

The Contribution of the Radioactive Gas, Radon, To the Effective Dose Received By the Population of Mioveni City, Arges County, Romania

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ABSTRACT : An essential role in the total effective dose of indoor environments is played by the indoor concentration of radon and thoron . In the present paper, the measurement of indoor radon, soil gas radon concentration, and the drinking water radon concentration was carried out in Mioveni city, Arges county, Romania. Measurements were carried out using RAD-7, a solid-state detector with its special accessory (desiccant, tubes, connective tubes, water kit etc.), an active detector, Airthing Wave plus, which is the first battery-operated smart indoor air quality monitor with Radon detection, including sensors for temperature, air pressure, humidity, TVOCs, and CO₂ and also with a CR-39 nuclear tracks passive detector. The indoor radon concentration varies from almost 36 to 149 Bq/m³ with an average value of 86 Bq/m³. The soil-gas radon concentration varies from 2 to 11,65 kBq/m³ with an average value of almost 6,5 kBq/m³. Radon concentration in water samples varies from 1,7 to 57,12 Bq/liter with an average value of 19,69 Bq/liter. The results were involved in the calculation/estimation of the annual effective dose, ingestion dose, and inhalation doses. The annual effective dose varies from 2,3537 to 7,8162 mSv⁻¹ with an average value of 4,7144 mSv⁻¹. The annual ingestion dose due to drinking water was found to vary from 0,043 to 0,144 mSv⁻¹ with an average value of 0,05 mSv⁻¹. The annual inhalation dose was found to vary from 1,6 to 6,62 mSv⁻¹ with an average of 3,80 mSv⁻¹.

KEYWORDS: Active detector, Annual effective dose, Radon, Radon concentration, Solid-state radon detector,

I. INTRODUCTION

Radon gas is produced by the decay series of radium (²²⁶Ra) which is also a decay product of uranium (²³⁸U) decay series. Radon is an inert, water-soluble gas, having no color or smell. Radon further decays into Polonium (²¹⁸Po) which emits an alpha particle of 5,5 MeV energy. Radon is present in small trace amount almost everywhere on earth, being distributed in soil, groundwater and in the lower levels of the atmosphere. Radon reaches by different processes and accumulates in houses, underground mines working places and so on. Most of the time the most affected are the rooms/dwellings/work places that have direct contact with the ground. Rooms/dwellings on the upper floors should not be neglected either, the radon being able to reach inside them through various access ways, among which we list: pipes, cracks in the walls, leaks in the vents etc.

The contribution of radon is approximately 55% of the total internal radiation exposure to human beings [2]. Radon in the outdoor environment air is very quickly diluted. But when it enters into the indoor air, it accumulates in houses/dwellings and causes serious damage to the people living there. The major source of radon in the indoor air is the uranium prone area present in the soil and rocks beneath the house as well as the drinking water, building material and the cooking gas we use [3]. Environmental Protection Agency mentioned radon as the second largest cause of lung cancer, after smoking [4]. Many research studies were made discovering a huge number of lung cancer induced by the inhalation/ingestion of radon and its progenies [5]. The average annual effective dose was estimated to be 2,4 mSv from natural sources, out of which, approximately 1,0 mSv is because of the exposure of radon [6]. The International Agency for Research on Cancer has categorized accumulated radon in dwellings as a the first carcinogen group. The presence of radon in water is due to its radium content, bedrock and the soil surrounding it. The concentration of radon is

generally low in the surface water as compared to the underground water because of the presence of granite, sand and gravels [7]. Soluble radon found in water is important as secondary source for the indoor radon exposure. This soluble radon, degas from the usable water inside the dwellings and becomes airborne[8] It is easy to understand that the indoor radon concentration increases simultaneously with the high radon content in water. Through the presence of radon in water, inhalation and ingestion cause exposure principally to lungs and stomach. Inhalation is expected to cause much health risk. The chances of inhaling are much higher than ingesting [9]. This paper aims to investigate the indoor radon concentration, the soil gas concentration and the concentration in drinking water in Mioveni city, Arges county in Romania. The zone mentioned is homebirth place and searching for radon past information, there are no clues that the zone would give surprising high values of the radon concentration. The measurements aim to estimate the annual effective dose as well as inhalation and ingestion doses to the people in dwellings (whether we are talking about blocks of flats or houses).

