MEASUREMENT OF ^{222}Rn BY ABSORPTION IN POLYCARBONATES – RESEARCH AND PRACTICE

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Abstract

In the last few years, we have gathered experience in measurement of ^{222}Rn in air, water and soil-gas by absorption in polycarbonates. The method employs the remarkable absorption ability to noble gases of some polycarbonates, like Makrolon$^®$ (the basic constructive material of CDs/DVDs). This report summarizes the key results of research and practical applications of this method. Calibration procedures, including a posteriori calibration, are described. Comparison between retrospective measurements (by CDs) and estimates based on conventional prospective ^{222}Rn measurements in dwellings are shown. The method is also used for measurement of ^{222}Rn in water and soil-gas. Comparisons with conventional methods for ^{222}Rn measurement in air, water and soil-gas are made and a good agreement is obtained. The sensitivity of the method covers the whole range of concentrations that are of practical interest. The potential for wide scale practical applications of this method and possible directions for further progress are discussed.

Introduction

The remarkable absorption ability of some polycarbonates like Makrofol$^®$, Makrolon$^®$ or equivalents has been employed for quantitative measurements of ^{222}Rn first in 1999 (Pressyanov et al., 1999). Since then several studies were dedicated to the development of practical methods for measurements of radioactive noble gases – mainly ^{222}Rn (Pressyanov et al. 2000; 2004a; 2007), but also ^{85}Kr and ^{133}Xe (Pressyanov et al. 2004b). As a part of the progress in this field a method for precise retrospective ^{222}Rn measurements by CDs/DVDs has been proposed (Pressyanov et al., 2001; 2003). In the last AARST 2007 symposium the performance of this method in laboratory studies has been summarized (Pressyanov, 2007). Herewith, we present results from progress achieved in measurements under real conditions. In the last year, a number of retrospective measurements in dwellings were made. The procedure of *a posteriori* calibration was tested and utilized. For six of the studied dwellings data from past ^{222}Rn measurements by conventional methods were available. A comparison between CD-results and estimates based on conventional measurements was made and a very good correlation was observed.

Methods based on polycarbonates have also been successfully used for [222]Rn measurements in water (Pressyanov et al., 2007) and soil gas. We present pilot results obtained under real conditions. Again, we observe a good correlation with conventional methods. Based on experimental facts we tend to conclude that polycarbonates can be applied for quantitative [222]Rn measurements in the three media of interest – air, water and soil gas. The estimated sensitivity suggests that it covers the whole range of [222]Rn concentrations in air, water and soil gas that are of practical interest.

Materials and Methods

In the last decade, we have used different specimens made of Makrofol or Makrolon for [222]Rn measurements. However, the results presented in this report were obtained mainly by measurements with commercial CDs. For retrospective measurements in real dwellings, we have used CDs voluntary provided by homeowners. By interviewing them, the CDs were dated as accurately as possible. In all cases, we managed to date the CDs with accuracy of 1 year or better. This successful dating was possible because of the relatively small size of the study and the possibility for personal communication with the involved inhabitants. From some dwellings more than one CD was obtained, each of different origin and/or age. The CD-specimens used for measurements in water or soil gas were normally exposed for 1-2 weeks.

Further, all CDs were treated in the same manner. First, they were left to degas for at least 2 weeks (this is important when CDs are exposed for relatively short time – e.g. in water or soil gas). After that, a surface layer was removed by chemical pre-etching (CPE) at 30^0C with aqueous solution of 52% KOH (m/v) and 40% methanol (m/v). In the current study a layer with thickness of 80μm was removed (which is just enough to ensure that no tracks from Rn-progeny deposited on the surface are formed), but in order to expand the upper limit of measureable concentrations a thicker layer could be removed. After that pieces of the CDs were etched electrochemically (ECE) at 3.3 kV effective high voltage (6 kHz frequency) at 25^0 C for 3 hours. A HV generator produced by Ekotronik Ltd. (Prague, Czech Republic, www.ekotronic.cz) was used. The etching solution was mixture of ethanol with 6N KOH solution with 1:4 volume ratio. After ECE the CDs were scanned by a computer scanner and the tracks were counted by dedicated software. The track density at a certain depth (> 80μm) is proportional to [222]Rn concentration integrated over the exposure time.

