A COMPARISON OF RETROSPECTIVE RADON GAS MEASUREMENT TECHNIQUES CARRIED OUT IN THE SERBIAN SPA OF NIŠKA BANJA

by

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Indoor radon retrospective concentrations were obtained and compared using two radon measurement methods. Both methods rely on the measurement of the long-lived radon progeny ^{210}Pb, collected either on the surfaces (surface trap technique), most frequently glass, or in a volume trap, usually sponge from furniture (volume trap technique). These techniques have been used to retrospectively estimate radon gas concentrations that have existed in dwellings in the past. The work presented here compares the results provided by the surface trap technique devised at the University College of Dublin, Ireland, and the volume trap technique devised at the Scientific Research Center, Mol, Belgium. The field campaign was carried out by the research team of the ECE Laboratory of the Vinča Institute of Nuclear Sciences at the spa of Niška Banja, identified as a region of Serbia with a high indoor radon and ground water radium and radon content.

Key words: indoor radon, retrospective measurement, surface trap, volume trap

INTRODUCTION

Niška Banja, a spa located 9 km east of Niš, Serbia (fig. 1), has been known since the Roman times. It is one of the forty spas in the country, famous for its hot springs with high radium and radon concentrations. The town is located in the quarternary alluvium of the river Nišava, along the border of a karstic limestone re-

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Figure 1. Location of Niška Banja, Southern Serbia

gion and a thick stratum of travertine (groundwater spring deposits). The formation of travertine, in conjunction with high radium concentrations in the spring-waters of Niška Banja, could be the cause of high radium concentrations in the soil of the region. The measured radium concentration in the soil amounts to approximately 900 Bq/kg [1]. Due to the high radium content of the soil, a number of radon surveys have been conducted, both indoors and outdoors [2-5], and the average indoor radon concentrations were found to be largely dependant upon the type of bedrock on which the dwellings were built. Dwellings built on the travertine plains were found to have, on average, radon concentrations of approximately 1.5 kBq/m^3, with some dwellings reaching 2-6 kBq/m^3. For dwellings built on alluvium sediments, the average indoor radon concentrations were found to be 635 Bq/m^3, less than half of those built on travertine deposits, but still well above the Serbian national average of 400 Bq/m^3. The average outdoor radon gas level per annum was found to be 57 Bq/m^3, far higher than the typical worldwide value of 10 Bq/m^3 [6]. Due to the high values of indoor and outdoor radon concentrations measured, the Niška Banja region has been classified as an area with a high natural radon concentration.

The majority of radon gas measurements carried out indoors as a part of these surveys were contemporary radon gas measurements [7]. However, from 2004 to 2005, a number of retrospective radon gas measurements were also conducted in various dwellings throughout the region. Both surface trap and volume trap measurements were made and this paper compares the two types of measurements in dwellings where in the same indoor space both measurements were made.

RETROSPECTIVE MEASUREMENT TECHNIQUES

Retrospective assessment of radon exposure is a technique which was developed in recent years. Retrospective methods are based on measuring ^{210}Pb or ^{210}Po due to radon which has accumulated in solid or porous media over many years and which is, therefore, in principal proportional to the cumulative radon exposure. By means of the retrospective method, past changes in radon exposure are measured, for example, in homes previously inaccessible for radon testing or subject to temporal and spatial changes in radon concentration. Basically, two methods of retrospective radon assessment exist. These methods are based on the build-up of low levels of ^{210}Pb and, consequently, ^{210}Po activity that can be related to long term average radon concentration. In the surface trap technique, airborne radon decay products deposit on glass surfaces. Subsequently, they can be implanted through alpha recoil and remain fixed in the near surface layer of the glass where they can be detected through alpha decay by means of track-etch detectors or pulse ionization chambers [8]. Any bulky, sufficiently porous material, with known age and history, can serve as a volume trap. It suffices that radon gas can diffuse freely through the material on a time scale shorter than that of radon half-life, so that the radon concentration inside the volume trap is continuously representative of the radon concentration outside the volume trap. The material should, however, be such that radon decay products can not migrate into or out of the volume trap. Moreover, the volume trap material should not contain a significant natural ^{210}Po background, so as to disturb the measurements.

