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NATURAL RADIATION DUE TO RADON. CASE STUDY: RADON CONCENTRATION IN HOUSES FROM APUSENI MOUNTAINS

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ABSTRACT. As a radioactive decay product from rocks, in natural conditions, radon is a source of radiation for the population. Moreover, the awareness regarding the fact that this type of natural radiation is of great radiological health significance for the general population has increased since high concentrations of indoor radon were detected. The first step to prevent the risk of exposure to radon is to identify the sources and find the appropriate mitigation method. Once formed by the disintegration of heavy elements in the Earth's crust, radon diffuses into the soil and water, and then it is transported to the atmosphere. Being a noble gas, which doesn't take part in chemical reactions, radon is present in the environmental factors, such as air, water, soil and can accumulates in enclosed spaces with restricted air circulation. The aim of this paper is to determine the total amount of radon to which household members of two houses from Avram Iancu and Câmpeni (Alba County) are being exposed. The radon concentration values were within the national and international proposed limits in the field of radioprotection.

Key words: *radon concentration, soil permeability, dose, radon risk*

INTRODUCTION

An element with great mobility, formed from the decay of ^{226}Ra (a member of the ^{238}U decay chain) radon (^{222}Rn) is the most important radioactive gas because of its longest half-life of 3.82 days. This time is long enough so that a great percent of the radon atoms formed in the ground within approximately few meters from building foundation can reach the indoor environment. In some cases, radon from much larger distances than one meter can be important, if high-permeability transport routes (such as gravely soil or fissures in the ground) are available. In contrast, the thoron (^{220}Rn) formed from decay of ^{224}Ra (a member of the ^{232}Th decay chain), reaches air in substantially lesser amount because its short half-life (only 55.6 s) limits the distance that it can travel before decaying (Nazaroff *et al*., 1988).

Indoor radon exposure of important population groups and associated health risks continues to be a major issue in EU countries and in radon affected areas (Scivyer, 2007).

It is stated that from the total annual radioactive dose received by the population, over 40% is due to inhalation and ingestion of radon and its decay products (ICRP, 2012).

Research on radon concentration in buildings located in uranium mining areas where radon potential is high is of great national and international interest. In many countries authorities target monitoring all houses and implementing effective methods of reducing radon where levels are elevated (Scivyer, 2007). Moreover, several factors affecting the behaviour of radon in indoor air should be carefully studied. Radon concentration in buildings can vary widely strongly depending on the underlying geological formations and characteristics of the house, such as house structure, building materials, insulation or living habits. The main sources of indoor radon gas are represented by the soil under the house and building materials. In addition, water dissolved radon may also contribute to the levels of indoor radon (Gunby et al., 1993; Cosma and Ristoiu, 1996).

Excepting tritium, the European Commission Drinking Water Directive (98/83/EC) does not give maximum activity concentrations for individual nuclides. It sets a maximum effective dose of 0.1 mSv/y from ingestion of water from the public supply. The radon and its daughters are excluded 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 supply with radon concentrations above 100 Bq/l, Member States should set a reference level for 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. On radiological protection grounds for excessive concentrations, exceeding 1000 Bq/l, remedial action is deemed.

Human body is exposed to radon by inhaling (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 from 100 Bq/l to 50000 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).

Study area

In order to identify radon concentration with health risk potential, two areas from Apuseni Mountains were selected as case study: Avram Iancu and Câmpeni (Alba County). Both of them belong to the North Apuseni Mountains major unit and have a different local geological setting. This fact makes possible the estimation of relation between geological setting and indoor radon concentration.

Avram Iancu is situated in the southern part of the Bihor Mountains, between the hydrographic basin of Arieșul Mic and Crișul Negru rivers. This area is characterized by the presence of uranium mineralization. The geological structure belongs to Biharia Nappe System and consists of several metamorphic rock series and un-metamorphosed mollase-type sediments with granite-granodiorite intrusions. Local geology consists of two lithological formations (ophiolitic formation in the lower part of stratigraphic sequence and calcareous-tufaceous series in the upper part of stratigraphic sequence) (Zajzon et al. 2015).

Câmpeni is located at about 25 km est of Avram Iancu to the border of Northern and Southern Apuseni Mountains. In this area surface geological deposits mainly consist of Upper Cretaceous limestones.