The calibration is made by exposing the CD-specimens to controlled [222]Rn concentrations. After exposure the CDs must also be left to degas for at least 2 weeks, before CPE and ECE. The calibration factor (CF) is defined as *net track density/integrated [222]Rn concentration*. One feature of the CD method is the possibility for individual *a posteriori* calibration of each CD. For this purpose the CD taken for analysis is cut in pieces. One is then exposed to controlled [222]Rn concentrations and the others not. Further, after etching and counting α-tracks individual *a posteriori* CF is determined using the difference between the track densities of the exposed for calibration piece of the CD and that of the original piece of the same CD.

For measurements in soil gas the calibration factor for air was applied. For measurements in water an independent calibration by exposure to controlled ^{222}Rn concentrations in water was made.

Results

Calibration

In this report we emphasize on *a posteriori* calibration for measurements in air. Pieces of CDs that have already been exposed in dwellings were put in 50 L calibration chamber, where artificial ^{222}Rn concentration was created by a certified ^{222}Rn source. During exposure reference measurements were made by a calibrated radonometer, AlphaGUARD (Genitron GmbH, Germany). In these experiments the individual CF of each CD was obtained. In addition, the results were used to compare the variance in the properties of different CDs as radon detectors. Results are shown in Fig. 1. As seen, the maximum deviation of an individual CF from the average one is about 20%.
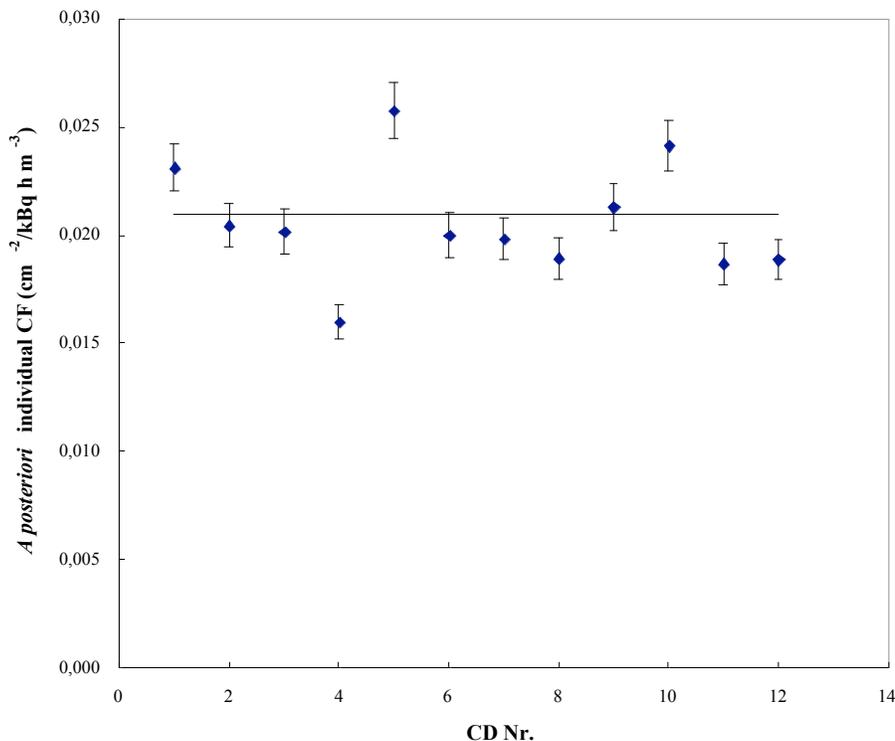

Fig. 1. Individual *a posteriori* calibration factors of "old" CDs. The error bars represent counting statistics uncertainty. The horizontal line is *a priori* CF for a set of new CDs.

