

# A short-time method to measure the radon potential of porous materials

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## ABSTRACT

The radiological risk associated with the use of solid materials has been traditionally established according to their radon exhalation rates, the accumulation chamber technique being the most widely used for the determination of this quantity. However, this coupled methodology has two important drawbacks: the calculated exhalation rate value depends strongly on the experimental setup used; hence widely varying values can be calculated for the same material. Furthermore, this technique usually requires long monitoring times (between 1 and 4 weeks).

In this paper, we present a fast and reproducible method for the determination of radon potential (as an alternative to the exhalation rate) based on the application of the accumulation chamber technique. Radon potential is proportional to the emanation coefficient, and can be calculated within measuring times of less than 24 h. The theoretical basis is developed and the experimental setup is discussed in detail in this paper.

The procedures for the determination of different experimental parameters (leakage constant, slope correction) are shown as essential steps for the later determination of the radon potential. In addition, the robustness of the developed methodology is demonstrated, and the reproducibility tests carried out with the general system performance are shown. Finally, the radon potential for different materials is determined, allowing its prompt categorization according to its associated radiological risk.

## 1. Introduction

It is well known that radon and its short-lived decay products in both outdoor and indoor atmospheres are the most important contributors to human exposure to ionizing radiation from natural sources, especially the indoor atmosphere is sometimes troublesome due to possible radon accumulation in houses, buildings and workplaces. This contribution is around  $1.1 \text{ mSv y}^{-1}$ , which represents 50% of the total dose (UNSCEAR, 2000). This significant level of radon radiology implies the need to control it in the workplace and/or housing. To do that, it is important to evaluate the role and contribution of the different materials that can act as radon sources inside buildings for work and residence (soils, building materials, NORMs, wastes, etc.).

Several parameters, such as exhalation and air ventilation rates at the studied site and the occupation factor, determine the true radon radiological risk associated with such materials. As a consequence, a quantitative classification of these materials according to their true associated radiological risk is not possible. A common classification could be established on the basis of the

corresponding material exhalation rate. Nevertheless, the exhalation rate is a function of diffusion length, which, in turn, depends on several physical parameters of the material (humidity, porosity and geometry). Furthermore, the obtained value of this quantity is dependent on the methodology used for its characterization (Petropoulos et al., 2001; Tuccimei et al., 2006), i.e., it is not an absolute magnitude. An alternative approach to quantify the potential risk instead of the true radiological risk could be based on the so-called “radon potential”,  $\Omega$ , (1), which represents the radon activity generated inside the material that can be transported across it towards the surrounding atmosphere.

$$\Omega = \varepsilon C_{Ra} \text{ (Bq kg}^{-1}\text{)} \quad (1)$$

with  $\varepsilon$  being the emanation factor and  $C_{Ra}$  ( $\text{Bq kg}^{-1}$ ) the activity concentration of  $^{226}\text{Ra}$  of the material.

In contrary to the exhalation rate, the radon potential is a characteristic of the material that is not influenced by the experimental conditions during its determination. The radon potential appears, then, as a suitable magnitude for classifying/comparing porous materials in relation to its potential radon radiological risk.

Several authors have described the determination of the exhalation rate and emanation factor of materials using both laboratory and field measurements. Nevertheless, big differences

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have been found among their data, as well as in the experimental conditions employed (Baykara et al., 2001; Bossew, 2003; Singh et al., 1999; Mahur et al., 2008). These methods are generally based on accumulation techniques, and they present the additional problem of requiring accumulation periods within a range of 1–4 weeks (Sakoda et al., 2008). Furthermore, the presence of leakages within the chamber modifies the effective time constant and the saturation concentration reached. As a consequence, the exhalation rate and emanation coefficient could be underestimated (Ferry et al., 2002). Another typical problem of this technique is the bound exhalation: when the radon concentration within the chamber air increases, the concentration gradient driving the diffusion decreases. This provokes a redistribution of the radon profile leading to a decrease in the determined exhalation rate value (Cosma et al., 2001).

The major aim of this work is the improvement of the accumulation technique in order to get a fast, accurate and reproducible method for the measurement of radon potential. It will be shown that the same methodology permits the determination of radon potential, once the experimental setup is provided and the corresponding mathematical modeling has taken into account the effects described above.

