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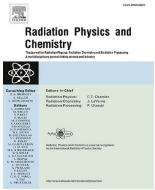

Comparative analysis of techniques for estimating radon exhalation from building materials

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ABSTRACT

Evaluating the radon emissions from building structures to determine optimal strategies for either preventing radon infiltration into buildings or decreasing its concentration in occupied areas is essential. This research presents a study of different methodologies for determining the radon exhalation rate in three samples of granite: two techniques based on passive detectors (Electret Ion Chamber (EIC) and Solid State Nuclear Track Detectors (SSNTD)) and one technique based on the use of a continuous radon detector. The operating principle of the three methodologies is based on the accumulation of radon inside an experimental container, hermetically sealed. Passive detectors offer the concentration measurement, integrated over time, while the continuous detector monitors the increase in concentration hour by hour. In this last case, three mathematical models have been applied to adjust the experimental results.

According to a previous radiological characterization of the building material samples, those exceeding the legal limit of the radioactive activity index (I = 1) have been analyzed: Rose Porriño granite, Blue Vizag granite and Julia Cream granite. The obtained results indicate that the five methodologies used present radon exhalation rate values within a consistent range for each sample. Therefore, the protocols detailed in this work could be used interchangeably to calculate the radon gas exhalation rates in building materials, providing flexibility in selecting the most suitable method depending specific experimental requirements or constraints.

1. Introduction

Radon is a radioactive gas that occurs naturally from the decay of uranium. This gas tends to accumulate in closed spaces such as homes and buildings. The greatest hazard of its inhalation is that high exposure has been associated with an increased risk of developing lung cancer according to the World Health Organization (WHO, 2009). Despite the associated risks, there is a lack of awareness and regulations regarding the measurement and mitigation of this gas.

It is naturally present in soils, depending on the composition and type of rock; in water, when gas dissolves in underground streams, lakes or rivers or in building materials whose raw materials have uranium and radium traces. Recent studies demonstrate the importance of analyzing the contribution to the increase in radon concentration inside homes due to some construction materials as granites, which are widely used due to its robustness. The exhalation of radon from construction materials is a result of their containing traces of uranium, specifically ^{238}U, which undergoes a decay process leading to the formation of radon. The higher radon concentration in certain materials is attributed to the presence of radium (^{226}Ra) according to uranium (^{238}U) decay chain. Overall, it is estimated that construction materials may contribute up to 20% of the total radon concentration in households (Chinchón-Payá et al., 2011). Granitic rocks are those with the highest concentration of ^{238}U; therefore, presenting higher concentration of ^{226}Ra and ^{222}Rn.

Building materials are the major sources of radon in newly built and highly air-tightened apartments, owing to the energetic efficiency and lack of ventilation. Its exhalation remains constant over time; therefore, it is crucial to identify and manage radon exhalation levels from building materials during the design stage. Doses received from construction materials are attributed to both external exposures, from gamma radiation emitted by elements present in the construction materials and internal exposure, related to the inhalation of radon gas and its decay products exhaled from these materials.

The use of natural materials and byproducts of industrial processes containing some of the mentioned radionuclides is common in construction. Therefore, it becomes necessary to characterize the most used construction building materials. Furthermore, measuring radon gas exhalation in the final product is essential. The activity concentration

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index and radium contents can be used to manage the exposure to gamma radiation originating from radionuclides in building materials. However, as the radon exhalation of building materials is affected by final product state (varnishes, paints, and other treatments applied to the material) it is difficult to directly relate radium activity with radon exhalation. Therefore, different countries are conducting research and measurements to verify the radon exhalation of building materials. Finland, Germany or Czech Republic or Spain (Federal Ministry for the Environment, N.C, 2018; Decree 422/2016 Prague, 2016; Guide ST12.2, 2010) use the radioactive activity index to determine the radiological risk from the ^{226}Ra, ^{232}Th and ^{40}K activities (Directive, 2013/59/EURATOM). Additionally, the US Environmental Protection Agency (USEPA) propose for concrete, new management standards based on the radium content and the radon exhalation rate of the concrete (K.K. Nelson et all, 1996).

The European Union underscores the significance of monitoring construction materials to assess the doses received (internally or externally) by individuals due to their exposure to the radioactive content of these materials. The primary building materials include concrete, bricks, natural stones, gypsum, and phosphogypsum (EUROPEAN COMMISSION, 1999). Moreover, various studies demonstrate that granite is the material that exudes the most radon, followed by stone, sand, and marble (Youssef et al., 2015). Hence, it is crucial to have an extensive database regarding the exhalation of various construction materials.