II. EXPERIMENTAL MEASUREMENTS AND THEORETICAL DATA/APPROACH

Study area- geology data : Mioveni city is located in the central area of Argeş County, on the coordinates 45°12' north latitude and 25°03' east longitude. Mioveni is located in the great Subcarpathian unit of the Getic Plateau or the Getic Piedmont (which continues the Muscelele Argeş), more precisely in a subunit of it, the Cânduşti Piedmont. The geomorphology of Cânduşti Piedmont is complex, being composed of terraces, long and smooth peaks, fringed by wide and steep valleys of rivers: Doamnei (at the administrative limit with Mărăcişeni commune), Târgului (at the administrative limit with Dărmăneşti commune) and the river Arânăt of the NW of the Racoviţa neighborhood and of the city of Mioveni). The soil cover is the result of the interaction of physical and geographical factors, is quite diverse, but in general the special influence of the relief rock and groundwater is noticeable. The soils are divided into several zonal and intrazonal units which constitute the pedological potential exploited as a basis for the development of biocenoses and various cultures in relation to the environmental conditions. We distinguish the following types of soils: brown clay-alluvial soils, luvic brown soils, regosols, erodisols. National Institute of Statistics [10] "Last but not least, the uranium deposits in the county - the energy source of the future" should not be neglected." Taking these into account, measurements can indicate surprising values of radon concentration.

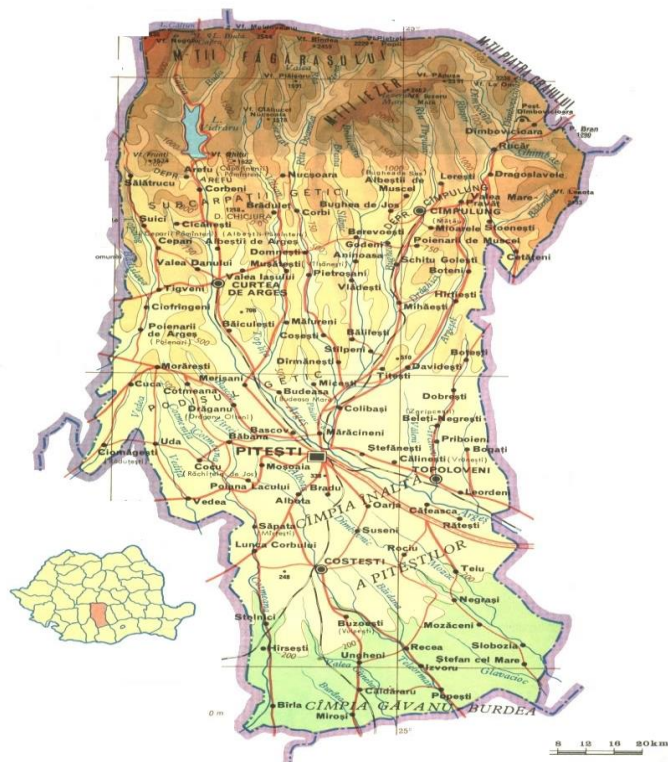


Figure 1-Mioveni city, Arges county

Experimental method : For the experimental data (for radon concentration in air) measurements were carried out with 3 (three) different types of detector, as follows:

- RAD-7 detector was used for the measurement of radon concentration in indoor, soil and water. Silicon, as a semiconductor material it is used by RAD-7 to convert the alpha radiation into electrical energy. The detector inside the RAD-7 distinguishes the alpha particles from ^{218}Po and ^{214}Po with an energy range of 6,0 MeV and 7,9 MeV, respectively, into their respective windows. [12]

