Numerical Simulation of Radon Migration and Exhalation Behavior During Closed-Loop Measurement of Radon Exhalation Rate

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Abstract

Accurate measurements of the radon exhalation rate helps identify and evaluate radon risk regions in the environment. Among these measurement methods, the closed-loop method is frequently used. However, traditional experiments are insufficient or cannot analyze the radon migration and exhalation patterns at the gas–solid interface in the accumulation chamber. The CFD-based technique was applied to predict the radon concentration distribution in a limited space, allowing radon accumulation and exhalation inside the chamber intuitively and visually. In this study, three radon exhalation rates were defined and two structural ventilation tubes were designed for the chamber. The consistency of the simulated results with the variation in the radon exhalation rate in a previous experiment or analytical solution was verified. The effects of the vent tube structure and flow rate on the radon uniformity in the chamber; permeability, insertion depth, and flow rate on the radon exhalation rate; and the effective diffusion coefficient on back diffusion were investigated. Based on the results, increasing the insertion depth from 1 to 5 cm decreased the effective decay constant by 19.55%, whereas the curve-fitted radon exhalation rate decreased (lower than the initial value) as the deviation from the initial value increased by approximately 7%. Increasing the effective diffusion coefficient from 2.77$\times 10-7to7.77\times$10-6 m2 s-1 made the deviation expand from 2.14% to 15.96%. The conclusion is that an increased insertion depth helps reduce leakage in the chamber, subject to notable back-diffusion, and that the closed-loop method is reasonably used for porous media with a low effective diffusion coefficient in view of the back-diffusion effect. The CFD-based simulation is expected to provide guidance for the optimization of the radon exhalation rate measurement method and, thus, the accurate measurement of the radon exhalation rate.

Full Text

Preamble

Numerical Simulations for Radon Migration and Exhalation Behavior During Measurement of Radon Exhalation Rate with Closed-Loop Method

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Highlights - Transient variation during radon exhalation rate measurements was simulated - The effects of porous medium and measurement parameters were examined - The discrepancy between various radon exhalation rates was quantified and analyzed - Simulation results are expected to optimize the closed-loop measurement method

Abstract: Accurate measurements of the radon exhalation rate help identify and evaluate radon risk regions in the environment. Among available measurement methods, the closed-loop method is frequently used. However, traditional experiments are insufficient for analyzing radon migration and exhalation patterns at the gas–solid interface within the accumulation chamber. A CFD-based technique was applied to predict radon concentration distribution in a limited space, enabling intuitive and visual examination of radon accumulation and exhalation inside the chamber. In this study, three radon exhalation rates were defined and two structural ventilation tube designs were implemented for the chamber. The consistency of simulated results with variation in radon exhalation rate from previous experiments and analytical solutions was verified. The effects of vent tube structure and flow rate on radon uniformity in the chamber; permeability, insertion depth, and flow rate on the radon exhalation rate; and the effective diffusion coefficient on back diffusion were investigated. Results showed that increasing insertion depth from 1 to 5 cm decreased the effective decay constant by 19.55%, whereas the curve-fitted radon exhalation rate decreased (falling below the initial value) as deviation from the initial value increased by approximately 7%. Increasing the effective diffusion coefficient from 2.77×10^{-7} to 7.77×10^{-6} m^2 s^{-1} expanded the deviation from 2.14% to 15.96%. The conclusion is that increased insertion depth helps reduce chamber leakage but is subject to notable back-diffusion, and that the closed-loop method is reasonably applied to porous media with low effective diffusion coefficients when considering the back-diffusion effect.

The CFD-based simulation is expected to provide guidance for optimizing the radon exhalation rate measurement method and thus enabling accurate measurement of the radon exhalation rate.