For calibration in water we used a water sample with high ^{222}Rn concentration (taken from the mineral spa "Momin prohod" in Bulgaria). The reference measurement was done by gamma spectrometer with HPGe detector (relative efficiency 24.9%,

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resolution (FWHM) 1.9 keV at 1332 keV reference gamma line of ^{60}Co). The pieces for calibration were exposed for 48 h in hermetic vessels. During the exposure regular gamma spectrometry measurements confirmed that there was no leakage of radon. The results obtained for CF at 80μm depth in air and in water are:

- CD in air, *a priori* determined CF (for a large group of unused new CDs): 0.021 ± 0.003 cm^{-2}/kBq h m^{-3}. The uncertainty in this and others CFs is due to counting statistics, variation between different CDs and uncertainty in reference concentrations;
- CD in air, *a posteriori* determined CF (for used CDs exposed in dwellings - Fig. 1): 0.0206 ± 0.0027 cm^{-2}/kBq h m^{-3};
- CD in water (for a limited number of CDs): 0.067 ± 0.010 cm^{-2}/kBq h m^{-3}.

Notably, the average CF in *a posteriori* calibration of "old" CDs is the same as the *a priori* CF obtained for a set of new CDs without previous "radon history".

Real measurements and comparison

In real retrospective measurement the following questions were first addressed:

a) Whether different CDs taken from the same place give comparable results?

b) Will CD results match with independent estimates of retrospective ^{222}Rn concentrations based on past ^{222}Rn measurements by conventional methods?

To answer the first question we tried, whenever possible, to obtain more than one CD for analysis. In two dwellings, one with high and another with average ^{222}Rn concentrations we succeeded to obtain four CDs of different origin and age. The results obtained with these CDs are shown in figures 2a and 2b. As can be seen, different CDs give the same retrospective ^{222}Rn concentration within the experimental uncertainty. This conclusion is supported by results from other houses, from which we have obtained two or three different CDs for analysis.
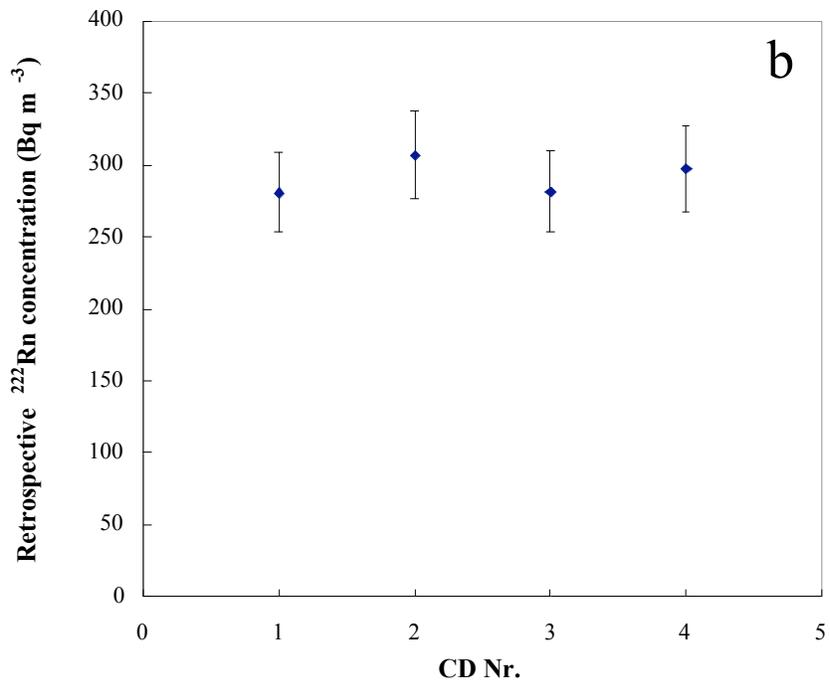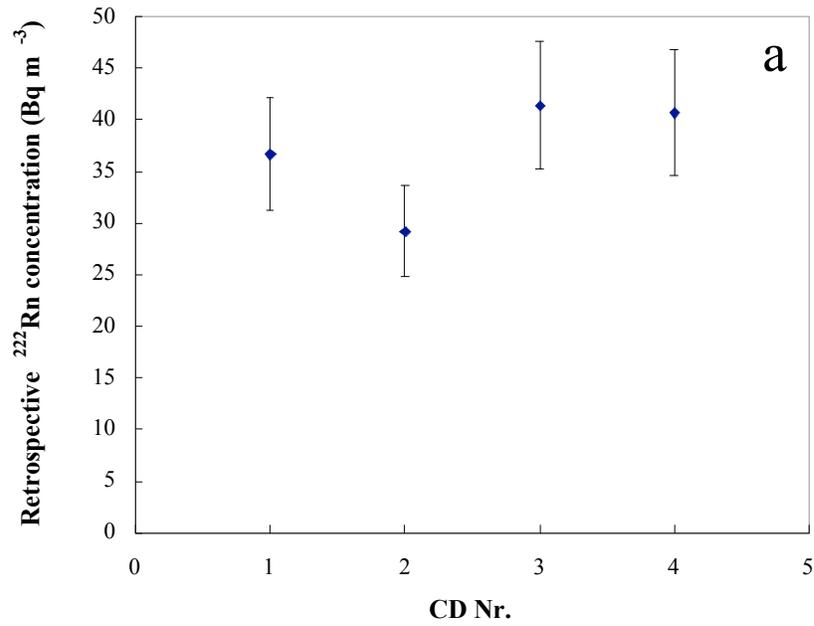
Fig. 2. Average retrospective ^{222}Rn concentrations measured by different CDs from the same place, a) dwelling with average ^{222}Rn concentration, b) dwelling with elevated ^{222}Rn concentration.

