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RADON ASSESSMENT WITH SOLID-STATE NUCLEAR TRACK DETECTORS IN BUCHAREST AND ITS SURROUNDING REGION

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Abstract. Radon contributes with more than 50% to the dose equivalent to the public coming from natural sources, and is a major risk factor for lung cancer occurrence (10–14%, in Europe, p.a.) which should be investigated. Insufficient data on radon levels exist for Romania up to now and further extensive surveys are needed. The paper presents a series of in-situ indoor radon data in various locations in the Bucharest region, in long-term exposures, using a relative method with CR-39 nuclear track detectors, developed formerly at NIPNE.

*Key words***:** indoor radon, risk assessment, solid-state nuclear track detectors, CR-39.

1. INTRODUCTION

Of the three isotopes of radon, radon (^{222}Rn) and thoron (^{220}Rn) contribute with more than 50% to the dose equivalent to the public coming from natural sources. Radon is a major risk factor for lung cancer occurrence (10-14% of the cases in Europe, p.a.). Previous measurements in Romania are scarce and show a radon level above the European average, while, according to the WHO, the lack of sustained preoccupation situates us among the "weakly radon developed" countries with respect to the response to this hazard. However, after the completion of this work, new radon data for some regions of Romania were published, which are included in our discussion.

Radon and thoron, and their decay products (its "progeny") are permanently present everywhere in our natural environment. They can be found in higher concentrations in the confined atmospheres of buildings and underground spaces, mines, spas; the main source of radon is the soil, but significant contributions may come from building materials and groundwater.

In temperate and cold regions, heat conservation and reduced ventilation habits increase the radon levels, especially in the cold months (winter), determining, e.g. for Romania, an increase of the measured levels by a factor of about 2 [1].

The present paper deals only with ^{222}Rn (radon) measurements using the SSNTD technique in several locations in Bucharest and the surrounding region.

Radon originates from the uranium-radium natural decay series (^{238}U-^{226}Ra) and is a noble gas, which escapes into air easily from the material in which it is formed. Uranium and radium occur widely in soil, rocks and water – thus the air we inhale outdoors or indoors contains radon. Average radon levels are of 8 Bq/m^3 for continental outdoors, at 1m from the soil, and only 0.04 Bq/m^3 above the sea. Indoor radon concentrations reach orders of magnitude higher values, typically 12–300 Bq/m^3 up to maximum values of several thousands of Bq/m^3 in radon prone areas, e.g. in Spain, UK, Germany, etc. [2]. For Romania, recently published data show values in the limits of 15–1005 Bq/m^3, with an arithmetic mean of (115±48) Bq/m^3 for measurements performed in winter, spring and summer seasons for several regions in Transylvania [3].

Radon decays to a number of short-lived radioactive decay products, which may attach to aerosols or remain unattached; both may deposit in the lung if inhaled, and will irradiate the tissue for long time - a far greater risk than the gas itself. Dosimetric discussions need the knowledge of the **radioactive equilibrium factor** between radon and its progeny in the specific point, expressed as the ratio of the total alpha particle energy emitted by the given radon - progeny mixture to the total energy emitted by the same concentration of radon gas in equilibrium with its progeny. For most indoor environments of interest, the equilibrium factor between radon and its progeny is fairly constant (40-50%) [2], but it can reach extreme values in some locations, down to 20% and up to 80%. Most of the scientific discussion on radon is usually expressed in terms of radon concentrations, rather than in decay product concentrations, for 2 main reasons:

– it is by far easier to measure concentrations of radon gas than the progeny, especially for long term measurements; and

– owing to the higher dose conversion factor of the unattached fraction of radon progeny in lung dosimetry models, the effective dose relates more to radon gas concentration than to the equilibrium equivalent radon concentration.

Unless the progeny is measured by a spectrometric method, an average equilibrium factor for ‘stationary’ conditions of about 0.4-0.45 is to be assumed for the indoors and about 0.7 for the outdoors. Dosimetric considerations are beyond the scope of the present paper.

2. SAMPLES AND TECHNIQUE

Our lab uses a long-time exposure passive method of measurement for indoor radon, based on CR-39 solid-state nuclear track detectors (SSNTD), encapsulated in special monitoring devices/radon probes (MD), developed by A. Danis [4], Fig. 1. The volume of the radon probe (MD) is (15.0±0.2) cm^3.

The CR-39 track detector sheets, manufactured by Page Ltd, England, have been cut to (3.5×1) cm^2 size rectangles, and taped to a holder in the middle of the monitoring device (5).