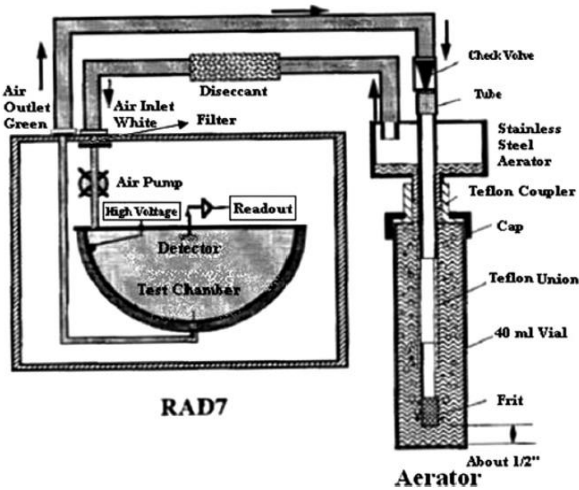


Figure 2- RAD-7 scheme [13]



Figure 3 – RAD-7 soil kit

- Airthing Wave plus is the first battery-operated smart indoor air quality monitor with radon detection, including sensors for temperature, air pressure, humidity, TVOCs, and CO₂. [1]

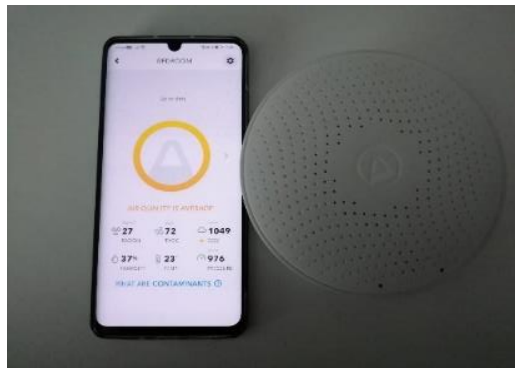


Figure 4- Airthing Wave Plus radon detector

- CR-39 Radon detector: The method involves the use of a plastic container that plays the role of a storage chamber and contains a CR-39 detector inside. Figure 5 presents those aspects.