In six of the studied houses past [222]Rn measurements were available. They were done by conventionally used diffusion chambers with α-track detectors of Kodak-Pathe LR-115 type II. The diffusion chambers used are traceable to a primary international [222]Rn standard (Picolo et al., 2000). There is a very good correlation between the results obtained from CDs and those estimated from past measurements by diffusion chambers (Fig. 3). We should note however, that 5 out of the 6 dwellings are from risk areas, without any change and repair for the period of CD storage and therefore great variations in the average annual concentrations are not expected. The remaining one (that at about 20 Bq m^{-3}) is the flat of one of the authors (DP) with well-known radon history. More comparisons will be made in the future, but the pilot results seem very promising.
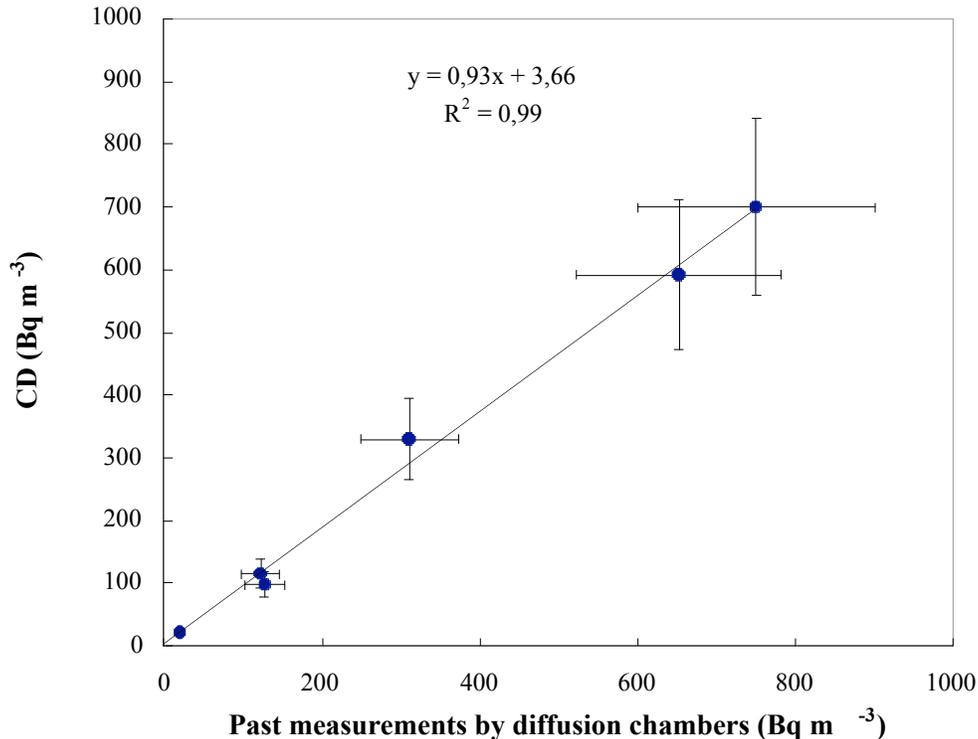

