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Application of scintillation counting using polycarbonates to radon measurements

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Radiation Measurements

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highlights are the control of

• An approach for ²²²Rn 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/m³) 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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This work proposes an approach for radon (^{222}Rn) measurement based on scintillation counting of polycarbonate specimens. The proposed technique takes advantage of the high absorption ability of polycarbonates to 222Rn and of the fact that radiation emitted by 222Rn 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 ²²²Rn measurements is demonstrated. The first application of the proposed technique is in the a posteriori calibration of compact discs for retrospective 222 Rn measurements, where it is sometimes necessary to measure ²²²Rn concentrations higher than 2 MBq/m³ for several days. It is demonstrated that the application of the proposed technique increases the range of applicable 222 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 222 Rn measurements. The applicability of the technique is demonstrated in more than 110 radon-in-soil-gas measurements, which were performed in different terrains, covering 222 Rn concentrations between 3 and 1200 kBq/m³. 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

The compact disc method (CD method) for retrospective mea-surements of ²²²Rn was proposed by [Pressyanov et al. \(1999, 2000,](#page--1-0) [2001\)](#page--1-0) and ever since has been applied intensively for measurements of 222Rn in human dwellings and working environment ([Pressyanov et al., 2012, 2010](#page--1-0)). The method relies on the fact that

the polycarbonate material, of which the compact discs (CDs) are made, absorbs ²²²Rn and is a solid state nuclear track detector. Thus, the track density at depths greater than 80 μ m beneath the CD surface is proportional to the integrated 222 Rn concentration to which the CD was exposed [\(Pressyanov et al., 2003, 2004;](#page--1-0) [Pressyanov, 2010](#page--1-0)). Currently the CD method seems to be the most accurate retrospective ²²²Rn measurement method ([Pressyanov](#page--1-0) [et al., 2012\)](#page--1-0). The accuracy of the method is mainly due to two reasons: first, the signal (track density) at depths greater than 80 μ m does not depend at all on the radon progeny concentrations Finally author.

Finally although stress is the $(K, M\text{lim})$ in the air and, second, the method allows determination of the

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individual sensitivity to 222 Rn of each CD. The latter is termed a posteriori calibration of the CDs and facilitates the establishment of \overline{a} traceability chain of the 222 Rn measurements by CDs to reference standards [\(Pressyanov et al., 2013](#page--1-0)). 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 222 Rn concentration to which the CD was exposed. Then, a second quarter of the same CD is cut and exposed in the laboratory to known, high 222 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 222 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 222 Rn concentrations, of the order of $1-2$ MBq/m³ or higher. However, such ²²²Rn concentrations are on (or beyond) the upper limit of the measurement range of reference ²²²Rn monitors. For example, the upper limit of the RAD7 222 Rn detector is 0.75 MBq/m³ ([Durridge, 2011](#page--1-0)) and that of the AlphaGUARD portable ²²²Rn monitor is 2 MBq/m³ ([Saphymo,](#page--1-0) [2014\)](#page--1-0). Thus, when higher than 2 MBq/m^{3 222}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 210Pb/210Po which cannot be compensated. Therefore, new techniques for high ²²²Rn concentration measurements are necessary to make the routine a posteriori calibration of CDs time and cost efficient.

Soil-gas radon (222 Rn) measurements, on the other hand, provide basic information for risk mapping [\(Kemski et al., 2006\)](#page--1-0). In conjunction with geological and soil investigations they can serve for classification and regionalization of the geologically induced risk for high indoor 222Rn concentrations [\(Kemski et al., 1996\)](#page--1-0). The approach to use soil-gas 222 Rn concentration for mapping of the geogenic ²²²Rn potential has been applied in some countries like Germany [\(Kemski et al., 2001\)](#page--1-0) and Hungary ([Szab](#page--1-0)ó [et al., 2014](#page--1-0)). In situ soil-gas ²²²Rn measurements also provide important information when ²²²Rn mitigation is to be performed (see, for example [Cosma et al. \(2015\)](#page--1-0)). Soil-gas 222Rn 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. \(2009\)](#page--1-0) and the references therein) or precursors of volcanic seismic activity ([Cigolini et al., 2013](#page--1-0)). An extensive review of the relationship between soil and spring gases (incl. 222 Rn) and both tectonic and seismic activities is published by [Toutain and Baubron \(1999\).](#page--1-0) 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 ²²²Rn in soil gas measurements in earthquake research is published by [Papastefanou \(2002\)](#page--1-0).

The objective of this work is to propose and demonstrate feasibility of a new technique for 222 Rn measurements. The technique uses polycarbonate pellets (PC pellets) to sample ²²²Rn from the environment and scintillation counting by liquid scintillation spectrometer to determine ²²²Rn activity. The technique is intended to provide time and cost efficient $22\overline{2}Rn$ 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 ²²²Rn measurements are discussed.

2. Methods and materials

2.1. Radon sampling by absorption in polycarbonates

When polycarbonate specimens are used to sample ²²²Rn from the environment it is useful to define sampling efficiency ε_{s} ([Mitev](#page--1-0) [et al., 2014b](#page--1-0)):

$$
\varepsilon_{\rm S} = \frac{A_{\rm PC}}{A_{\rm env}},\tag{1}
$$

where A_{PC} is the ²²²Rn activity absorbed in the polycarbonate and A_{env} is the ambient ²²²Rn 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. \(2009\)](#page--1-0) for details). The two most common cases are exposure to constant ambient ²²²Rn 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 λ being the ²²²Rn 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:

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

• Sampling efficiency in Mode B exposure:

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

In the equations above ν_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](#page--1-0)).

For the purpose of the measurement of the absorbed activity one usually defines counting (i.e. detection) efficiency ε_c ([Mitev](#page--1-0) [et al., 2014b](#page--1-0)):

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

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

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