## 2. Theoretical framework review

The study of the accumulation of radon exhaled by a sample into a sealed chamber can be divided into two interdependent processes: (1) transport within the sample and (2) accumulation in the chamber after crossing the interface. The continuity condition of the radon concentration in the sample–air interface is the term that relates both processes of the problem. This coupled equations' system can be solved in an analytical way after introducing several approximations.

### 2.1. Transport within the sample

The generation and transport processes within porous materials are described in a general way by an equation of balance for each phase in the sample (soil, water and interstitial air). The complexity of the problem can be reduced by transforming these equations into another for a simple phase (air), using expanded definitions of transport coefficients and porosity (Rogers and Nielson, 1991).

In order to resolve this equation analytically, it is necessary to assume the following approximations:

- One-dimension transport across z-axis.
- Diffusion coefficient is homogeneous in the sample.
- $^{226}\text{Ra}$  concentration is homogeneously distributed in the material.
- Diffusive transport only (the pressure is constant within the chamber).
- Exhalation only occurs in the interface sample–air ( $z = 0$ ).

Under these conditions, the transport equation can be written in the following way:

$$\frac{\partial C_{Rn}}{\partial t} = D_e \frac{\partial^2 C_{Rn}}{\partial z^2} + \lambda_{Rn} \Phi_0 - \lambda_{Rn} C_{Rn} \quad (2)$$

With b.c.

$$C_{Rn}(z = 0) = C_{Rn}^a(t) \quad (3,4)$$

$$\left. \frac{\partial C_{Rn}}{\partial z} \right|_{z=-z_0} = 0$$

where  $C_{Rn}$  is the radon concentration in the air-filled pores ( $\text{Bq m}^{-3}$ ),  $C_{Rn}^a$  is the radon concentration in the air of the chamber ( $\text{Bq m}^{-3}$ ),  $\lambda_{Rn}$  is the radon decay constant ( $\text{s}^{-1}$ ) and  $D_e$  is the effective diffusion coefficient ( $\text{m}^2 \text{s}^{-1}$ ), which is written as  $D_e = D_b/\beta$ .

$D_b$  is the bulk diffusion coefficient ( $\text{m}^2 \text{s}^{-1}$ ) and  $\beta = (1-s+sL)p + \rho k_a$  is the effective porosity,  $s$  is the humidity saturation (fraction of water-filled pores),  $L$  is the Ostwald coefficient,  $p$  is the porosity,  $\rho$  is the bulk density ( $\text{kg m}^{-3}$ ) and  $k_a$  is the adsorption coefficient to solid phase (generally depreciable).  $\Phi_0$  is related to the source term of the material ( $\text{Bq m}^{-3}$ ):

$$\Phi_0 = \frac{\rho}{\beta} C_{Ra} \varepsilon = \frac{\rho}{\beta} \Omega \quad (5)$$

$\varepsilon$  is the emanation fraction and  $C_{Ra}$  is the  $^{226}\text{Ra}$  activity concentration ( $\text{Bq kg}^{-1}$ ). The term  $\Omega$  (Eq. (1)) is the previously defined *Radon potential*.

Eq. (3) is the continuity condition and it provides the temporal dependence of the concentration profile in the sample as a function of the Rn concentration inside the chamber. To solve this equation it is necessary to consider that the evolution of the concentration profile within the sample from the initial steady state to final steady state is a quasi-static process. This way, the solution will be a succession of steady states modulated by this boundary condition (Cosma et al., 2001).

In these conditions, the concentration profile in the sample is written in this form (Porstendöfer, 1994):

$$C_{Rn}(z, t) = (C_{Rn}^a(t) - \Phi_0) \text{tgh} \left( \frac{z_0}{l_0} \right) \text{senh} \left( \frac{z_0}{l_0} \right) + (C_{Rn}^a(t) - \Phi_0) \cosh \left( \frac{z_0}{l_0} \right) + \Phi_0 \quad (6)$$

$$l_0 = \sqrt{\frac{D_e}{\lambda_{Rn}}} \quad (7)$$

with  $z_0$  being the sample thickness (m) and  $l_0$  the diffusion length (m).