The surface trap technique

A technique using CR-39 detectors (polyallydiglycol carbonate) and LR-115 detectors (cellulose nitrate) known as the "CR-LR difference technique" aimed at large-scale fieldwork was used in this investigation to measure ^{210}Po [9]. The alpha sensitivity of CR-39 (plastic material), of the same type typically used in ambient radon detectors, makes activity measurements on glass using an alpha track-etch detector possible, fulfilling at the same time the need for a practical and economical way of estimating long term radon concentration. This method allows measurement without removing the glass from the site. Another advantage is that such detectors are passive and require no power supply. In this technique, a few square centimeters of each of the alpha track detectors are mounted side by side on a chosen flat glass surface. The glass is cleaned and a piece of CR-39 plastic is affixed to its surface and left in place for a few weeks. For standard etching conditions, LR-115 is sensitive to alpha particles in an energy window in the approximate range of 1.2-4.8 MeV, while CR-39 is sensitive to alphas with energies of less than 1 MeV, to those much greater than the maximum energy (7.68 MeV) emitted by the radon progeny. As the ^{210}Po alpha energy is 5.3 MeV, the LR-115 will not record tracks from the surface trapped ^{210}Po, but will, nevertheless, produce tracks proportional to the intrinsic alpha activity naturally present in the glass. The CR-39 track density will thus be a mixture of tracks due to the implanted ^{210}Po alpha emissions and those from the intrinsic activity of the glass. After being deployed for t hours, the following expression gives the surface ^{210}Po activity for a glass surface onto which, side by side, CR-39 and LR-115 detectors were mounted

$$A_{\text{Po}} = \frac{CR - b \, LR}{k \, t}$$

where A_{Po} means ^{210}Po activity in Bq/m^2 and CR and LR represent net tracks per cm^2 for CR-39 and LR-115 detectors, respectively; $b = CR/LR$ is the intrinsic alpha activity track density ratio for unexposed glass, and k is the pulse ionization chamber factor which deter-

mines CR-39 sensitivity to ^{210}Po. The values of b and k depend on the actual etching regimes and track acceptance criteria used. For standard procedures, values of $b = 1.97$ and $k = 0.081$ tracks per cm^2 / Bqh per m^2 were obtained [9].

The typical exposure period used in this work for the alpha track detectors mounted on glass was about three to four months. The plastic is then removed and the alpha tracks resulting from implanted ^{210}Po that have been etched into its surface are counted. The alpha-particles from the glass produce latent damage tracks in the plastic that can be easily developed and measured. The track generation rate is then a measure of the alpha-activity of the glass surface. Radon concentrations obtained in this way, correlated to the radon decay products, can be used to create radon exposure histories [9-11].

The Cr-39 alpha track densities recorded for these exposures were mainly within the range of 5 to 25 tracks per mm^2. Measured A_{Po} values ranged from about 3 to 30 Bq/m^2, with individual values as high as 50 Bq/m^2.

The use of the surface trap technique in retrospective radon measurements aimed at epidemiological studies is now becoming more common. Besides Serbia [12-15, 3], studies involving measurements on glass have been performed in USA, Ireland, Norway, Germany, and Sweden [16-22, 10, 23].

The volume trap technique

The other technique in use is the volume trap measurement technique. It, too, utilises the measurement of ^{210}Po surface activity arising from the decay of the long-lived radon progeny ^{210}Pb. However, in this case, the ^{210}Po activity is not measured on a glass surface or mirror. Instead, the ^{210}Po activity measured is that which has been deposited in spongy materials such as mattresses or cushions. This idea was first proposed in 1994 [24]. Spongy materials allow the diffusion of radon gas through their pores but, in normal circumstances, radon progeny cannot diffuse easily into the material, so that the progeny deposited at depth in the material is attributed entirely to the radon gas itself. Thus, the ^{210}Po activity in the centre of the material correlates well with the radon gas it has been exposed to over its lifetime, allowing for a retrospective radon gas measurement to be made.