MATERIALS AND METHODS

Two houses were diagnosed in terms of radon potential. These were monitored by radon measurements in indoor air, radon measurements of the soil around the house and radon measurements in the water that is used in each household. For that purpose, the houses in the Apuseni Mountains area - Avram Iancu and Câmpeni, were chosen.

Radon in soil and permeability measurements

In order to determine the contribution of the soil gas to indoor radon concentration, radon in soil and the soil permeability were measured. LUK 3C radon detector equipped with Lucas cells of 145 ml was used for measuring radon in soil. This method is based on sampling soil gas and measuring the activity concentration of ^{222}Rn at non-equilibrium state with its daughters (Plch, 2012). Soil gas sampling was performed by a Janet syringe (150 ml volume) connected to a steel probe inserted in soil at 80 cm depth. The detection limit is about 1 $kBq \cdot m^{-3}$ (3σ) and the uncertainties are in the range of tens of $kBq \cdot m^{-3}$ ± 10%. Comparison measurements confirmed the results of the study in this interval (Matolin, 2010). The permeability of soil, was measured with a RADON-JOK device. The principle of determining the soil permeability k [m^2] from the emptying time t [s] is the protocol of Radon-Jok (Radon v.o.s., Czech Republic) which is based on the Darcy law (Barnet et al, 2008).

For further estimation of radon risk in a given location, radon potential of the soil (RP) can be estimated using the following expression:

$$RP = (C_{Rn} - 1) \times (-log k^{-10})$$

where: C_{Rn} [Bq/m^3] is the measured soil radon concentration, and k [m^2] is the permeability of soil.

According to the method of the radon risk assessment of the building site, the values of radon potential can range between RP < 10 (for low risk); 10 < RP < 35 (for medium risk) and RP > 35 (for high risk) (Neznal et al., 2004).

Radon in indoor air

The indoor measurements of the present survey were performed with the help of nuclear track detectors provided by Radosys Ltd. Hungary, which uses CR-39 chips for passive monitoring of radon in indoor air. Each device consists of a cylindrical plastic vial provided with an appropriate lid and a 1 cm^2 CR-39 chip. The radon present in indoor air enters the vial through the space created between the lid and the vial body. Once inside, the alpha particles emitted during the radon decay hit the CR-39 chip leaving tracks. After 3-month exposure, all detectors are returned to the lab and etched in a 6.25 M solution of NaOH and analysed using a Radosys microscope. The track density found is used to calculate the radon exposure and the indoor concentration for each location investigated. The measurements protocol has already been described in previous papers (Cucoș et al., 2012).

Radon in water

The water samples were collected by means of 500 ml plastic bottles. The bottles were filled completely and tightly closed to prevent the entry of air into the bottle and radon gas escaping from the bottle. The samples were transported to the laboratory in order to determine the radon concentrations with the minimum delay and were measured at the earliest possible time. The radon in water was measured using a LUK-3A device specially adapted for radon in water measurement. This equipment called LUK-VR consists of a 500 ml scrubber in which a known quantity of water (300 ml) is introduced. Before measuring, the water was tempered to room temperature.

RESULTS AND DISCUSSIONS

Radon in soil and permeability measurements

The concentration of radon at six points, respectively the permeability of the soil were determined around the house from Avram Iancu (N: 46,22823 and E: 22,47859) (Table 1).

Table 1. *Radon concentration in soil and soil permeability in the house located in Avram Iancu*

Nr.	Radon concentration (kBq/m^3)	Soil permeability k(m^2)
1	11,91±0,63	2,10E-12
2	16,6±0,8	3.8E-11
3	23,06±1,1	4.5E-11
4	22.06±1,08	4,30E-11
5	24,99±1.15	2,90E-11
6	14,06±0.69	4,60E-12

For the Câmpeni house (N: 46,22150 and E: 23,04834) the determinations for radon concentration and soil permeability were performed in five points (**Error! Not a valid bookmark self-reference.**).

Soil permeability was determined using the Darcy equation, where F = 0.149.

The results of applying the above mentioned equation indicated that there is an average risk of radon emission with a value of 34 for the house of Avram Iancu and a value of 27 for the house in Câmpeni (RP <10 low risk, 10 <RP <35 medium risk, RP> 35 high risk) (Neznal et al., 2004).