The sample exhalation rate,  $E$ , is defined as the radon activity flow crossing the interface sample–air ( $\text{Bq m}^{-2} \text{s}^{-1}$ ) and can be calculated by means of Fick's first law as  $z = 0$ :

$$E = -D_b \left. \frac{\partial C_{Rn}}{\partial z} \right|_{z=0} = \frac{D_b}{l_0} (\Phi_0 - C_{Rn}^a) \text{tgh} \left( \frac{z_0}{l_0} \right) = E_0 - \omega C_{Rn}^a(t) \quad (8)$$

$$\omega = \frac{D_b}{l_0} \text{tgh} \left( \frac{z_0}{l_0} \right) = \beta \lambda_{Rn} l_0 \text{tgh} \left( \frac{z_0}{l_0} \right) \quad (9)$$

$$E_0 = \omega \Phi_0 = \rho \lambda_{Rn} l_0 \text{tgh} \left( \frac{z_0}{l_0} \right) \Omega \quad (10)$$

The exhalation rate depends on the radon in the chamber air (Eq. (8)). This situation leads to a decrease in the exhalation rate as the radon concentration increases. This effect is known as bound exhalation, and is quantified by the term  $\omega C_{Rn}^a$  in Eq. (8). It is possible to consider the extreme case where the sample thickness is very small against diffusion length,  $z_0 \ll l_0$ . In this situation, the free exhalation,  $E_0$ , is not dependent on the diffusion length, but only on sample characteristics:

$$\text{tgh} \left( \frac{z_0}{l_0} \right) \xrightarrow{z_0 \ll l_0} \frac{z_0}{l_0} \quad (11)$$

$$E_0 = \lambda_{Rn} \varepsilon C_{Ra} \rho z_0 = \lambda_{Rn} \Omega \frac{m}{S} \quad (12)$$

$S$  is the exhalation surface ( $\text{m}^2$ ) and  $m$  is the sample mass (kg), and plane geometry for the source has been assumed (i.e.,  $V = Sz_0$ ). Typically, the diffusion length in porous material is above the 0.25–0.30-m range (Keller et al., 2001; Mujahid et al., 2005);

therefore, this approximation will be applicable to a sample thickness of less than 0.05 m.

On the other hand, if the sample thickness is great against the diffusion length, then  $z_0 \gg l_0$ . Now, the free exhalation rate is a function of the diffusion length. This is the typical case of semi-infinite material:

$$\operatorname{tgh}\left(\frac{z_0}{l_0}\right) \xrightarrow{z_0 \gg l_0} 1 \quad (13)$$

$$E_0 = \lambda_{Rn} \varepsilon C_{Ra} \rho l_0 \quad (14)$$

## 2.2. Accumulation inside the chamber

The Rn accumulation process inside the chamber is governed by a balance equation (Butterweck-Dempewolf and Schuler, 1996), with the source term as an implicit time function, is shown in Eq. (8):

$$\frac{dC_{Rn}^a}{dt} = \frac{E(t)S}{V_c} - \lambda_{Rn} C_{Rn}^a - \lambda_v C_{Rn}^a \quad (15)$$

$$C_{Rn}^a(t=0) = C_{Rn}^0 \quad (16)$$

where  $V_c$  is the effective chamber volume available for accumulating radon ( $\text{m}^3$ ) and  $\lambda_v$  is the leakage constant of the experimental setup ( $\text{s}^{-1}$ ).

After introducing Eqs. (8) in (15) for  $z_0 \ll l_0$ , the solution can be written as

$$C_{Rn}^a(t) = C_{Rn}^{sat} + (C_{Rn}^0 - C_{Rn}^{sat})e^{-\lambda_{eff}t} \quad (17)$$

$$\lambda_{eff} = \lambda_{Rn} + \lambda_b + \lambda_v \quad (18)$$

$$\lambda_b = \lambda_{Rn} \beta \frac{V_0}{V_c} \quad (19)$$

$$C_{Rn}^{sat} = \frac{E_0 S}{\lambda_{eff} V_c} \quad (20)$$

with  $V_0$  being the sample volume ( $\text{m}^3$ ),  $\lambda_{eff}$  the effective time constant ( $\text{s}^{-1}$ ),  $\lambda_b$  the bound exhalation constant ( $\text{s}^{-1}$ ),  $C_{Rn}^{sat}$  the final steady-state concentration ( $\text{Bq m}^{-3}$ ) and  $C_{Rn}^0$  the initial background concentration ( $\text{Bq m}^{-3}$ ).

The solution (Eq. (17)) represents an exponential growth of the radon concentration inside the chamber with an effective time constant, which depends on the leakages of the chamber and the sample characteristics (bound exhalation constant). The ratio of the sample volume to chamber volume and the sample effective porosity determine the magnitude of the effect of bound exhalation on the measure. The leakage constants are generally the most important parameters to be determined in these experiments. Indeed, the combined contributions of all sink terms can be described through the determination of  $\lambda_{eff}$ .