The ISO 11665-9, *Test methods for exhalation rate of building materials* (ISO 11665–9:2019), determines two methodologies for the determination of the radon exhalation rate of building materials. The principle is based on flushing the radon activity concentration from a free volume during a specific period, using nitrogen, and by collecting it on an adsorbent material. This method, although precise, requires a high investment cost, in addition to the repeated use of nitrogen, as well as long sampling periods (sample preparation and testing). For this reason, based on the operating principle of these methodologies, in which the accumulation of radon in an airtight chamber is analyzed, a study of other methodologies to determine radon levels is proposed.

The main objective is to develop protocols for measuring the radon exhalation rate faster than those established in the ISO 11665–9:2019. It is verified whether the results obtained with new methodologies are adequate and could be established as preferential, thus reducing sampling times.

2. Methods and materials

2.1. Material selection

The selection of building materials is a critical decision in building design and safety considerations. Within granite options, materials sourced from various regions offer a range of qualities, including aesthetic appeal and durability. Notably, granites such as South African granite, Pearl White granite, Rose Porriño granite, Blue Vizag granite, Julia Cream granite, and Absolute Black granite are the most relevant materials in building (EX RADON CSN, 2021).

According to Directive (2013)/59/EURATOM, the external exposure to gamma radiation emitted by indoor construction materials (in addition to external outdoor exposure) is limited to 1 mSv per year. This limit will not be exceeded if the activity concentration index (I) for gamma radiation emitted by construction materials is less than 1 ((Real Decreto 1029/2022 and Directive, 2013/59/EURATOM)). This limit is determined by the activity of ^{226}Ra, ^{232}Th and ^{40}K. The value of 1 for the activity concentration index can be used as a conservative screening tool to identify those materials that may lead to exceeding the reference level established.

For this reason, granites exceeding the threshold of I = 1 have been singled out to analyze their potential radon exhalation rates: Rose Porriño, Blue Vizag and Cream Julia. The radium activity of these samples are greater than 130 Bq·kg^{-1} for the Rose Porriño and Cream Julia granite and around 20 Bq· kg^{-1} for the Blue Vizag granite.

From each granite sample, two different sizes have been chosen for sampling: 19 cm × 19 cm and 6 cm × 6 cm. Sizes and mass are shown at Table 1.

2.2. Selection of radon exhalation detectors

The selection of equipment was based on the type of detector (size, availability, and electrical dependency), as well as whether the measurement reading provided was direct or indirect. After the analysis, one active method continuous detector RAD-7 (Durridge Inc.) (Fig. 1 (A)) and two passive methods: E-PERM electrets (Rad Elec Inc.) (Fig. 1 (B)) and CR-39 alpha tracks (Radosys) (Fig. 1 (C)) were chosen. The following section describes the operating principle for each of these methods.

For the measurement of radon by passive methods (B) and (C), given that the same radon-impermeable jar is used, the size of the samples is $1.22 \cdot 10^{-2}$ m^2. In the case of the samples analyzed with the active method; the size is bigger, with a value of $8.82 \cdot 10^{-2}$ m^2. For the calculations of the exhalation rate, the correction is made according to the size of the sample analyzed and the volume of air inside each experimental equipment. Other parameters necessary to consider are: the radon decay constant, experimental chambers leakage, as well as the initial concentrations inside the container (in this study, background radon concentration).

2.2.1. Continuous monitor detector

RAD-7 is a solid-state alpha detector, a semiconductor material that converts the alpha radiation emitted by radon into electrical signals (RAD7, Durridge). The measurement procedure is based on placing the material piece inside a container and creating a closed loop circuit between the container and the detector. Air is pumped from inside the container through RAD-7 by Teflon tubes before being returned to the container as shown in Fig. 1 (A). The hermeticity of the experimental equipment (container) is calculated according to ISO 11665-13 (2017). Considering the initial radon concentration in 30 L deposit, the system losses constant resulted $\lambda_l = 0.0035\, h^{-1}$ (Noverques Medina, 2022). The measurement period is 14 days hour by hour. This continuous detector can also discern the energy of the ^{222}Rn and ^{220}Rn isotopes separately, so only the concentration due to radon 222 will be quantified.

Based on the results of radon concentration measured within the container, the exhalation rate E (Bq·m^{-2}·d^{-1}), is calculated through three different estimation models.

