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# Application of scintillation counting using polycarbonates to radon measurements



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#### HIGHLIGHTS

• An approach for <sup>222</sup>Rn measurement by scintillation counting is proposed.

• Its application to *a posteriori* calibration of compact disks is demonstrated.

• Pilot results for application to radon in soil-gas measurements are presented.

• The approach is very suitable when high radon activity concentrations (>2 MBq/m<sup>3</sup>) are to be measured for several days.

• The approach may be suitable when fast, screening radon-in-soil gas measurements are necessary in a large number of points.

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#### ABSTRACT

This work proposes an approach for radon (<sup>222</sup>Rn) measurement based on scintillation counting of polycarbonate specimens. The proposed technique takes advantage of the high absorption ability of polycarbonates to <sup>222</sup>Rn and of the fact that radiation emitted by <sup>222</sup>Rn and its progeny causes the polycarbonate material to emit light. The theoretical background behind the proposed technique is presented and its application to two types of <sup>222</sup>Rn measurements is demonstrated. The first application of the proposed technique is in the *a posteriori* calibration of compact discs for retrospective <sup>222</sup>Rn measurements, where it is sometimes necessary to measure <sup>222</sup>Rn concentrations higher than 2 MBq/m<sup>3</sup> for several days. It is demonstrated that the application of the proposed technique increases the range of applicable <sup>222</sup>Rn concentrations in the *a posteriori* calibration of CDs and adheres to its time and cost efficiency. The second application of the proposed technique is to soil-gas <sup>222</sup>Rn measurements, which were performed in different terrains, covering <sup>222</sup>Rn concentrations between 3 and 1200 kBq/m<sup>3</sup>. It is found that the measurements by scintillation counting of polycarbonates are consistent with the reference measurements by diffusion chambers with Kodak Pathe LR II detectors and very good linear correlations between the techniques are observed. The results from this study imply that the scintillation counting of polycarbonates may be suitable when fast, screening radon-in-soil gas measurements are necessary in a large number of points.

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#### 1. Introduction

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The compact disc method (CD method) for retrospective measurements of <sup>222</sup>Rn was proposed by Pressyanov et al. (1999, 2000, 2001) and ever since has been applied intensively for measurements of <sup>222</sup>Rn in human dwellings and working environment (Pressyanov et al., 2012, 2010). The method relies on the fact that the polycarbonate material, of which the compact discs (CDs) are made, absorbs <sup>222</sup>Rn and is a solid state nuclear track detector. Thus, the track density at depths greater than 80  $\mu$ m beneath the CD surface is proportional to the integrated <sup>222</sup>Rn concentration to which the CD was exposed (Pressyanov et al., 2003, 2004; Pressyanov, 2010). Currently the CD method seems to be the most accurate retrospective <sup>222</sup>Rn measurement method (Pressyanov et al., 2012). The accuracy of the method is mainly due to two reasons: first, the signal (track density) at depths greater than 80  $\mu$ m does not depend at all on the radon progeny concentrations in the air and, second, the method allows determination of the

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individual sensitivity to <sup>222</sup>Rn of each CD. The latter is termed a posteriori calibration of the CDs and facilitates the establishment of a traceability chain of the <sup>222</sup>Rn measurements by CDs to reference standards (Pressyanov et al., 2013). It is performed as follows: First, a quarter of the CD that is to be analyzed is cut. The quarter is etched and the density of the tracks is determined. This track density is proportional to the integrated <sup>222</sup>Rn concentration to which the CD was exposed. Then, a second guarter of the same CD is cut and exposed in the laboratory to known, high <sup>222</sup>Rn concentration. After this additional exposure the second quarter is etched and its track density is determined. The track density increment due to the *a posteriori* exposure is then determined. The calibration coefficient of the CD is the proportionality coefficient between the track density increment and the integrated <sup>222</sup>Rn activity concentration to which the CD was exposed in the *a posteriori* exposure.

To complete the *a posteriori* calibration in a reasonable time (days) the CDs should be exposed to high <sup>222</sup>Rn concentrations, of the order of 1–2 MBq/m<sup>3</sup> or higher. However, such <sup>222</sup>Rn concentrations are on (or beyond) the upper limit of the measurement range of reference <sup>222</sup>Rn monitors. For example, the upper limit of the RAD7 <sup>222</sup>Rn detector is 0.75 MBq/m<sup>3</sup> (Durridge, 2011) and that of the AlphaGUARD portable <sup>222</sup>Rn monitor is 2 MBq/m<sup>3</sup> (Saphymo, 2014). Thus, when higher than 2 MBq/m<sup>3</sup> <sup>222</sup>Rn concentrations are necessary for *a posteriori* calibration of CDs these cannot be monitored with a reference instrument and alternative technique might be useful in that range. In addition, the routine use of reference instruments like RAD7 and AlphaGuard in the *a posteriori* calibration leads to their increasing contamination with <sup>210</sup>Pb/<sup>210</sup>Po which cannot be compensated. Therefore, new techniques for high <sup>222</sup>Rn concentration measurements are necessary to make the routine a posteriori calibration of CDs time and cost efficient.