The experimental determination of  $\lambda_{eff}$  and  $C_{Rn}^{sat}$  requires fitting experimental values of  $C_{Rn}^a$  as a function of time under continuous monitoring conditions. Obtaining the radon potential is immediate with the combination of Eqs. (12) and (20):

$$\Omega = \frac{\lambda_{eff} V_c C_{Rn}^{sat}}{\lambda_{Rn} m} \quad (21)$$

Since the determination of  $\Omega$  is neither dependent on other parameters nor other methods, it could be said that this method (continuum monitoring until reaching steady-state conditions) is an absolute method for measuring the radon potential in materials.

## 3. Short-time accumulation periods

As previously explained, the determination of different parameters such as the exhalation rate or emanation factor is time-consuming, requiring between 1 and 4 weeks of continuous monitoring for their determination. In order to reduce the typical accumulation period, a linear approximation can be made during the first hours of accumulation (Stranden, 1983):

$$C_{Rn}^a(t) \cong C_{Rn}^0 + (C_{Rn}^{sat} - C_{Rn}^0) \lambda_{eff} t \quad (22)$$

The slope of this equation,  $b$  ( $\text{Bq m}^{-3} \text{s}^{-1}$ ), is related to the exhalation of the sample, and this in turn is related to the radon potential:

$$\Omega = \frac{b + \lambda_{eff} C_{Rn}^0 V_c}{\lambda_{Rn} m} \quad (23)$$

As the time used for the determination of the slope increases, the deviation between the fitted slope,  $b$ , and the initial slope,  $b_0$ , increases. This deviation is a systematic consequence of approximating the exponential growth to a linear law. The slope that truly shows the radon potential is the initial slope (i.e., where the short measurement time approach can be used); thus, it is necessary to correct the fitted slope,  $b_{fitted}$ , by means of a slope correction factor (SCF):

$$SCF = \frac{b_0}{b_{fitted}} \quad (24)$$

A good approximation to the fitted slope for an accumulation period,  $t_0$ , could be the average value of all slopes with measurement times of less than  $t_0$ . Thus,

$$b_{fitted} = \frac{1}{t_0} \int_0^{t_0} m(t) dt = \frac{1}{t_0} \int_0^{t_0} \frac{dC_{Rn}^a}{dt} dt = \frac{a}{\lambda_{eff} t_0} (1 - e^{-\lambda_{eff} t_0}) \quad (25)$$

$$a = \lambda_{eff} (C_{Rn}^{sat} - C_{Rn}^0) \quad (26)$$

Eq. (25) is not defined for  $t=0$ , nevertheless, it is possible to evaluate its limit, which corresponds to the initial slope:

$$\lim_{t_0 \rightarrow 0} (b_{fitted}) = a \quad (27)$$

Therefore, the SCF is written in the following form:

$$SCF = \frac{b_0}{b_{fitted}} = \frac{\lambda_{eff} t_0}{1 - e^{-\lambda_{eff} t_0}} \quad (28)$$

As expected, this factor only depends on the measuring time used for fitting and the effective time constant of the measure.

Using this approach requires knowing the effective time constant in the chamber, which is a combination of the sample ( $\lambda_b$ ) and chamber ( $\lambda_v$ ) characteristics. The bound exhalation constant can be determined by using Eq. (19). Nevertheless, obtaining the leakages constant requires a special test, which can be made in two ways: (1) natural decay of an initial radon concentration (López-Coto et al., 2007), and (2) accumulating and monitoring the radon exhaled by a sample until steady-state conditions have been reached. The second way introduces a greater uncertainty than the first due to the combined effect of the parameters involved in the determination.

## 4. Detection limit. Minimum detectable radon potential ( $MD\Omega$ )

In samples with low radon potential, it is possible that the radon concentration generated by the sample can be masked by the typical background fluctuations and, so, the result obtained for the sample would be overestimated. In the literature, they usually refer to the detection limit of the radon detector.

With the aim of establishing a lower limit for this method, it is necessary to define the minimum detectable radon potential ( $MD\Omega$ ) concept as “the minimum radon potential that produces a net increase in the radon concentration significantly distinguishable from the background”. This parameter is simply the application of the ISO’s decision limit (DL; ISO 11929-5; 2006) to our working conditions.