Fig. 3. Comparison between retrospective [222]Rn concentrations measured by CDs and these determined using past measurements with diffusion chambers.

Two strategies for measurements in water and soil-gas by polycarbonates are possible, depending on whether a radiometric laboratory capable of measuring beta and gamma radiation is available closely. If it is, the exposed specimens can be used to sample [222]Rn and later to be measured for beta or gamma radiation from [222]Rn progeny related to radon absorbed in the polycarbonate (Pressyanov et al., 2007). The measurement should start shortly after the specimen is taken (within a few days, but at least 3 h after the end of exposure, so that the [222]Rn progeny plate-out decays). The second strategy we consider more promising for the large radon community. In it the tracks formed inside the polycarbonate sample due to absorbed radon and its progeny are developed and counted. After being exposed in water or soil gas, the samples are forwarded to an etching

laboratory for analysis. This allows measurements in any water supply source (incl. directly in the source – without water sampling) or any terrain where soil gas radon is usable for evaluation of the radon potential. To test this approach for soil-gas, we have exposed four CDs in parallel with conventional diffusion chambers in four points in the region of Sofia, Bulgaria. In order to prevent direct contact with subsoil water the specimens were packed in polyethylene envelopes. The influence of polyethylene packaging had been studied in dedicated laboratory experiments. These studies have demonstrated that [222]Rn penetrates by diffusion through the envelopes and that there is no statistically significant difference in the signal from packed and unpacked polycarbonates exposed under the same conditions. The detectors were buried at 60 cm depth and left for two weeks. The results are shown in Fig. 4. Given the small size of this study and the fact that the concentrations are close or slightly above the typical background concentrations of [222]Rn in soil-gas, we consider the agreement reasonably good.
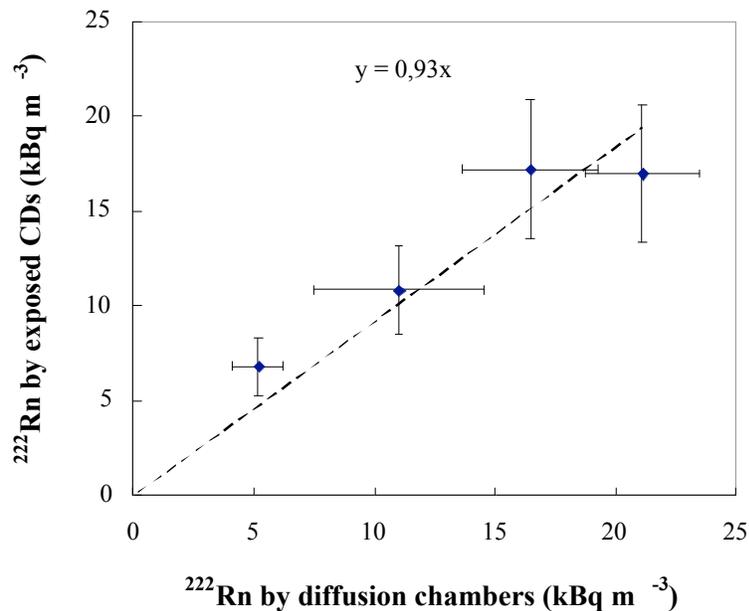