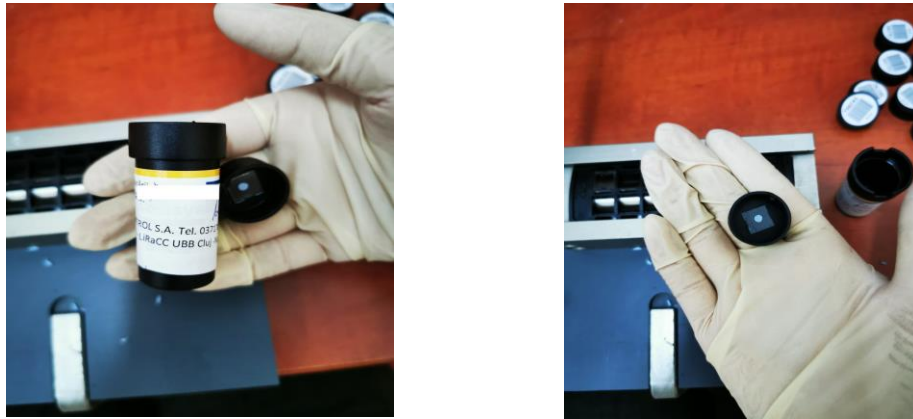


Figure 5 – CR-39 Radon detector

The detector has an area of 1 cm² and the storage chamber has a height of 55 mm and a diameter of 26 mm. CR-39 trace detectors are made of a polymer (alidiglycol) and are sensitive to radiation with energies in the range of 0,2-20 MeV. As alpha particles pass through the polymer, energy transfer leads to the breaking of molecular chains along the particle's path. In order to visualize the traces resulting from the interaction of the incident particle with the detector material, a chemical development of it is required. CR-39 detectors have an antistatic treatment applied in the manufacturing process. The soil gas radon concentration was measured at a depth of roughly 80 cm (respecting the norms/legislation in force [14]) using a 10 mm diameter probe. The soil probe is immersed in the soil with gentle strokes of a hammer. The soil probe is connected to drierite which is connected to the RAD-7 device. Drierite is an important accessory that absorbs the moisture from the soil gas. Hence the soil gas passing through the drierite is moisture free. The gas from the soil probe is sucked for the 5 min pumping phase and then the data is received in the form of bar charts and cumulative spectra of the sample which can be printed with the infrared printer. During the soil gas sampling, was used sniff mode with grab sampling. For the determination of radon concentration in water, RAD H₂O was used. A watt-250 protocol along with Grab mode was chosen on the RAD-7 for the 250 ml samples. Airthing Wave plus radon detector was left in each dwelling for 1 week. CR-39 Radon detectors were placed for 3 months (from June to September).

III. DOSE ESTIMATION

Mean effective dose : The annual effective dose (H) for the inhabitants was calculated from the experimentally determined value of radon concentration (CR_n) using expression:

$$H(mSv/y) = E_{\text{internal}}(mSv/y) + E_{\text{external}}(mSv/y) \quad (1)$$

Mean internal effective dose for ingestion and inhalation

For the estimation of ingestion and inhalation doses, the indoor waterborne radon in the dwellings is a serious concern rather than other contaminated materials in it.

The annual mean effective dose for ingestion (E_{igestion}) and inhalation (E_{ihalation}) were calculated from experimentally determined value of radon concentration in air (CR_n) water (CR_nW) by using expression (2),(3) and (4) respectively given by reference [6]:

$$E_{\text{igestion}}(mSv/y) = CR_{nW} \times CW \times EDC \quad (2)$$

$$E_{\text{ihalation}}(mSv/y) = CR_n \times F \times O \times DCF \quad (3)$$

$$E_{\text{internal}} = E_{\text{igestion}} + E_{\text{ihalation}} \quad (4)$$

Where, CW is the weighted estimate of water consumption (720 l/y) [16], EDC is the effective dose coefficient for ingestion (3,5 nSv/Bq), F is the global average (0,4) of equilibrium factor for radon and its progeny [6], O is the global average indoor occupancy factor (7000 h/y) [6] and DCF is the dose conversion

factor ($6,3 \times 10^{-6}$ mSv/h/Bq/m³) for radon and its progeny [6], $D\gamma$ is the gamma dose for indoor measurements.

Mean external dose

$$E_{\text{external}} = O \times D\gamma \times 10^{-3} [\text{mSv/y}]$$

(5)

IV. RESULTS AND DISCUSSION

Table 1 shows the indoor radon concentration, soil gas concentration and drinking water radon concentration founded in Mioveni city. Were carried out 15 measurements of each parameter for radon concentration: air, soil and water. Various areas of the city were submitted to measurements, both houses and apartments. Attempts were made to perform measurements in apartments located on the ground floor of the building, but also on the upper floors to highlight the differences in values found, as well as the various factors that could have an impact on them. The measured indoor radon concentration varies from 36 Bq/m³ to 149 Bq/m³ with an average of almost 86 Bq/m³ and a geometric mean of 77,86 Bq/m³. Higher radon concentration in dwellings is due to the construction material as well as their ventilation condition.. The measured radon concentration is major than the world's average of 40 Bq/m³ [6], but the average value is also well below to the recommended level of 300 Bq/m³ [15]. The measured soil gas radon concentration varies from almost 2 kBq/m³ to 11,65 kBq/m³ with a mean value of 6,3 kBq/m³ and a geometric mean of almost 5,5 kBq/m³. The soil gas concentration was found to be high enough supposing to be a prone area. The radon concentration in drinking water used by the members in respective dwellings was also measured for calculating the ingestion doses received. The drinking water radon concentration varies from 1,7 to 57,12 Bq/liter with a mean value of almost 20 Bq/l and a geometric mean of 14 Bq/l. The radon concentration in drinking water was above the EPA recommended limit of 11 Bq/l.