This idea is associated with the concept of minimum detectable concentration (MDC), which can be defined as radon concentration that differs significantly from the background (i.e., the radon concentration corresponding to the lower limit of detection (LLD) of the whole operational setup). This way, it is possible to write the MDC as

$$MDC = C_{Rn}^0 + 3\sigma_{background} \quad (29)$$

where  $\sigma_{background}$  is the standard deviation of the background ( $Bq\ m^{-3}$ ). This definition ensures that the MDC is different to the typical background fluctuations within a 99% confidence interval.

If the averaged background concentration is less than the detection limit of the radon detector, it is necessary to use this last value in order to determine the MDC.

With these definitions it is possible to evaluate the  $MD\Omega$  for this method in its two versions: long-time and short-time periods.

#### 4.1. Long-time periods

This is the classic case of accumulation as far as the steady-state condition (Eq. (16)). In this case, the saturation concentration must be at least the MDC in order to differentiate it from background fluctuations, thus

$$MD\Omega|_{Long-time} = \frac{\lambda_{eff} V_c MDC}{\lambda_{Rn} m} \quad (30)$$

The value of this limit increases along with the expansion of the chamber volume, the leakages constant, the bound exhalation constant, the initial background concentration and the background fluctuations, and it decreases as the mass amount increases.

#### 4.2. Short-time periods

For short-time periods (Eq. (22)), the net change in the radon concentration during the accumulation period,  $t_0$ , must be at least  $MDC - C_{Rn}^0 = 3\sigma_{background}$ ; thus the minimum detectable slope must be  $3\sigma_{background}/t_0$ . Hence, the  $MD\Omega$  can be written in the following form:

$$MD\Omega|_{Short-time} = \frac{(3\sigma_{background}/t_0) + \lambda_{eff} C_{Rn}^0 V_c}{\lambda_{Rn} m} \quad (31)$$

This expression follows, qualitatively, the same dependence on the chamber and sample characteristics as the expression found for long-time periods. Nevertheless, the background fluctuations are of greater importance than in long-term periods. This way, it is possible to ensure that the detection limit for short-time periods is higher than those corresponding to long-time periods, as expected for conventional radiometric detectors.

### 5. Experimental setups

In order to perform the experimental measures involved in this work, three radon accumulation chambers were developed, each with different nominal volumes. The materials used in the construction of the chambers have been selected according to

their diffusion performances in order to ensure maximum impermeability of the walls.

For radon concentration monitoring purposes, two radon detectors have been used. The AlphaGUARD PQ2000PRO of Genitron Instruments (AG) is an ionization chamber with a working range of  $2-2 \times 10^6 Bq\ m^{-3}$  and a temporal resolution of 10 min. This detector had been factory-calibrated and is traceable to PTB and NIST. The other detector is the Model 1027 of Sun Nuclear Corporation (SNC). This detector is based on the radon progeny electrodeposition with a working range of  $4-4 \times 10^3 Bq\ m^{-3}$  and a temporal resolution of 1 h. It had been calibrated at the Huelva University Radon Chamber, the reference detector being the NIST-traceable AG, and a calibration factor very close to the ideal value obtained (López-Coto et al., 2007).

The A chamber consists of a hollow aluminium cylinder with a nominal volume of 0.23 L, which is screwed onto the radon diffusion entry window of the AlphaGUARD. A rubber gasket seals the two parts of the setup (Ferry et al., 2002). The effective volume for accumulation is formed by the inner volume of the detector (0.59 L) and the rest of the cylinder not occupied by the sample.

The B and C chambers had been made with a high-density polyethylene (HDPE) barrel of 60 and 220 L of nominal volume, respectively, and are sealed with a rubber gasket and a mechanical pressure closing. Although these chambers can be operated with both detectors, the B chamber (60 L) was operated with the SNC and the C chamber (220 L) with the AG in order to carry out simultaneous measurements and save time.

The sample has cylindrical geometry with different diameters, and the sample thickness is always less than 5 cm. The walls of the sample container ensure that radon exhalation from the materials only appears on their upper surface.

Humidity saturation,  $s$ , and porosity,  $p$ , have been obtained via the determination of the grain density,  $\rho_g$  ( $kg\ m^{-3}$ ), the bulk density,  $\rho_b$  ( $kg\ m^{-3}$ ) and the amount of water,  $w$  (kg), contained within a volume,  $V$  ( $m^3$ ), of the sample:

$$p = 1 - \frac{\rho_b}{\rho_g} \quad (32)$$

$$s = \frac{w}{pV\rho_w} \quad (33)$$

where  $\rho_w$  ( $kg\ m^{-3}$ ) is the water density. Using these parameters, the diffusion length can be calculated using the experimental expression proposed by Rogers and Nielson (1991). Hence, it has been possible to test that all the diffusion lengths of the material are substantially longer than the sample thicknesses. In these conditions, it is possible to ensure that the operational requirements shown in Section 2.1 are satisfied.