The measurement material for the volume trap technique is typically a 100 cm^3 polyester foam sample from mattresses or cushions taken from a dwelling. In order to exclude any surface effects, the sample is normally dissected from inside the exposed material. Once the sample has been removed from the material, it is kept in a radon-free environment for approximately 138 days (one half-life of ^{210}Po), in order to reach radioactive equilibrium. The next stage of the process is to chemically separate the ^{210}Po activity from the material. This is done via several steps [25, 26].

Firstly, the samples are dissolved in NaOH (14 molar) and hydrolyzed for 5 to 10 hours. After cooling, HCl (8 molar) is added and the solution dried under infra-red lights. This residue is then dissolved in HCl (12 molar) in order to eliminate all NaOH. The final residue is then dissolved in 4.2 ml of HCl (12 molar) and the sample is made up to 100 ml using water. This solution is mixed with 100 mg of ascorbine acid and covered by silver plate, turned onto its head, and then the polonium is autodeposited onto the plate for over 48 hours. The activity of ^{210}Po is then determined via alpha spectrometry using a PIPS detector. The amount of ^{210}Po lost during this procedure is determined by using ^{208}Po as a tracer material. Using a conversion factor that takes into account the porosity and the age of the sample material, retrospective radon gas concentration is then calculated from the said ^{210}Po activity. These detectors have been used in surveys in the past [27, 23] and a subsequent study [15] found this measurement technique to be very reliable.

However, like all radon measurement techniques it has its disadvantages. The major one being that this technique is destructive (*i. e.* that the material is removed from the dwelling and destroyed during analysis, as opposed to the surface trap technique), so that occupants are sometimes reluctant to supply the needed samples. In addition, it has been established that, in very dusty circumstances, direct penetration of radon progeny from the outside of a sample to the centre is possible, so that extra caution is needed in these conditions.

FIELD CAMPAIGN: DEPLOYMENT AND RETRIEVAL OF DETECTORS ON SITE

All of the dwellings in Niška Banja were examined by a research team of the Electrochemical Etching Laboratory of the Vinča Insitute of Nuclear Sciences, Belgrade, Serbia, and surface trap monitors installed at choosen sites within them. The locations and the glass surfaces for the surface monitors within the rooms were selected according to appropriate criteria. In addition to this, a detailed questionnaire for each room of interest was set up, recording a number of other relevant factors related to the aerosol concentration, ventilation rate, and surface to volume ratio, past and present. These details were then used in conjunction with the modified Jacobi room model [28], in order to better estimate the average radon concentration of the glass objects regarding the period they have been exposed to over their lifetime in the room.

In addition, during these investigations, a number of these rooms had a sample of sponge material taken from them for a volume trap analysis, the age of the material also beeing noted in the questionnaire.

These samples were then sent to, treated and analysed at the radiochemical laboratory of the Scientific Research Center (SCK CEN), Mol, Belgium.

Surface trap monitors were left *in-situ* for a period of approximately 3 months before being removed by an experienced person and sent back for analysis to the experimental laboratory of the School of Physics at the University College Dublin (UCD), Ireland, in an electrostatically sealed bag. Surface trap detectors were etched as soon as they reached UCD. Due to the extremely low intrinsic background of the transported material, relatively short period in post and high ^{210}Po activity, the background corrections of the detectors exposed were not calculated.

After being etched, the CR-39 and LR-115 detectors were counted and a retrospective estimate of radon concentration obtained. These results were then compared to the volume trap measurement results made by the research team of the SCK CEN.

RESULTS

Surface trap measurements of a total of 39 dwellings were conducted. At the least, one surface monitor was placed in each of them, with the majority having two surface trap measurements per dwelling installed. This resulted in a total of 61 surface trap measurements.