The gamma radiation level was measured for indoor as well as outdoor environments using pocket gamma survey meter for each dwelling. Gamma radiation for indoor varies from 0,09 to 0,17 μSv/h with the mean of 0,12 μSv/h, while for outdoor, it varies from 0,09 to 0,11 μSv/h with the mean of 0,10 μ Sv/h.

Table 1- Dwellings and measurements results

Dwelling code	Gamma dose [μSv/h]		Average radon concentration				
	Indoor	Outdoor	Indoor conc. [Bq/m ³] RAD-7	Soil gas conc. [kBq/m ³] RAD-7	Drinking water [Bq/l] RAD-7	Airthings Wave Plus [Bq/m ³]	CR-39 passive Radon detector [Bq/m ³]
1	0,11	0,10	37	2,300	2.35	41	29/48
2	0,12	0,11	39	2,000	1.7	58	37/61
3	0,15	0,11	149	8,550	57.12	114	74/122
4	0,09	0,11	89	3,100	28.5	97	51/84
5	0,10	0,10	36	1,980	21.3	50	30/50
6	0,11	0,09	112	6,120	17.45	79	53/87
7	0,12	0,09	138	11,650	20.63	96	57/94
8	0,17	0,10	107	9,400	31.85	70	44/73
9	0,16	0,10	66	6,300	22.3	53	36/59
10	0,13	0,10	121	8,750	30.58	110	67/111
11	0,12	0,10	74	5,400	8.15	78	47/78
12	0,12	0,10	88	10,000	16.44	94	55/91
13	0,12	0,10	100	8,110	17.535	99	61/101
14	0,14	0,10	55	6,230	7.15	117	72/119
15	0,09	0,10	72	5,050	12.30	77	48/79*
Weighted mean ± SD	-	-	85,53	6,329	19,690	82	-
Geometric mean	-	-	77,87	5,487	14,344	78,52	-

Standard Deviation	-	-	35,90	3,071	14,002	24,21	-
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* Example of calculation: The value measured in dwelling no.15 was 48 Bq/m³, but using the seasonal correction factors mentioned in [14], appendix 2, there will result out a value of almost 79 Bq/m³ for indoor radon concentration in dwelling no. 15.

Figure 6 shows the variation of indoor gamma radiation to the outdoor gamma radiation level for each dwelling site.

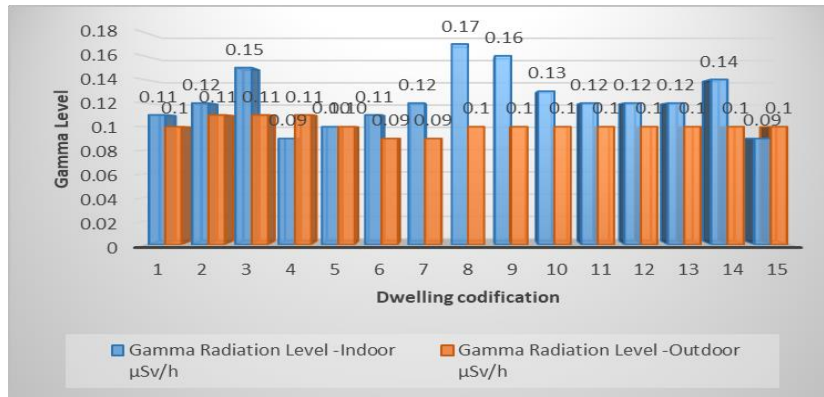


Figure 6- Gamma radiation level [µSv/h]

Table 2 shows the indoor radon Annual Effective dose, the inhalation dose and the ingestion doses for the dwellings.