Although it is not necessary for radon potential determination, the  $^{226}Ra$  activity of the samples has been measured using a Canberra GX3519 gamma spectrometer (38% relative efficiency) including a 15-cm Fe passive shielding (Pérez-Moreno et al., 2002). This value, coupled to the radon potential, allows the calculation of the emanation factor of the samples (1).

### 6. Results

#### 6.1. Leakages constant

This parameter, which is necessary for the determination of the effective time constant of the chamber, has been determined by monitoring the temporal evolution of radon concentration inside the chamber, following the natural decay of an initial radon concentration. Fitting the experimental data to Eq. (17), without

source term, it is possible to calculate the leakage constant of the chamber (López-Coto et al., 2007).

In order to test the reproducibility of this parameter, 5–10 replicates for each chamber have been performed. Both averages and standard deviations are shown (Table 1).

As expected, the leakage constant decreases with increasing volume of the chamber. However, it is possible to see by comparing the standard deviation that the screwed closing system is less reproducible than the pressure-based closing system.

## 6.2. Slope correction factor

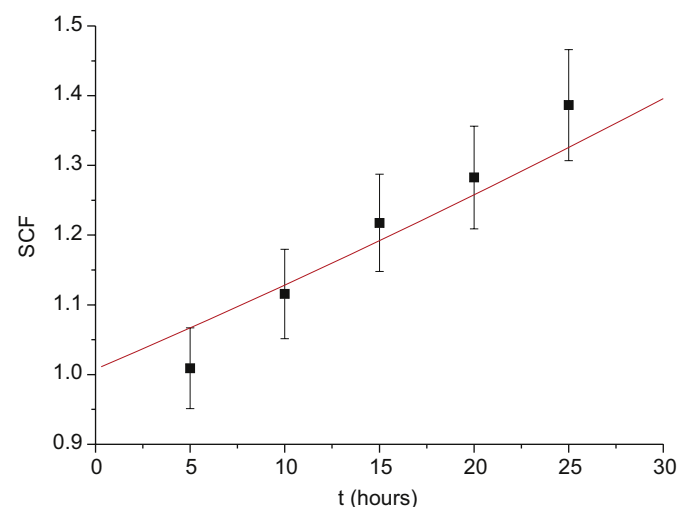
In order to correct the deviation between the linear law and the exponential growth, it is necessary to introduce the SCF as previously explained. The robustness of the theoretical model (28) has been tested and the experimental slope correction determined. To do that, different accumulation times have been used and tested against the long-time measurement method through the analysis of a phosphate rock sample and the A chamber. The results are shown in Fig. 1. It is possible to see the good agreement between the experimental and theoretical values. This experience allows the use of this theoretical factor in order to correct the fitted slope to a selected accumulation time.

## 6.3. Free exhalation rate vs sample thickness

According to the theoretical model, a linear relationship can be established between the free exhalation rate and the sample thickness. Hence, an experiment was carried out in the C chamber using a phosphate rock sample with  $C_{Ra} = 1600 \pm 20 \text{ Bq kg}^{-1}$ . Fig. 2 shows the normalized free exhalation rate for several sample thicknesses. This normalized exhalation rate,  $\hat{E}$  (m), has

**Table 1**  
Leakages constants of the chambers with the standard deviation for 5–10 replicates

Chamber	$\lambda_v \text{ (s}^{-1}\text{)}$
A	$(4.5 \pm 0.7) \times 10^{-6}$
B	$(3.70 \pm 0.19) \times 10^{-7}$
C	$(2.81 \pm 0.14) \times 10^{-7}$



**Fig. 1.** Theoretical and experimental slope correction factor.

been calculated from Eq. (12) in order to obtain the emanation factor as the slope of the fitted function:

$$\hat{E} = \frac{E_0}{C_{Ra} \rho_b \lambda_{Rn}} = \varepsilon z_0 \quad (34)$$

The aforementioned dependence is in good agreement with the prediction as it is possible to deduce through the correlation factor ( $r^2 = 0.9801$ ) obtained. The emanation factor obtained this way,  $\varepsilon = 0.14 \pm 0.01$ , is in good agreement with the literature and the radon potential is  $\Omega = 224 \pm 22 \text{ Bq kg}^{-1}$ . The independent term has no statistical significance, showing that there is no systematic offset, as expected.