Eighteen dwellings had volume trap measurements made in them, with a significant portion of them having at least two measurements made per dwelling, resulting in a total of 27 volume trap measurements.

All in all, there were 20 rooms from 16 different dwellings (10 bedrooms, 9 living rooms, 1 storageroom) where at least one surface trap measurement and one volume trap measurement was made. The results obtained can be seen in tab. 1. For rooms that had more than one volume trap detector placed in them, the overall average of the radon concentrations from the detectors was used.

The ^{210}Po surface activity measurements have been normalised to 20 years and a summary of the volume and surface trap measurements can be seen in tab. 2.

The ratio of the volume trap to surface trap measurements was then calculated and the results presented in fig. 2. The volume trap estimate differs greatly from the surface trap estimate for room number 12, as can be seen in the graph. Both the volume and surface trap results were revisited for this sample and it was noted that the volume trap sample was particularly dusty. Since the dust could have had an affect on the estimated retrospective radon gas concentration, this sample result was ommited from the rest of the analysis.

The same ratios as before, without room 12, are displayed in a more appropriate scale in fig. 3.

Figure 3 indicates that the majority of the ratios of the surface to volume measurements are close to one. The ^{210}Po surface activities normalized to 20 years for the 19 rooms were then compared to the vol-

Table 1. Volume and surface trap measurements from dwellings in Niška Banja

Dwellings		Volum trap	Surface trap	
Room number	Location	Radon concetration [Bqm^{-3}]	^{210}Po concentration (normalised to 20 years) [Bqm^{-2}]	Radon concentration [Bqm^{-2}]
1	Bedroom	510	13.32	570
2	Bedroom	850	21.42	998
3	Bedroom	6000	30.77	1347
4	Bedroom	7200	64.03	2459
5	Living room	26600	560.48	26107
6	Living room	13900	128.69	10.624
7	Living room	8750	13.03	1076
8	Living room	1700	21.10	983
9	Storage room	1400	45.00	1926
10	Bedroom	2000	24.75	1153
11	Living room	2800	138.08	5304
12	Living room	41800	117.93	453
13	Bedroom	480	31.33	1204
14	Livin room	1100	36.31	2715
15	Bedroom	3400	56.04	1846
16	Living room	360	4.92	189
17	Bedroom	610	18.60	714
18	Bedroom	2000	8.05	309
19	Living room	510	12.56	482
20	Bedroom	12400	128.00	5943

Table 2. Summary of volume and surface trap measurements

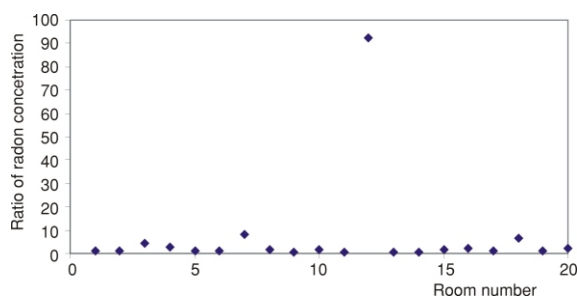| Technique	Activity	Minimum	Maximum	Average
Volume trap	^{222}Rn activity [Bqm^{-3}]	360	41800	6719
Surface trap	^{210}Po activity [Bqm^{-2}]	5	560	74
	^{222}Rn activity [Bqm^{-2}]	189	26107	3320

Figure 2. Ratio of volume to surface trap estimated radon concentration

ume trap retrospective radon gas estimates in order to determine the degree of correlation that existed be-

tween these two different methods. A reasonably good correlation (0.77) was found (fig. 4).