2.2.1.1. Model 1: Integrated fitting. This model relies on fitting the experimental curve for 14 days measurement time. From the following equation (1), the emission rate E (Bq·d^{-1}), is calculated (Park et al., 2023):

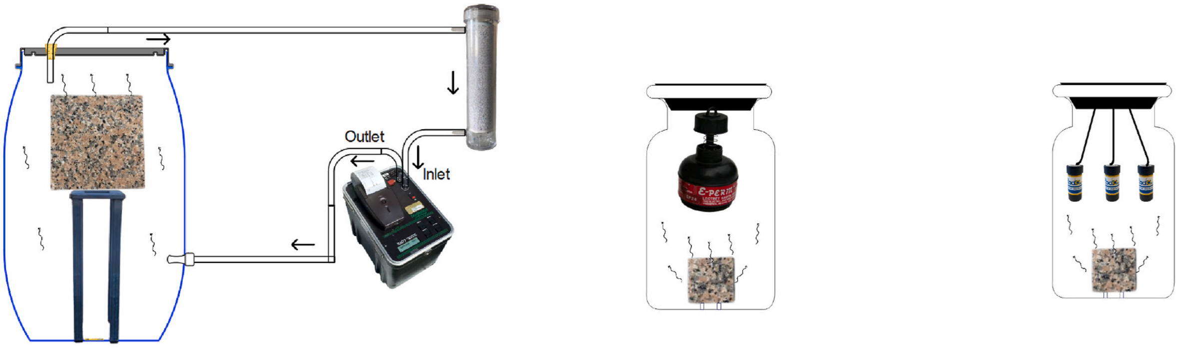
$$C_{Rn}(t) = C_{Rn_0} \cdot e^{-(\lambda+\lambda_l)\cdot t} + \frac{\frac{E}{V} + \lambda_l \cdot C_{Rn\ BG}}{(\lambda+\lambda_l)} \cdot \left(1 - e^{-(\lambda+\lambda_l)\cdot t}\right) \tag{1}$$

Where $C_{Rn}(t)$ is the radon concentration (Bq·m^{-3}); $C_{Rn\ BG}$ is the background radon concentration (Bq·m^{-3}), C_{Rn0} is the initial radon concentration in the container (Bq·m^{-3}), in this work equal to $C_{Rn\ BG}$; λ is the radon decay constant (d^{-1}); λ_l is the container leakage rate (d^{-1}); V is the volume of free air inside the container (m^3) considering the volume occupied by the sample and by the metallic support where the sample is placed;, and t is the time value for each calculated concentration (d). Considering the surface exhalation of the granite sample, exhalation rate E (Bq·m^{-2}·d^{-1}) is calculated.

2.2.1.2. Model 2: Simplified fitting. This model is a simplification of Model 1 according to Park et al. (2023) once the equilibrium is reached in the closed circuit. Currently, in this set-up experimental methodology,

Table 1
Dimensions of samples for radon exhalation measurements.

	Sample size (6 cm × 6 cm)			Sample size (19 cm × 19 cm)		
	Mass (kg)	Area (m^2)	Volume (m^3)	Mass (kg)	Area (m^2)	Volume (m^3)
Rose Porriño	0.19	$1.22{\cdot}10^{-2}$	$7.32\ {\cdot}10^{-5}$	1.93	$8.82{\cdot}10^{-2}$	$7.58{\cdot}10^{-4}$
Blue Vizag	0.19	$1.22\ {\cdot}10^{-2}$	$7.69{\cdot}10^{-5}$	1.95	$8.77{\cdot}10^{-2}$	$7.54{\cdot}10^{-4}$
Cream Julia	0.20	$1.24{\cdot}10^{-2}$	$7.32{\cdot}10^{-5}$	1.93	$8.82{\cdot}10^{-2}$	$7.58{\cdot}10^{-2}$



(A) (B) (C)

Fig. 1. Experimental set-up used for radon exhalation calculation.

the radon concentration after ten days approximately stabilizes and reaches the steady state. The exhalation rate, E, is calculated by equation (2):

$$E = \frac{V}{A} \cdot (\lambda \cdot C_{Rnss} + \lambda_l \cdot (C_{Rnss} - C_{Rn\ BG})) \tag{2}$$