Soil-gas radon (<sup>222</sup>Rn) measurements, on the other hand, provide basic information for risk mapping (Kemski et al., 2006). In conjunction with geological and soil investigations they can serve for classification and regionalization of the geologically induced risk for high indoor <sup>222</sup>Rn concentrations (Kemski et al., 1996). The approach to use soil-gas <sup>222</sup>Rn concentration for mapping of the geogenic <sup>222</sup>Rn potential has been applied in some countries like Germany (Kemski et al., 2001) and Hungary (Szabó et al., 2014). In situ soil-gas <sup>222</sup>Rn measurements also provide important information when <sup>222</sup>Rn mitigation is to be performed (see, for example Cosma et al. (2015)). Soil-gas <sup>222</sup>Rn measurements are also important in geology, where anomalies in the soil-gas concentrations in faults are considered as potential earthquake precursors (see, for example Cicerone et al. (2009) and the references therein) or precursors of volcanic seismic activity (Cigolini et al., 2013). An extensive review of the relationship between soil and spring gases (incl. <sup>222</sup>Rn) and both tectonic and seismic activities is published by Toutain and Baubron (1999). In view of their importance in various fields, a variety of radon soil-gas measurement techniques have been developed. These include grab sampling, integrating and continuous measurements performed by variety of detectors like scintillation detectors, solid state nuclear track detectors and spectrometers. An extensive review of the instrumentation for <sup>222</sup>Rn in soil gas measurements in earthquake research is published by Papastefanou (2002).

The objective of this work is to propose and demonstrate feasibility of a new technique for <sup>222</sup>Rn measurements. The technique uses polycarbonate pellets (PC pellets) to sample <sup>222</sup>Rn from the environment and scintillation counting by liquid scintillation spectrometer to determine <sup>222</sup>Rn activity. The technique is intended to provide time and cost efficient <sup>222</sup>Rn measurements for the purpose of *a posteriori* calibration of CDs. We also present results from pilot applications of the proposed technique to radon-in-soil

gas measurements, which imply that the technique may be applicable when large screening campaigns for express radon in soil-gas measurements are necessary. In the next sections the theoretical basis of this type of measurement is outlined and the methods' applicability to *a posteriori* calibration of CDs is demonstrated and possible applications to soil-gas<sup>222</sup>Rn measurements are discussed.

#### 2. Methods and materials

#### 2.1. Radon sampling by absorption in polycarbonates

When polycarbonate specimens are used to sample <sup>222</sup>Rn from the environment it is useful to define sampling efficiency  $\varepsilon_s$  (Mitev et al., 2014b):

$$\varepsilon_s = \frac{A_{PC}}{A_{env}},\tag{1}$$

where  $A_{PC}$  is the <sup>222</sup>Rn activity absorbed in the polycarbonate and  $A_{env}$  is the ambient <sup>222</sup>Rn activity contained in a volume, equal to that of the polycarbonate sample:  $A_{env} = VA_V$ , with  $A_V$  being the ambient radon activity concentration. The sampling efficiency is a measure of the sampler capabilities. It can be determined from the solution of the diffusion equation taking into account the radioactive decay (see Pressyanov et al. (2009) for details). The two most common cases are exposure to constant ambient <sup>222</sup>Rn concentration  $A_V(t) = A_V = \text{const}$  (Mode A exposure) and exposure to concentrations that decrease due to radioactive decay  $A_V(t) = A_V e^{-\lambda t}$  (Mode B exposure with  $\lambda$  being the <sup>222</sup>Rn decay constant). The corresponding sampling efficiencies for PC pellets with the shape of a circular cylinder of radius *R* and height *H* are:

• Sampling efficiency in Mode A exposure:

$$\varepsilon_{s}(t_{s}, t_{d}) = 32\lambda K \sum_{k=0}^{\infty} \sum_{m=1}^{\infty} \left( \left( \frac{L_{D}}{H\nu_{m}} \right)^{2} + \left( \frac{L_{D}}{\pi R(2k+1)} \right)^{2} \right) \frac{\left( 1 - e^{-\lambda_{2k+1,m}t_{s}} \right)}{\lambda_{2k+1,m}} e^{-\lambda_{2k+1,m}t_{d}}$$
(2)

• Sampling efficiency in Mode B exposure:

$$\varepsilon_{s}(t_{s}, t_{d}) = 32\lambda K \sum_{k=0}^{\infty} \sum_{m=1}^{\infty} \left( \left( \frac{L_{D}}{H\nu_{m}} \right)^{2} + \left( \frac{L_{D}}{\pi R(2k+1)} \right)^{2} \right) \frac{\left( e^{-\lambda t_{s}} - e^{-\lambda_{2k+1,m} t_{s}} \right)}{\lambda_{2k+1,m} - \lambda} e^{-\lambda_{2k+1,m} t_{d}}$$
(3)

In the equations above  $\nu_m$  is the m-th root of the Bessel function of 0-th order and  $\lambda_{j,m} = \lambda(1 + (\nu_m L_D/R)^2 + (j\pi L_D/H)^2)$ ;  $t_s$  is the sorption (exposure) time and  $t_d$  is the desorption time, where the desorption is considered in radon-free air. *K* and  $L_D$  are the partition coefficient and the diffusion length of radon in the polycarbonate material. Both *K* and  $L_D$  depend on the temperature (*T*), i.e. K = K(T)and  $L_D = L_D(T)$  (Pressyanov, 2009; Dimitrova et al., 2012).

For the purpose of the measurement of the absorbed activity one usually defines counting (i.e. detection) efficiency  $\varepsilon_c$  (Mitev et al., 2014b):

$$\varepsilon_c = \frac{n_0}{A_{PC}},\tag{4}$$

where  $n_0$  is the net counting rate of the instrument that is due to

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