Table 2- Doses

Dwelling code	Annual effective dose [mSv/y]	Internal dose [mSv/y]	External dose [mSv/y]
1	2,4204	1,6504	0,77
2	2,5776	1,7376	0,84
3	7,8162	6,7662	1,05
4	4,6574	4,0274	0,63
5	2,3537	1,6537	0,70
6	5,7918	5,0218	0,77
7	7,0253	6,1853	0,84
8	6,0258	4,8358	1,19
9	4,1095	2,9895	1,12
10	6,3648	5,4548	0,91
11	4,1494	3,3094	0,84
12	4,7925	3,9525	0,84
13	5,3286	4,4886	0,84
14	3,4425	2,4625	0,98
15	3,8610	3,2310	0,63
Overall weighted average	4,7144±1,6773	-	-
Average worldwide [6]	1,15	-	-

Figure 7 shows the variation of the Annual Effective dose for each dwelling site. The annual effective dose varies from 2,3537 to 7,81 mSv/y. The average value is above the worldwide average value of 1.15 mSv/y [6] but it is above the limit of the recommended action level of 6 mSv/y [17].

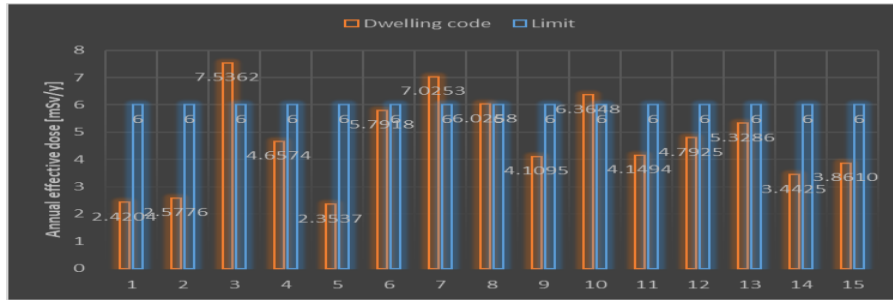


Figure 7- Annual effective dose [mSv/y]

Figure 8 shows the correlation graphic for the measured indoor radon concentrations and soil-gas radon concentration of the study area- Mioveni city. A positive correlation ($R^2=0,7997$) was observed between indoor radon concentration and soil-gas radon concentration, which shows that the soil gas radon concentration also contributes to indoor radon concentration.

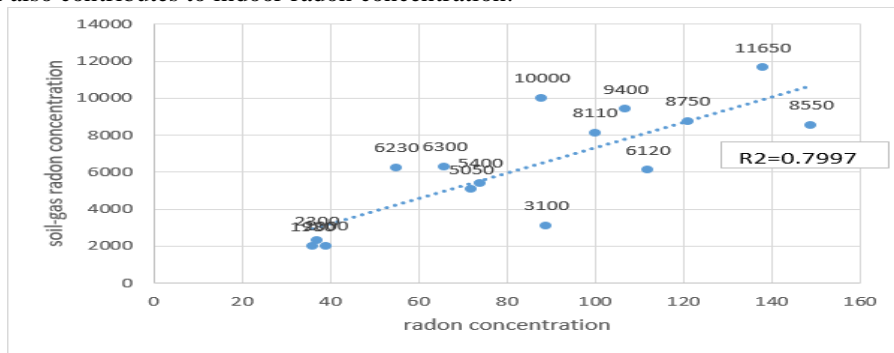


Figure 8- Correlation indoor radon concentration and soil-gas concentration

Figure 9 shows the correlation graphic for the measured indoor radon concentrations and drinking water radon concentration. The indoor radon concentration was also found positively correlated ($R^2=0,7346$) with the drinking water radon concentration. Both of the graphs show that the indoor radon concentration depends upon the radon in the nearby soil and radon in drinking water.