Where, A is the surface exhalation area (m^2) of the granite sample and $C_{Rn\ ss}$ is the steady-state radon concentration ($Bq{\cdot}m^{-3}$), given by equation (3) as an average of C_{Rn} values once steady-state has been reached:

$$C_{Rnss} = \frac{\sum_{i=i\ SS}^{n} C_{Rn\ i}}{n} \tag{3}$$

2.2.1.3. Model 3: Slope-based fitting. This model utilizes the slope of radon concentration during the first 24 h (Stajic, J. M, 2023). An adjustment is made to calculate the exhalation rate E ($Bq{\cdot}m^{-2}{\cdot}d^{-1}$) based on the slope (S) of the radon growth curve during the initial 24 h. Using equation (4), the result of the radon exhalation rate for the measured sample is obtained.

$$E = \frac{V}{A} \cdot (S + C_{Rn_0} \cdot (\lambda + \lambda_l) - \lambda_l \cdot C_{Rn\ BG}) \tag{4}$$

The initial slopes S of radon build-up curves were determined by linear regression of data obtained by RAD7 during the first day of measurements.

2.2.2. Electrets ion chamber

The electret detector is composed of an electret that is attached to a radon accumulation chamber. One short term E-PERM detector (E-PERM User, 2024) has been placed in the impermeable jar, measuring the accumulation of radon inside exhaled by the building sample as shown in Fig. 1 (B). The exposure time with this methodology is 3 days. The detector configuration chosen (ST electret and S chamber) detect less than 3% of the thoron activity, so its contribution to the measured concentration is not significant.

From the obtained radon concentration results the estimation of the exhalation rate E, measured in $Bq{\cdot}m^{-2}{\cdot}d^{-1}$ is obtained according to equation (5) (Kotrappa et al., 2008))

$$E = \frac{V{\cdot}\lambda}{K{\cdot}A} \cdot (C_{Rn} - C_{RnBG}) \tag{5}$$

Where V is the free air volume within the container (m^3) considering the sample size and the electret volume; λ radon decay constant (d^{-1}); K is a constant that depends only on time of exposure in day units; A the exhalation area of the material (m^2) and C_{Rn} is the concentration of radon in air that accumulates inside the jar, integrated according to the exposure time ($Bq{\cdot}m^{-3}$).

2.2.3. Solid-state nuclear track detector

Each chamber was equipped with three SSNTD RSKS CR-39 detectors, sensitive to alpha particles emitted by radon. These particles create tracks in the plastic that are proportional to the number of alpha particles passing through the detector. The count of these tracks is conducted through a chemical development process and subsequent examination under a microscope. The measurement procedure for radon exhalation involves placing the selected material inside the jar, adding 3 alpha tracks detectors, as shown in Fig. 1 (C). The measurement period should be at least 20 days (Radosys User Manual, 2013). At the standard radon chamber the standard air gap acts as a barrier for the thoron gas, so the Rn-channel is almost non sensitive for thoron at all.

This methodology, despite requiring long exposure times, serves as a verification between both passive methods, checking if in both cases the results obtained are in the same range. The mass exhalation rate E_m is calculated according to the following equation (6)

$$E_m = \frac{\rho{\cdot}V{\cdot}\lambda + C_{Rn0}{\cdot}k{\cdot}V{\cdot}(e^{-\lambda{\cdot}t} - 1)}{\frac{m{\cdot}k}{\lambda}{\cdot}(e^{-\lambda{\cdot}t} + t{\cdot}\lambda - 1)} \tag{6}$$

Where ρ is the trace density ($traces{\cdot}cm^{-2}$) produced during the exposure time; V is the volume of free air inside the jar (cm^{-3}); λ is the radon decay constant (d^{-1}); C_{Rn0} is the initial radon concentration within the jar ($Bq{\cdot}m^{-3}$); k is the calibration coefficient of the chamber (($traces{\cdot}cm^{-2}$)/($Bq{\cdot}d{\cdot}cm^{-3}$)); t is the period of time during which the tracks have been measured (d), and m is the sample mass (kg). Considering the sample mass, the exhalation rate E ($Bq{\cdot}m^{-2}{\cdot}d^{-1}$) is obtained.

3. Results

In this section results from the different described methodologies of exhalation measurements are compared, and then the three methods of measurement are analyzed to value which one is the most suitable for Rn exhalation measurement from building materials. The exhalation rate has been calculated using the equations proposed, considering the air volume inside the equipment as well as the dimensions of the granite sample of each experimental configuration. The background concentration, used in calculations as initial and external air concentration is: 26 ± 4 Bq·m^{-3}.

3.1. Comparison of continuous monitor detector models

In this section, the results of radon concentration based on the methodology corresponding to RAD-7 are presented: three different mathematical models to obtain the exhalation rate have been compared. Fig. 2 shows the results obtained by applying the three models to Rose Porriño granite sample.