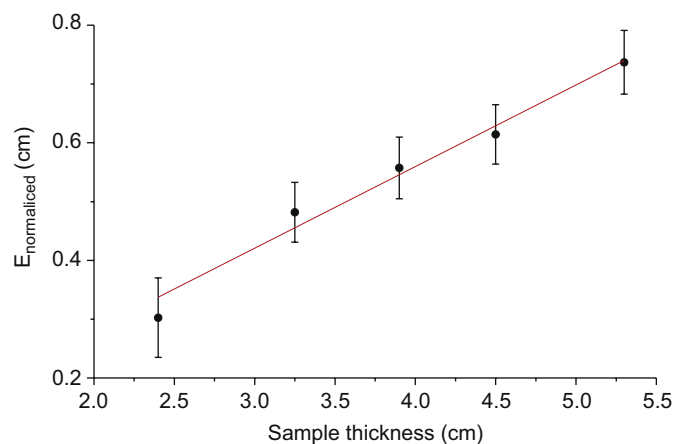
## 6.4. Reproducibility test

A phosphate rock sample has been measured 10 times in each chamber (Table 2). It can be seen that all the results agree within the statistical dispersion. Indeed, those values are in good agreement with the radon potential obtained for this sample in Section 6.2. This shows that this methodology can be reproduced independently in the chamber used, and that small-volume chambers give more precise results.

## 6.5. Radon potential of different materials

To prove the usefulness of this method, 64 samples of varied origins have been measured. Table 3 shows the results obtained for the radon potential, radium concentration, emanation factor and  $MDS\Omega$  with their standard deviations, which indicates the statistical dispersion of each sample.

The materials arising from the uranium mining industry present a radon potential 5 times higher than those generated in the production of phosphate fertilizers, which in turn have values 30 times higher than typical soils. In contrast, the building materials from this zone have a radon potential similar to these soils. That could allow us to establish an exposure risk classifica-



**Fig. 2.** Variation of the exhalation factor with sample thickness.

**Table 2**  
Radon potential and standard deviation determined for 10 replicates of a phosphate rock sample

Chamber	$\Omega \text{ (Bq kg}^{-1}\text{)}$
A	$227 \pm 9$
B	$215 \pm 13$
C	$222 \pm 15$

**Table 3**  
Radon potential, radium concentration, emanation factor and minimum detectable radon potential with their standard deviations for different kinds of materials

Material	$\Omega$ (Bq kg <sup>-1</sup> )	$C_{Ra}$ (Bq kg <sup>-1</sup> )	$\varepsilon$ (%)	$MD\Omega$ (Bq kg <sup>-1</sup> )
Huelva soils (10)	13 ± 1	32 ± 20	37 ± 11	3.0
Canary Island soils (4)	12 ± 3	42 ± 16	29 ± 10	2.5
Phosphogypsum stacks cover (9)	6 ± 3	22 ± 8	27 ± 7	2.9
Phosphogypsum (15)	127 ± 29	656 ± 149	20 ± 4	8.9
Phosphorite (13)	227 ± 9	1600 ± 40	14 ± 1	9.7
Uranium mining wastes (3)	552 ± 404	5500 ± 3600	10 ± 2	9.2
Building materials (10)	2.4 ± 1.3	<i>N. M.</i>	<i>N. M.</i>	2.1

The number in brackets represents the analysed samples for each kind of material.

tion: material from uranium mining industry > phosphate rock ≫ typical soils ~ building materials.

These results show that a wide range of materials can be measured by the method proposed; only materials with very low radon potential being problematic from the methodological point of view.

It is worth noting that, according to Eq. (12), the determination of radon diffusion lengths has been not required to obtain a categorization of the materials studied. If necessary, the exhalation rate could be determined using Eq. (10), although it would require the inclusion of both diffusion length and the problem geometry (as shown in the calculations). It can be seen that the exhalation rate for the same kind of material (i.e., same radon potential) might vary along several orders of magnitude as a function of these factors.