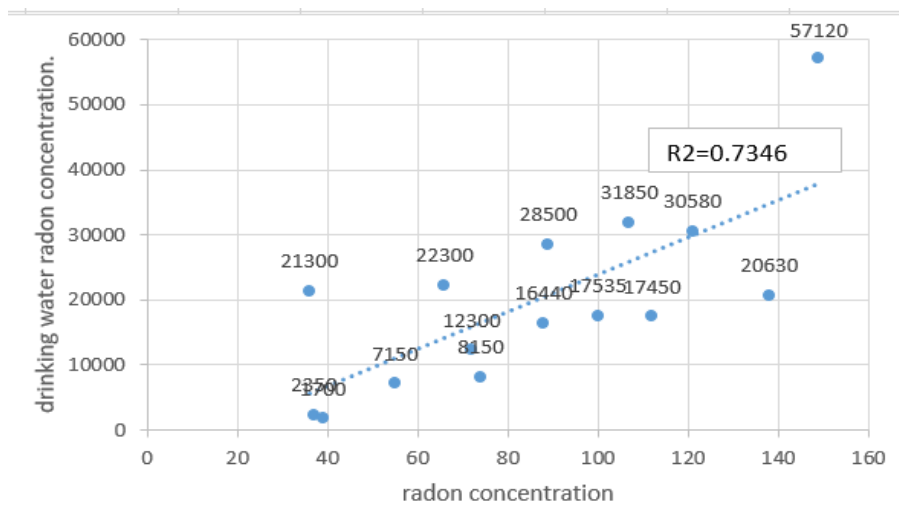


Figure 9- Correlation indoor radon concentration and drinking water concentration

IV. CONCLUSIONS

It is concluded that the radon concentration is relatively high in soil-gas and drinking water of the study area due to possible existing of uranium prone area under territory of Arges conty. Results have shown a positive correlation between soil gas radon concentration and indoor radon concentration. In a similar way, positive correlation has been found between radon concentration in drinking water and indoor radon concentration. This leads to the conclusion that along with the soil underneath the dwellings and building material, soil nearby the houses and drinking water also contributes to the indoor radon concentration in the study area. It is of great interest that all water samples were taken from the city's general water supply system.

Building material : Models have been developed for the dose assessment of gamma radiation emitted from construction materials. These models use a reference room that is assumed to have the dimensions 3m x 4m x 2,5 m and that is made from concrete [18]. The level of radon in the room can be calculated using Eq. (6) (see Ref. [18])

$$C_{bm} = \frac{1}{(\lambda + \alpha) \times V} \times \sum E_i \times A_i \quad (6)$$

where

C_{bm} = Radon contribution to indoor air concentration from construction material

λ = Radon decay constant = 0.00755

n = Air exchange rate = 0,5 volumes/hour

V = Room or buildings inner volume = 30 m³

E_i = Radon exhalation rate = 13 Bq/m²h

A_i = Surface area of walls made using the construction material in the room = 55,4 m²

The contribution of the construction material to the radon concentration in the room can be calculated using Eq. (6) above:

$$C_{bm} = (1/((0.00755 + 0.5) * 30)) * (13 * 55.4) = \mathbf{50 \text{ Bq/m}^3}$$

For workplaces and municipal buildings, the airflow demands are typically higher, up to 1,0 air change per hour. This additional ventilation may reduce the above contribution from the construction material to the total indoor radon concentration to approximately 25 Bq/m³ for the same volume facility.

Soil : It is possible to calculate how much soil air is entering the building from the ground every hour by using Eq. (7) (see Ref. [19]):

$$C_{house} = \frac{1}{(\lambda + \alpha) \times V} \times C_{soil} \times L \quad (7)$$

where

C_{house} Measured radon concentration in the house (Bq/m³)

C_{soil} = Measured radon concentration in the ground under the building (Bq/m³)

λ = Radon decay constant

V = Volume of air in the house (m³)

n = Air exchange rate (air exchange/h)

L = Volume of soil air entering the building per hour (m³/h)

Using for example some numbers for the parameters below described, the calculated volume of soil air entering the building is:

$$L = (500 * (0.00755 + 0.5) * 500) / 2\ 000 = \mathbf{63,4 \text{ m}^3/\text{h}}$$

As a rough indication of the household's water contribution to the radon concentration in a relatively small indoor space, it is generally accepted that 1000 Bq/l of radon in the water contributes to 100–200 Bq/m³ of radon in indoor air, if the water consumption is 1 m³/day [20].