3.2. Model 1: Integrated fitting

In Fig. 2 (A), the experimental radon concentration results are depicted as blue data points obtained from RAD-7 ($CRn\,exp$). The theoretical curve, according to equation (1) is represented by a blue line ($CRn\,t$), and the upper limit (UL) and lower limit (LL).) are represented by dashed green and red lines, respectively. The exhalation rate is obtained through the fitting of this curve, yielding the optimal value for the theoretical fit to closely resemble the experimental data. The upper limit (UL) and lower limit (LL) have been calculated based on the uncertainty associated with each measurement. The same procedure as described is used for calculating these limits, employing the uncertainty from the experimental concentrations. Therefore, the exhalation rate from Model 1 is: 34 ± 4 (Bq·m^{-2}·d^{-1}).

3.2.1. Model 2: Simplified Fitting

According to Model 2, the simplified fit requires the steady-state concentration (CRn_{SS}). The radon concentration grows exponentially until it reaches a steady state concentration, where it remains approximately stable after 8 days of exposure.

In Fig. 2 (B), the experimental radon concentration data is represented by blue dots ($CRn\,exp$), and three dashed lines represent the average radon concentration for the steady state—blue for the actual data (CRn_{SS}), and green and red for the upper limit (CRn_{UL}) and lower limit (CRn_{LL}), respectively. Additionally, the highlighted area indicates the region where the concentration stabilizes, and the average at that point is considered to calculate the steady-state concentration.

The radon concentration in the air stabilizes after 250 h (10 days) until the end of the test, as observed in Fig. 2 (B). The results obtained during the following 83-h period have been used for the calculation of the exhalation rate. Applying from Model 2 the exhalation rate is: 32 ± 4 (Bq·m^{-2}·d^{-1}).

3.2.2. Model 3: Slope-based fitting

According to Model 3, the slope for the first 24 h of radon increasing concentration is used. Fig. 2 (C) shows the experimental radon concentration ($CRn\ exp$) from the first 24 h that are selected. The experimental concentration data are displayed in blue, the slopes for the upper limit (Slope $_{UL}$) in green and lower limit (Slope $_{LL}$) in orange are plotted. According to equation (7), the radon exhalation is 32 ± 2 (Bq·m^{-2}·d^{-1}).

For the Julia Cream granite and Blue Vizag granite same procedure has followed. According to Julia Cream granite results, there is an accumulation of radon concentration in air inside the experimental device, from background levels until 724 ± 65 Bq·m^{-3}. However, at Blue Vizag granite sample, its experimental radon concentration, measured by RAD-7, are around background levels due to the poor exhalation of this material. So, there is no accumulation inside the experimental device and calculations of the exhalation rate were not carried out for the different models since there was no growth curve.

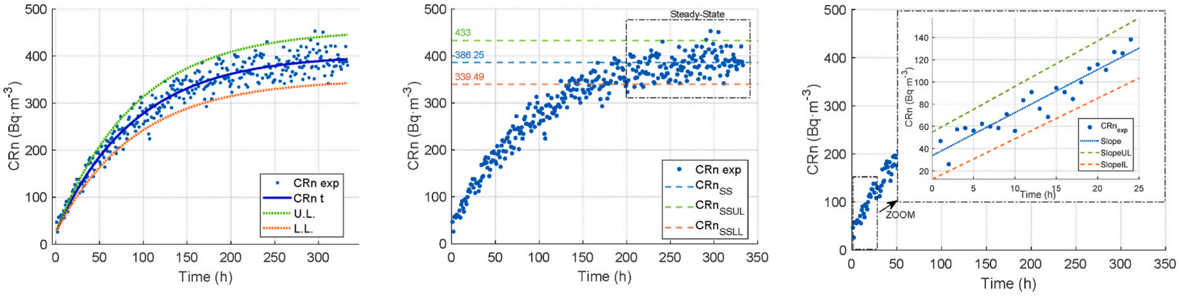
Table 2 shows the results of exhalation rate for the Rose Porriño and Julia Cream samples. The relative error between the three models has been calculated by choosing the value of Model 1 as the reference value, since it considers all the experimental data, and the other two models are a simplification of this one.

