 9.8 Bq l^{-1}, which is ~36% lower than that reported for Transylvania (Cosma et al., 2008). A possible explanation for this difference is given by the high share of water samples from the local network in the present study as well as the areas involved in the two studies. The present study shows radon activity concentrations in water 10 times lower than 100 Bq l^{-1}, value recommended at national and international leve for drinking water (EU, 2001; WHO, 2011; Law nr. 301/2015), yet similar to those reported for other countries by Duggal et al. (2017). Only 16 out of 2452 (0.7%) radon measurements in water samples recorded values above the value of 100 Bq l^{-1}.

Very low (0–10 Bq l^{-1}) concentrations of radon in water were obtained for most of the investigated sedimentary areas of the Transylvanian Depression (central Romania), and Banat Plain (Timiş and Arad counties). These areas correspond to a predominantly detrital sedimentation of Neogene age (Badenian-Sarmatian-Pannonian) in the Transylvanian Basin and the Neogene bays on the western margin of the Apuseni Mountains. These formations tend to generate low concentrations of Rn in water. Medium concentrations (10–30 Bq l^{-1}) were measured mainly on the western side of the Southern Carpathians. In the western and south-western part of the Transylvanian Depression, the concentrations are somewhat higher than in the eastern, central and northern part. In the Western Plain, the sediments are younger, represented by the deposits of the Pannonian Lake and the Quaternary alluvial deposits of the river network, originating from the western part of the Apuseni Mountains. This situation may explain the slightly higher concentrations in the Western Plain. High (30–100 Bq l^{-1}) and very high (>100 Bq l^{-1}) concentrations of radon in water in mezometamorphic and granitic areas from the Apuseni Mountains, Southern and Eastern Carpathians. Generally, high concentrations of radon, both in soil and in water, occur in areas with crystalline rocks and granite bodies.

The total number of sampling points where the radon concentration in soil was determined in the close vicinity of drinking water sources was 1702. The Pearson correlation coefficient for these sampling points indicates a poor correlation ($r = 0.16$). By centralizing these raw data on a cell level, 648 cells were identified containing at least one common sampling point, in which case the Pearson correlation coefficient suggests a moderate correlation between the two types of measurements ($r = 0.36$). Subsequently, a filtering of the results was attempted in terms of the number of measurements performed in each cell. The correlation coefficients, presented in Table 2, show an increase in the degree of association between the two types of variables measured with the increase in the number of values per cell, but even for $n \geq 6$, the correlation is moderate ($r = 0.47$). By comparing the Pearson correlation coefficients, there is a statistically significant difference between the values obtained for cells with 2 or 3 measurements and those with at least 5 measurements.

A similar analysis was attempted for log-transformed data on residential radon (under review elsewhere) and radon in soil determined in this study. In this situation no statistically significant correlation ($p > 0.05$) was observed, regardless of the level of integration. Even a stratified analysis which took into account the absence of concrete screed and the type of floor did not provide a statistically significant correlation between residential radon concentration and radon in soil. Other studies, reported by Al-Khateeb et al. (2017) fail to reach an agreement whether radon concentration in soil alone would be a sufficient indicator for radon concentration indoor.

4. Conclusions

The present study represents one of the first attempts to identify and map radon priority area according to the radon activity concentration in soil gas and drinking water in Romania. The map of radon in water indicates that there is no significant radiological problem from the perspective of inhaled/ingested radon due to water, except for 2 cells out of 694 cells investigated. Moreover, a radon concentration in water ranging from 0.3 to 352 Bq l^{-1} would insignificantly influence the indoor radon concentration, even if the water source was inside the house. Because the original bedrock of the water sources (especially in case of tap water) is hard to establish, it is difficult to establish a good correlation between radon in water and radon in soil gas from a spot measurement close to the water source. The map of radon in soil suggests that strictly radon measurements in the soil without any additional information (permeability, construction type, etc.) are not a satisfactory predictive indicator of radon indoors. Under these circumstances a similar wider survey would hardly bring different results, especially since the area investigated in the preset study incorporate a much-varied geology than the remaining uncharted territory of Romania. A new study, still in progress, is looking at the use of additional parameters (i.e. permeability, gamma measurements of soil samples, local geology, outdoor radon, indoor radon or architecture) at a much smaller scale in order to establish a more accurate correlation of geogenic radon with indoor radon risk.

Acknowledgements

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