The formula for the calculation of radon exhalation from the water into a building is given by the Eq. (8)

$$C_v = \frac{C_w}{24 \times (\lambda + n) \times V} \times \sum e_i \times W_i \quad (8)$$

C_v = Radon contribution from household water usage to indoor air, Bq/m³

C_w = Radon concentration in the water, Bq/m³

λ = Radon decay constant, 0.00755 h⁻¹

n = Air exchange in the building, h⁻¹

V = Building volume, m³

W_i = Volume of water used daily for purposes, m³/day

e_i = Share of radon that merges into the indoor air (value given by tables in specialty literature)

SSG-32 [21] provides recommendations on the process to determine compliance of building and construction materials containing radionuclides of natural origin with the reference level. The process includes the determination of the activity concentrations of radionuclides of natural origin, followed by the determination of an activity index.

An example of an activity index I that could be considered by the national authority is given by Eq. (9) (based on para. 4.20 of SSG-32 [21]).

$$I = \frac{C_{Ra}}{300 \text{ Bq/kg}} + \frac{C_{Th}}{200 \text{ Bq/kg}} + \frac{C_K}{3000 \text{ Bq/kg}} \quad (9)$$

where

C_{Ra} is the activity concentration of ²²⁶Ra in the construction material in Bq/kg,

C_{Th} is the activity concentration of ²³²Th in the construction material in Bq/kg,

C_K is the activity concentration of ⁴⁰K in the construction material in Bq/kg.

Guidance on the application of the activity index is provided in paras 4.21–4.27 of SSG-32 [21].

As shown in Figure 6, the ambiental gamma dose rate in 12 of the 15 dwellings (tested for radon concentration) has higher values inside the dwellings than outside. Dwelling no. 5 has an equal value for both indoors and outdoors gamma dose rate, and 2 of the dwellings have lower values indoors than outdoors.

Slightly higher values of the ambiental gamma dose rate may be due in particular to existing construction materials. Therefore it is imperative to take very seriously the problematic of it when it comes to dwelling refurbishment or/and dwellings/workplaces constructions.

Making a comparison between dwelling no. 2 and dwelling no. 3, both being on the ground floor of a 4-floor building, water radon concentrations are extremely different: 1,7 Bq/l versus 52 Bq/l. The only plausible answer is that the public water pipes for each section are positioned differently, at different depths, having different radiation sources (materials of nuclear interest) or/and made of different materials, proving once more that not only materials that are used inside the house are important for us and our health.

All the measurements that were carried out proved that all the numbers were beyond the recommended level of radon concentration:

- 300 Bq/m³ for radon concentration in air;
- 100 Bq/l for radon concentration in water;

For dwellings 3 and 7 actions must be taken immediately for reducing radon concentration. The numbers showed in the paper could not be true, because counting (for real) the time that a person stays in the dwelling may show different than 7000 h, and the real dose could be less than 7,5362 or 7,0253 mSv/y.

National Commission for Nuclear Activities Control (CNCAN), the regulatory body in Romania, by the order of the CNCAN president no. 185/2019 for the approval of the Methodology for determining the radon concentration in the air inside buildings and at workplaces specifies that the method of measuring radon concentration (for early screening measurements) are measurements using passive radon detectors. Active radon detectors can be purchased (at the time of speaking) from reputable manufacturers without difficulty. When discussing passive measurements (using passive detectors) things get a little complicated. Considering that the development of detectors after the exposure period (minimum 90 days - in the case of screening measurements) is performed by a CNCAN accredited laboratory. The value of the annual radon concentration is corrected with the correction factors provided in the Methodology [14].

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