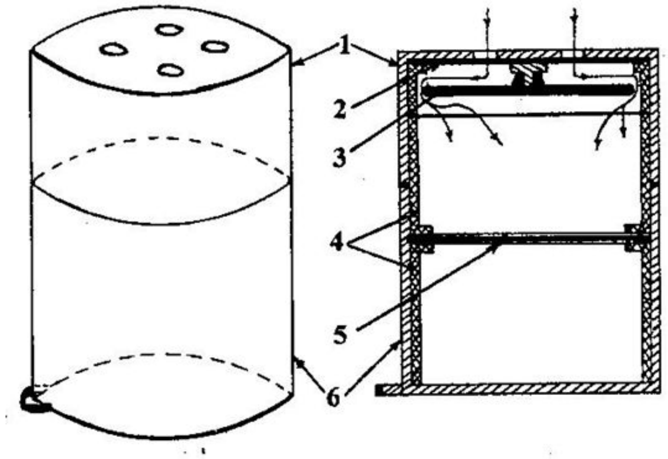

Fig. 1 – The radon monitoring device: general view and details: 1 – end cap with holes for radon entry; 2, 3, 4 – various holding elements for filter and SSNTD foil; 5 – SSNTD; 6 – end cap with ring.

A fine granularity paper filter covers the upper end-cap, allowing exclusive access into the sampler volume to radon gas (no daughter products or aerosols). Exposed to a radon-air mix environment the probe will sample the gas, and the alpha particles, emitted in the decay, will be registered by the nuclear track detector. After exposure, the detectors will be etched and the tracks can be counted with an optical microscope. The track density $\boldsymbol{\rho}$ is proportional to the radon activity concentration $C_{\mathrm{A,Rn}}$ and the time t_{exp}.

$$\rho\ [\text{tracks/cm}^2] = C_{\mathrm{A,Rn}}\ .\ v \cdot t_{\mathrm{exp}} \cdot \varepsilon_{\mathrm{d}}, \tag{1}$$

where v is the sampled volume, and $\boldsymbol{\varepsilon}_{\mathrm{d}}$ the efficiency factor.

The radon probes are calibrated in the lab, in a system designed and built at NIPNE [4,5], using a ^{226}Ra-^{222}Rn equilibrium gaseous source. The method has been attested recently [5] and by further intercomparison will be aligned to international reference. Measurement with this technique is based on a relative method, the reference being the track density value obtained in the controlled geometry of the Ra-Rn source in the calibration system.

The activity of the calibrated Ra source was 2 082±150 Bq.

Each set of measurements was accompanied by a set of witness samples from the same SSTND foil and a set of probes exposed for reference in a dedicated calibration system. The SSNTD samples were all etched together (calibration, in-situ experiment and unexposed witness samples) for 7 hours in a 30% NaOH bath at 70^0C. After counting the alpha particle tracks and calculating the track densities, we are able to correct for detector background (witness track density) and efficiency (calibration). The minimum calibration time was taken to ensure at least a count of 3 times the average background density level obtained in a separate experiment on 12 samples.

The final formula to obtain the radon concentration in the locations investigated is

$$C_{A,Rn,x} = C_{A,Rn,c} \cdot (t_{exp,c} / t_{exp,x}) \cdot (\rho_x / \rho_c). \quad (2)$$

The indices in eq.(2) stand for c – calibration, and x – in-situ experiment respectively, the quantities being the same as above.

The method has been proven reproducible within 3σ [5].

The reference samples for calibration were exposed for 90 days, in order to ensure a good statistic for the track density and compatible values with the readings expected to be found in the in-situ survey.

We have chosen to investigate several dwellings in the winter months, when the radon concentration is typically elevated, in bedrooms, living-rooms, closets, workplaces and basements in the Bucharest region, which is not a radon-prone area. They are situated at different locations, and represent various types: different floors and made of standard building materials – bricks, concrete, etc. Some buildings were new, but most were over 30 or even 50 years old. The basements were either with concrete floors or without any cover, some had ventilation, some were closed. We have exposed about 45 probes in this campaign (2005), most for 6 months (November to May). 2 samples were exposed for 9 months (September to May) and 2 for about 11 months. The basements were: one in Bucharest, with permanent natural ventilation and concrete floor, and other two in Ciorogirla, one with a concrete floor and another without any flooring. The home/office locations were: an old brick house, at ground level (bedrooms, living-room, closet) – in Bucharest, a brick building at first floor (office); a brick+concrete, ground floor apartment (bedroom, closet), about 30 years old, a concrete building, underground level (laboratory) and first floor (office), a concrete+light concrete bricks, a 3rd floor (living-room), in Magurele, and two higher level apartments in blocks of flats in Bucharest (4th, 7th floor, bedrooms, living-rooms, closets).