Fig. 4. Comparison between [222]Rn concentrations in soil gas measured by CDs and diffusion chambers buried at 60 cm depth.

For water, we have up to now made a comparison only within the "first strategy" mentioned above (by beta measurements of the sample), following the procedure used in previous laboratory studies (Pressyanov et al., 2007). The results are shown in Table 1. The parallel measurements of water samples were done by gamma-spectrometry with HPGe detector. Again, within the experimental uncertainty we have a good correspondence with parallel conventional measurements, based on water sampling.

Table 1. ^{222}Rn concentration in private water supply sources determined by exposure and measurement of polycarbonate specimens and by laboratory analysis of water samples.

Water source Nr.	^{222}Rn measured by polycarbonates in the source	^{222}Rn measured by gamma spectrometry of water samples
1	218 ± 21 Bq L^{-1}	206 ± 31 Bq L^{-1}
2	153 ± 17 Bq L^{-1}	192 ± 29 Bq L^{-1}
3	126 ± 40 Bq L^{-1}	130 ± 40 Bq L^{-1}

The evaluation of the sensitivity of the method (applied by α-track etching of exposed CDs) and its usable range are shown in Table 2. As seen, the method appears to cover the whole range of concentrations that are of practical interest in air, water and soil-gas.

Table 2. Useful range of concentrations that can be measured by CDs. The lower limit is the minimum detectable concentration. The upper limit corresponds to "saturation" track density of 2000 cm^{-2}. At that level many tracks overlap with others and this hampers track counting.

Measurement	Lower limit	Upper limit
CD exposed in air for 10y - α-tracks at depth 80μm beneath the surface	3 Bq m^{-3}	3000 Bq m^{-3}
CD exposed in air for 10y - α-tracks at depth 200μm beneath the surface	21 Bq m^{-3}	21300 Bq m^{-3}
CD exposed in water for 1 week - α-tracks at 80 μm	0.5 Bq L^{-1}	510 Bq L^{-1}
CD exposed in water for 1 week - α-tracks at 200 μm	3.3 Bq L^{-1}	3600 Bq L^{-1}
CD exposed in soil-gas for 1 week - α-tracks at 80 μm	1500 Bq m^{-3}	1.5×10^{6} Bq m^{-3}
CD exposed in soil-gas for 1 week - α-tracks at 200 μm	10500 Bq m^{-3}	1.05×10^{7} Bq m^{-3}

Conclusions

The presented results show the practical performance under real conditions of the method for ^{222}Rn measurements based on absorption in the polycarbonate material of CDs/DVDs. Results from a posteriori calibration and field studies reveal:

- The variation in individual CF of different studied CDs is within 20%. If one targets accuracy substantially better than 20% an individual *a posteriori* calibration can be recommended;

- Different CDs from one and the same dwelling give generally similar results. The pilot comparison with independently estimated retrospective ^{222}Rn concentrations shows a very good correspondence;
- Quantitative measurements in air, water and soil-gas are possible over practically the whole range of interest.

The good performance of the method under real conditions let us strengthen the conclusion that the considered method has a potential for precise measurements and for large-scale practical use. We plan to implement this method in real measuring campaigns and to continue, whenever possible, parallel analysis by conventional methods.

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