## 7. Conclusions

A general model describing the radon concentration evolution inside an accumulation chamber has been developed in order to design a fast and reproducible method for the determination of radon potential. This quantity can be considered an interesting alternative to the exhalation rate under certain conditions in order to compare the potential radon exposure radiological risk due to materials. The typical accumulation period has been reduced from several weeks to a few hours, saving a lot of measurement time. A detection limit has been defined as a quality indicator. According to the data obtained, it is possible to ensure that a wide range of materials can be measured with this technique without the need to deal with additional parameters and considerations such as diffusion length and the geometry of the problem, which can introduce differences of several orders of magnitude in the calculated exhalation rate.

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## References

- Baykara, O., Dogru, M., Inceoz, M., Aksoy, E., 2001. Measurements of radon emanation from soil samples in triple-junction of North and East Anatolian active faults systems in Turkey. *Radiat. Meas.* 39, 209.
- Bossew, P., 2003. The radon emanation power of building materials, soils and rocks. *Appl. Radiat. Isot.* 59, 389–392.
- Butterweck-Dempewolf, G., Schuler, Ch., 1996. An extended radon chamber model. *Environ. Int.* 22, 891–898.
- Cosma, C., Dance, F., Jurcut, T., Ristoiu, D., 2001. Determination of <sup>222</sup>Rn emanation fraction and diffusion coefficient in concrete using accumulation chambers and the influence of humidity and radium distribution. *Appl. Radiat. Isot.* 54, 467–473.
- Ferry, C., Richon, P., Beneito, A., Cabrera, J., Sabroux, J.C., 2002. An experimental method for measuring the radon-222 emanation factor in rocks. *Radiat. Meas.* 35, 579–583.
- International Standard ISO 11929-5, 2006. International Organization for Standardization, Geneva, Switzerland.
- Keller, G., Hoffmann, B., Feigenspan, T., 2001. Radon permeability and radon exhalation of building materials. *Sci. Total Environ.* 272, 85–89.
- López-Coto, I., Bolivar, J.P., Más, J.L., García-Tenorio, R., Vargas, A., 2007. Development and operational performance of a single calibration chamber for radon detectors. *Nucl. Instrum. Methods Phys. Res. A* 579, 1135–1140.
- Mahur, A.K., Kumar, R., Mishra, M., Sengupta, D., Rajendra, P., 2008. An investigation of radon exhalation rate and estimation of radiation doses in coal and fly ash samples. *Appl. Radiat. Isot.* 66, 401–406.
- Mujahid, S.A., Hussain, S., Dogar, A.H., Karim, S., 2005. Determination of porosity of different materials by radon diffusion. *Radiat. Meas.* 40, 106–109.
- Pérez-Moreno, J.P., San Miguel, E.G., Bolivar, J.P., Aguado, J.L., 2002. A comprehensive calibration method of Ge detectors for low-level spectrometry measurements. *Nucl. Instrum. Methods Phys. Res.* 491, 152–162.
- Petropoulos, N.P., Anagnostakis, M.J., Simopoulos, S.P., 2001. Building materials radon exhalation rate: ERRICCA intercomparison exercise results. *Sci. Total Environ.* 272, 109–118.
- Porstendöfer, J., 1994. Properties and behaviour of radon and thoron and their decay products in the air. *J. Aerosol Sci.* 25, 219–263.
- Rogers, V.C., Nielson, K.K., 1991. Multiphase radon generation and transport in porous media. *Health Phys.* 60, 807–815.
- Sakoda, A., Hanamoto, K., Ishimori, Y., Nagamatsu, T., Yamaokaa, K., 2008. Radioactivity and radon emanation fraction of the granites sampled at Misasa and Badgastein. *Appl. Radiat. Isot.*
- Singh, A.K., Sengupta, D., Ragendra, P., 1999. Radon exhalation rate and uranium estimation in rock samples from Bihar uranium and copper mines using the SSNTD technique. *Appl. Radiat. Isot.* 51, 107–113.
- Stranden, E., 1983. Assessment of the radiological impact of using fly ash in cement. *Health Phys.* 44, 145–153.
- Tuccimei, P., Moroni, M., Norcia, D., 2006. Simultaneous determination of <sup>222</sup>Rn and <sup>220</sup>Rn exhalation rates from building materials used in Central Italy with accumulation chambers and a continuous solid state alpha detector: influence of particle size, humidity and precursors concentration. *Appl. Radiat. Isot.* 64, 254–263.
- UNSCEAR, 2000. United Nations Scientific Committee on the Effects of Atomic Radiation. Sources, Effects and Risks of Ionizing Radiation.