We expected to obtain values close to the typical European average-to-low radon concentrations in the apartments, independent of building material, with some increase for the less ventilated places, and at ground floor. Building materials contain, in principle, significant radioactive element concentrations, e.g. between 1Bq/kg of ^{226}Ra in wood, 30-65Bq/kg in concrete and tile, to about 700Bq/kg in phosphogips [6], but the samples do not show differences assignable to this effect.

At the same time, the basement data should clearly show significantly increased values, especially where no concrete flooring exists. Our measurements have been performed basically in winter months (mid November to mid May). Choosing the "winter" 6 months, we determined the less favourable values for each location.

3. RESULTS AND DISCUSSION

The choice of samples was based on consideration of the different sources contributing to indoor radon. In principle, the basement locations should yield the highest radon values, as radon comes from the radioactive decays in the soil and underground water. The values will be higher, if the entry of the radon is favoured by cracks and lack of insulation, and if convection is favoured by a temperature/ pressure gradient. If the spaces are used as caves, they are probably less ventilated, and more radon can accumulate in time. If the floor is well insulated and uniform and/or a good ventilation is ensured, this will result in a substantial reduction in the concentration, proving that these measures are helpful choices for indoor level reduction (risk mitigation). Water and gas pipes can add to an increased radon level as well, but the effect of the latter can be usually neglected.

At ground level, in free atmosphere the typical radon concentration is about a few Bq/m^3, while indoor accumulation will yield 10–100 times more, to ~kBq/m^3.

The results obtained cover a wide range of values, but can be grouped in two sets for discussion.

The results on basement indoor radon are listed in Table 1. The beneficial influence of ventilation (T33-III, T16-VIII) and concrete floor (T33-III, T16-VIII, T4-Z, T34-II compare to T37-1, T10-X, T40-VI, T13-IX) is obvious. Special attention is due to the T33-III, T16-VIII samples, which stayed in a cave with a concrete floor and a permanently open window, even in winter. These two samples show a low value of radon concentration. The same is true for labs/offices situated in the basement (T23-13, T19-17) reported in Table 2.

Table 1

Radon concentration results in homes – basements/caves in Bucharest and Ciorogirla

Sample code	Location/ description	ρ [tracks/cm^2]	Time [days]	Radon concentration $C_{A,Rn,x}$ [Bq/m^3]
T33-III	Cave, concrete ventilation, Bucharest	936±39	192.875	200±28
T16-VIII	As above	902±39	192.875	193±27
Average	Cave,concrete, ventilation	"normal risk"		196.5
T34-II	Cave, concrete, Ciorogirla	2310±54	186.02	512±69
T34-II	same	2725±80	186.02	604±82
T4-Z	Cave, concrete, Ciorogirla	2733±80	186.02	606±83
Average	Cave,concrete,new,closed	"elevated risk"		574

Table 1 (continued)

T37-1	Cave, no flooring, Ciorogirla	4625±73	186.03	1025±137
T10-X	Cave, no floor, Ciorogirla	6829±87	186.03	1513±202
T13-IX	As above	5974±114	186.03	1323±178
T40-VI	As above	4531±100	186.03	1004±136
T40-VI	As above	4569±72	186.03	1012±136
Average	Cave, no flooring	"high risk" action mandatory		1175

The results from workplaces and homes are presented in Table 2.

Action levels recommended by ICRP[7] for indoor radon levels are in the range of 200–600 Bq/m^3 , and basically the value accepted is a political decision for every country, depending on geology and affordable costs.

Table 2

Results on radon concentrations in homes and workplaces in Bucharest and Magurele

Sample code	Location/ description	ρ [tracks/cm^2]	Time [days]	Radon concentration $C_{A,Rn,x}$ [Bq/m^3]
	Magurele, NIPNE -workplaces			
T23-17	NPD, basement, lab	763±37	183	172±24
T18-17	Same	803±37	186.785	177±25
T15-16	NPD, ground floor office	379±31	185.98	84±13
T24-15	Same	341±30	185.97	76±12
T35-7	ENPD, first floor, office	589±34	184.903	131±19
T28-11	Same	471±33	184.903	105±16
	Homes, concrete+bricks			
T19-18	Magurele, ground floor, room	295±30	183.67	66±11
T30-19	Same,no ventilation, closet	907±39	183.67	204±28
T8-□	Magurele, 3rd floor, room	899±39	182	204±28
T12-L	Magurele, 3rd floor, room	1225±43	182	277±38
T31-6	Bucharest, 7th floor, room	607±34	174.83	143±21
T11-2	Same	538±34	174.83	127±19
T42-1	Same	560±34	174.83	132±19
T26-4	Same	765±37	174.83	180±26
T22-21	Same, closet	817±38	177.04	190±27
T7-22	Same, closet	728±36	177.04	169±24
T27-:	Bucharest, 4th floor, bedroom	2468±56	192.67	528±71
T5-VII	Same	728±36	197.67	152±22
T2-⊥⊥	Same	632±35	197.71	132±19
T1-⊥	Same, living room	2364±55	197.71	493±67
	Old, brick house, ground floor, Bucharest	Mostly "elevated risk", caution/action recommended		
T38-10	Bedroom	3754±66	272	569±76
T25-12	Bedroom	3029±60	272	459±62
T14-14	Bedroom	1305±44	192.83	279±38
T20-8	Bedroom	1738±48	192.83	372±50