Table 2. *Radon concentration in soil and soil permeability in the house located in Câmpeni*

Nr.	**Radon concentration (kBq/m^3)**	**Soil permeability k(m^2)**
1	14.02 ± 0.67	4,10E-11
2	2.126 ± 0.09	4.8E-11
3	11,95 ± 0.57	4.5E-11
4	13,71 ± 0.72	4,30E-11
5	1,28 ± 0.06	4,80E-11

Radon in air

Three detectors were placed in each house in the main living quarters: bedroom, kitchen and living room. The results are present in Table 3.

An average radon concentration of 165.6 Bq/m^3 was obtained in the house from Avram Iancu, and an average radon concentration of 154 Bq/m^3 was determined in the house from Câmpeni.

Table 3. *Radon concentration in indoor air in houses located in Avram Iancu and Câmpeni*

Location	**Type of room**	**Radon concentration (Bq/m^3)**
Avram Iancu	living room	200
	bedroom	150
	kitchen	147
Cîmpeni	living room	198
	bedroom	165
	kitchen	101

Radon concentration in water

Five water samples were measured from Avram Iancu to determine the dose on people who use it for different purposes. The concentration values of radon in drinking water varies between 7.8 Bq/l and 13.8 Bq/l with an average value of 9.9 Bq/l. Six water samples from Câmpeni were analysed. The results indicated that radon concentration in drinking water varies between 6.9 Bq/l and 10.5 Bq/l with an average value of 8.6 Bq/l. The results are presented in Table 4. The sources of these waters are captured springs or distribution networks.

Table 4. *Radon concentration in water samples from Avram Iancu and Câmpeni*

Location	Source	Radon concentration (Bq/l)
Avram Iancu	spring	8,2
	spring	7,8
	captured springs	10,2
	captured springs	9,8
	spring	13,8
Câmpeni	spring	7,8
	distribution networks	6,9
	captured spring	7,2
	captured springs	9,8
	captured spring	9,4
	captured spring	10,5

Effective doses

The annual effective doses for ingestion and inhalation were estimated according to the parameters introduced by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2000). The doses obtained due to the indoor radon concentration in both study areas, Câmpeni and Avram Iancu, are the same: 0.95 mSv/year. These values are below the annual limit of the natural dose, which is 2.4 mSv/year but close to 1 mSv/year, which corresponds to the dose that the population receives for one year from natural radon.

The dose due to the intake of radon obtained in Avram Iancu is 0.035 mSv/year and 0.029 mSv/year respectively in Câmpeni. Contribution of the radon to the indoor air in Avram Iancu is 1 Bq/m^3 and 0.82 Bq/m^3 in Câmpeni. These values are insignificant to the contribution of the natural dose of radon: 0.0023 mSv/year for Avram Iancu, 0.0019 mSv/year for Câmpeni.

CONCLUSION

The results of this study clearly indicate that radon concentration in different environmental samples (air, soil, water) from Apuseni Mountain are mostly low and below the proposed reference level of the EU Commission Recommendation (EC, 2001).

Since there are no data on the hydrogeology of the area, the correlation between the geological environment and radon concentrations can only be estimated. Therefore, considering the two types of sedimentary rocks predominant in the geology of the two areas: clastic rocks and carbonate rocks there is the possibility to be a connection between underground waters from clastic and carbonate formations and higher radon concentrations in comparison to underground waters from karstic formations. Moreover, there can be assumed that areas with alternation of clay and sandstone could represent a more important source of radon in soil than limestone of Upper Cretaceous age. This assumption is documented by the lithological heterogeneity of clastic material with different radon potential versus limestone with a lower potential.

Although the highest radon concentrations were expected to be identified in Avram Iancu monitored area, considering its proximity to uranium mineralization, this was not the case, fact suggesting that geology is not the only factor influencing indoor radon concentrations. In Câmpeni, an area with no indication of geology suggesting the presence of radioactive rocks, radon concentrations are most similar to the ones identified in Avram Iancu area.

The risk of radon from the soil calculated from soil radon concentration and soil permeability has a value of 34 for the house in Avram Iancu and 27 for the house in Câmpeni. The results of radon risk indicate that these homes are in areas with medium risk (Neznal et al., 2004). The exhalation of radon from the soil in the studied areas does not add an additional contribution to the natural irradiation of the population.

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