Retrospectively estimated surface trap radon gas concentration was then compared to the volume trap radon concentration (fig. 5). It was found that the correlation between these two methods was 0.83. This value is better than the correlation between the ^{210}Po surface activity and the volume trap radon estimate.
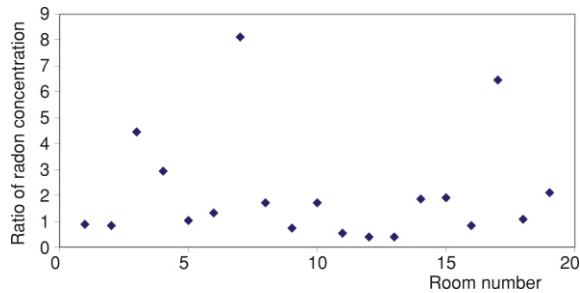

Figure 3. Ratio of volume to surface trap estimated radon concentration with room number 12 omitted
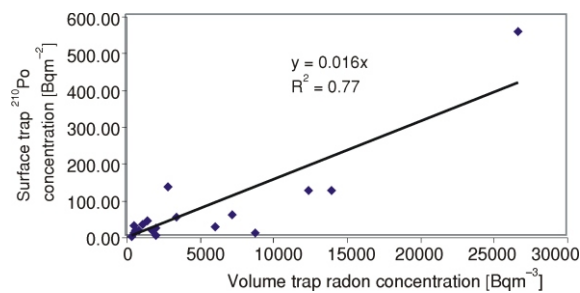

Figure 4. A comparison of the ^{210}Po surface trap (normalized to 20 years) to volume trap retrospectively estimated radon concentration
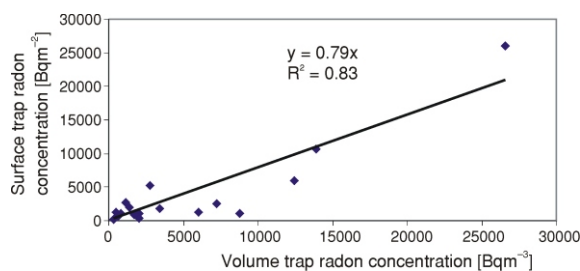

Figure 5. A comparison of surface to volume trap estimated retrospective radon concentration

CONCLUSIONS

This study indicates a very high degree of correlation (0.83) between two retrospective radon gas estimation techniques. Comparing the ^{210}Po surface activity measurements to the volume trap retrospective radon estimates, however, this correlation decreases.

Both methods confirmed the results obtained in previous research done in Niška Banja that the dwellings built on travertine plains were found to have very high radon gas concentration. On average, more than 3 kBq/m^2 were found in some dwellings by surface trap technique. By volume trap technique, on average, more than 6 kBq/m^3 was found in the dwellings built on the same travertine plains. On alluvium sediments, the average indoor radon concentration was below 1 kBq/m^2. This is less than half than that of those on travertine deposits, but still well above the Serbian national average.

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ПОРЕЂЕЊЕ РЕЗУЛТАТА МЕРЕЊА РАДОНА У НИШКОЈ БАЊИ РЕТРОСПЕКТИВНИМ ТЕХНИКАМА

Ретроспективне концентрације радона у затвореним просторијама одређене су и упоређене двема мерним методама. Обе методе заснивају се на мерењу дугоживећег потомка радона ^{210}Pb, депонованог било на стакленим површинама (површинско узорковање), или у унутрашњости сунђерастог материјала пореклом из намештаја (запреминско узорковање). Ове технике су употребљене да ретроспективно процене концентрацију радона која је у прошлости била присутна у том простору. У раду се упоређују резултати засновани на техници површинског узорковања развијеној на Универзитету у Даблину, Ирска и техници запреминског узорковања, развијеној у Научно-истраживачком центру у Молу, Белгија. Теренски рад обављен је у Нишкој Бањи, идентификованој зони са високим нивоом основног зрачења из природе и високим садржајем радијума у води, од стране истраживача ЕЦЕ лабораторије Института "Винча". Добијени резултати мерења радона површинским и запреминским узорковањем показали су високу корелацију, $R^2 = 0.83$.

Кључне речи: радон у затвореној просторији, ретроспективно мерење, површинско узорковање, запреминско узорковање