Table 2 (continued)

T32-5	Room	2948±59	192.85	630±85 !!!
T29-3	Room	2503±56	192.85	535±72
T17-V	Hallway	1423±45	192.83	304±42
T41-XI	Hallway	2165±53	192.83	463±63
T21-20	Hallway	1578±46	204.57	318±43
T39-9	Hallway	1558±46	204.57	314±43
T6-Δ	Closet	1928±50	181.9	437±59
T3-H	Closet	1732±48	181.9	393±53
T9-IX/	Another type of MD	1464±46	341.83	177±24
T36-2/	Different type of MD	1828±49	341.83	220±30

The latest WHO documents on radon are even more restrictive, introducing a lower warning level above 100 Bq/m^3 [8].

Accordingly, we will consider the concentrations < 200 as low compared to a probable average level in Romania, the values between 200 and 600 elevated and above 600 Bq/m^3 high risk levels, the threshold when special measures are mandatory (ventilation, insulation).

The systematic of the data obtained in this work is given in Fig. 2. The dotted lines represent the high limits from 2 different surveys: [9](limits: 43–477Bq/m^3, obtained by Dumitrescu et al, in a previous survey in winter (December) with track detectors, on 100 samples), and Cosma et al (updated 2009 [3,10]) (limits: 18-210 Bq/m^3, spring-summer in Bistrita-Nasaud, 27–225 Bq/m^3, winter- spring in Unirea, Alba and 6–149 Bq/m^3, in Agnita, Sibiu, same season, 20–680 Bq/m^3 in Cluj, winter. Note for comparison that in Stei, Bihor, a radon prone area, the high limit for a group of buildings was slightly above 1 000 Bq/m^3!)

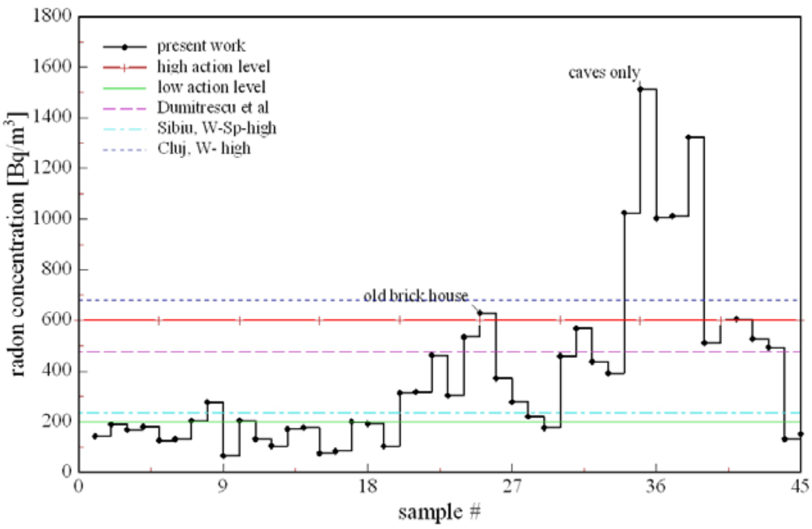

Fig. 2 – Radon concentration distribution on samples in homes and workplaces in the Bucharest region.

4. CONCLUSIONS

Action levels recommended by ICRP[7] for indoor radon levels are in the range of 200-600 Bq/m^3 and the most recent WHO[8] recommendations go even lower to 100 Bq/m^3 . Basically the value accepted after all is a political decision for every country, depending on the local geology and the affordable costs.

In this study we have found somewhat higher indoor radon levels for a non radon-prone area compared to the European average. The present data can be integrated in a national indoor radon database.This proves the necessity to embark on a systematic national survey and assessment program, as we proposed for the near future. For an extensive survey, it is mandatory to use automated read-out, as the technique is very time-consuming, and probes should be doubled for better accuracy.

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