As observed in the previous Table 2, the results obtained through all three models fluctuate within a similar range for each sampled granite piece. This consistency suggests a degree of agreement among the methodologies in assessing radon exhalation levels, bolstering confidence in the reliability of the findings across different analytical approaches. Since the relative error of Model 2 and Model 3 are below the commonly accepted threshold of 15% in environmental measurements, either of the alternate models could be used for calculating radon gas exhalation in construction materials. However, Model 1 has been chosen as the most suitable one because it includes all radon data collected over extended periods, providing a comprehensive understanding of its accumulation, and due to its notable reduction of testing periods to 24 h, which represents a significant advancement over previous methodologies and aligns favorably with the approach outlined in ISO 11665-9.

3.3. Analyses of measurement technologies

In this section, a comparative analysis will be conducted between Model 1 of RAD-7 and the other two methodologies based on the passive EIC and SSTND systems for measuring radon exhalation in construction materials. The error has also been calculated using the value obtained for RAD-7 Model 1 as the reference. This examination aims to discern the strengths and limitations of each approach, providing valuable insights into their effectiveness and applicability within the context of radon management.

As Blue Vizag granite showed a very low concentration of radon this



(A) (B) (C)

Fig. 2. Experimental data fitted to Model 1(A), Model 2 (B), and Model 3(C) (Rose Porriño granite).

Table 2
Exhalation rates for different granite samples measured by RAD-7.

	Methodology		Exhalation rates ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)	Error (%)
Rose Porriño granite	RAD - 7	Model 1: Integrated	34 ± 5	–
		Model 2: Simplified	32 ± 4	4
		Model 3: Slope	32 ± 2	7
Julia Cream granite	RAD - 7	Model 1: Integrated	60 ± 6	–
		Model 2: Simplified	59 ± 5	3
		Model 3: Slope	52 ± 4	14

Table 3
Comparison of radon exhalation rates according to different methodologies.

	Methodology	Exhalation rate ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)	Error (%)
Rose Porriño	RAD - 7	34 ± 4	–
	Electrets	33 ± 3	3
	SSNTD	37 ± 3	10
Julia Cream	RAD - 7	60 ± 6	–
	Electrets	60 ± 4	1
	SSNTD	66 ± 5	10

sample has not been analyzed using either electrets or SSNTD detectors. Results for Rose Porriño granite and Julia Cream granite are shown in Table 3. It is observed that the radon exhalation rate fluctuates within a similar range for the three different methodologies. The error obtained, with RAD7 as the reference, again falls below 15%, indicating that all three methodologies exhibit a comparable response for this type of measurement. This consistency across methodologies reinforces the reliability of the results and suggests their suitability for assessing radon exhalation in the tested materials.

4. Conclusions

In this research, different methodologies and procedures for measuring radon exhalation from three building materials that exceed the legal limit of the radioactivity activity index (I): Rose Porriño, Blue Vizag, and Julia Cream. Three methods have been studied to measure the exhalation rate: continuous monitor (RAD-7, Durridge), electrets (E-PERM, Rad Elec Inc.), and alpha tracks SSNTD (Radosys).

The experiments conducted with continuous RAD-7 monitor have been carried out by fitting the experimental results to three mathematical models, which have differences in the period of measurement and the way of processing results. From the results obtained with the RAD7, it is evident that all three models yield similar results and could be used indistinctly, since the calculated error between them remains below 15%. However, Model 1 is the most suitable one since it considers all the data collected and reduces measurement time in comparison with ISO 11665-9.

Furthermore, using the integrated Model 1 as a reference, the exhalation results are compared between the electrets detector and track methodologies. This rate varies within the same range for each granite sample: 32 ± 2 and 34 ± 5 ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) for Rose Porriño and 52 ± 4 to 60 ± 6 ($\text{Bq}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) for Julia Cream. The Blue Vizag sample, despite exceeding the threshold of the radioactive activity index, shows very low accumulation of radon inside the experimental setup, at background levels. It is considered that this could be attributed to the treatment applied to its finishing (painting, varnish, etc.).

Based on the developed protocols and the obtained results, it is concluded that all three techniques could be suitable for measuring radon exhalation in construction materials. The consistency of results across methodologies suggests their robustness and reliability, thereby providing flexibility in selecting an appropriate approach based on specific experimental requirements or constraints.

CRediT authorship contribution statement

P. Moreno: Writing – original draft, Validation, Software, Methodology. **A. Noverques:** Writing – review & editing, Validation, Software, Methodology, Investigation, Conceptualization. **B. Juste:** Writing – review & editing, Validation, Supervision, Methodology, Conceptualization. **M. Sancho:** Writing – review & editing, Validation, Supervision, Methodology, Conceptualization. **G. Verdú:** Writing – review & editing, Validation, Supervision, Software, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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