Keywords: Radon exhalation; Numerical simulation; Accumulation chamber

1 Introduction

Radon, a colorless and odorless naturally radioactive gas, is the second leading cause of lung cancer [?, ?]. Widely found in building materials, soil, rocks, and uranium mill tailings [?], concerns have been raised regarding radon released from these media into ambient air. An accurate radon exhalation rate can be used in environmental assessments to quantify and identify radon-risk areas [?]. Currently, radon exhalation has been studied extensively [?], with commonly used measurement methods including activated carbon [?, ?], open loop [?, ?], and closed loop [?]. For closed-loop systems, the effects of leakage from the accumulation chamber, back-diffusion at the gas–solid interface, and sampling flow rate during radon exhalation rate measurement are nonnegligible [?, ?]. Increased insertion depth has proven effective in reducing chamber leakage [?], though the back-diffusion effect is often ignored. Back-diffusion occurs when radon returns to the porous medium as radon concentration increases in the accumulation chamber [?], affecting radon exhalation at the medium surface as well as measurement of the radon exhalation rate [?]. Moreover, convection (or advection) driven by pressure gradients is an important factor influencing radon exhalation [?]. Conventionally, radon exhalation rate measurements for porous media with unknown physical parameters do not show discrepancy between measured and initial values. Limitations of experimental tests, such as control of environmental factors and experimental parameters, make experiments difficult or time-consuming. Although analytical solutions of 1D and 2D models can reflect radon migration patterns in steady or transient states, they are subject to various assumptions such as ignoring insertion depth, lateral diffusion, specific dimensions of the accumulation chamber or porous media, and constant uniform concentration distribution inside the chamber [?]. Diffusion driven by concentration gradients and convection driven by pressure gradients govern radon migration in porous media. However, traditional measurement methods cannot provide details of radon migration in the chamber and gas flow in the porous medium, which are key data for analyzing underlying reasons for deviation of measured radon exhalation rate from true values. Advancement of computational fluid dynamics (CFD) has made it possible to numerically solve the Navier-Stokes equations and has been maturely applied to scenarios such as soil, atmosphere, indoor environments, underground space pollutant migration, and nuclear reactor systems [?], providing researchers with solutions to complex scientific problems. Relevant simulations have been performed to predict radon concentration distribution and migration in limited spaces [?], showing good consistency between experimental and simulation results. Additionally, a two-phase flow and heat transfer simulation with a simplified porous media model was established for engineering purposes, and results showed good agreement with experimental data [?]. Therefore, CFD-based 3D numerical simulation exhibited spatial and temporal radon migrations in the accumulation chamber and porous media and revealed variation in radon exhalation at the gas–solid interface during measurement.

In this study, two types of vent tube structures (open-ended and half-open) in the accumulation chamber were modeled, and three radon exhalation rates (initial, transient, and fitted radon exhalation rate) were defined to quantify radon exhalation behavior. Using uranium mill tailings as the research object, a geometric model of a radon exhalation rate-measuring device and a mathematical model for radon migration were established. The CFD-based technique was employed to demonstrate radon migration patterns under coupling of multiple factors (flow rate, insertion depth, diffusion coefficient, and permeability), thus analyzing variations of fitted and transient radon exhalation rates at the gas–solid interface.

The simulated results are consistent with previous experimental phenomena and are expected to optimize design for accurate measurement of radon exhalation rate using the closed-loop method.

2.1 Theory of Radon Diffusion and Exhalation in Porous Media

One-dimensional steady-state diffusive migration of radon in a porous medium can be described using the following equation:

$$\frac{d^{2C}}{dx^2} - \frac{\lambda_{Rn}\eta}{D}C + \frac{\alpha}{D} = 0$$

where C is the radon concentration in the air (Bq m^{-3}), x is the vertical depth of the porous medium (m), D is the diffusion coefficient of radon in the porous medium (m^2 s^{-1}), η is the porosity (%), α is the free radon production rate (Bq m^{-3} s^{-1}), and λ_{Rn} is the decay constant of radon, 2.1\$×10^{-6}\$ s^{-1} [?]. The boundary conditions are $C(0) = C_a$ at the top and $C(x = h) = 0$ at the bottom of the porous medium, respectively (Fig. 1 [Figure 1: see original paper]). The radon concentration in boundary air was significantly lower than that in the porous medium, making it possible to assume $C_a = 0$ Bq m^{-3}. Upon substituting $l = \sqrt{D/(\lambda_{Rn}\eta)}$ for L, the analytical solution is:

$$C(x) = \frac{\alpha}{\lambda_{Rn}\eta}\left[1 - \frac{\cosh((h-x)/l)}{\cosh(h/l)}\right]$$

The radon exhalation rate for diffusion is then theoretically deduced:

$$E_{a-d} = \eta D \left. \frac{dC}{dx} \right|_{x=0} = \frac{\alpha l}{\eta} \tanh \left(\frac{h}{l} \right)$$

where E_{a-d} is the diffusive radon exhalation rate (Bq m^{-2} s^{-1}).

2.2 The Closed-Loop Method and Device for Radon Exhalation Rate Measurement

The closed-loop method is commonly used for measuring radon exhalation rate, involving radon accumulation in a chamber (Fig. 2 [Figure 2: see original paper]). Radon exhaled from the porous medium surface flows into a radon detector (e.g., RAD7 from Durridge Company Inc., USA) driven by an air pump and returns to the chamber through the flow-in vent tube. Inside the accumulation chamber, radon accumulation is described as:

$$V \frac{dC}{dt} = ES_a - (\lambda_{Rn} + \lambda_b + \lambda_l) VC$$

where t is the accumulation time (s), E is the radon exhalation rate (Bq m^{-2} s^{-1}), S_a is the exhaled area of the porous medium (m^2), λ_b is the back-diffusion rate of radon in the chamber (s^{-1}), λ_l is the leakage rate of radon in the chamber (s^{-1}), V is the chamber volume (m^3), and λ_e is the effective decay constant, equal to the sum of radon decay, back-diffusion, and leakage rates ($\lambda_e = \lambda_{Rn} + \lambda_b + \lambda_l$) (s^{-1}).

With boundary condition $C(0) = C_0$, and substituting $C_0 \approx 0$ Bq m^{-3}, the curve fitting solution is:

$$C(t) = \frac{ES_a}{\lambda_e V} (1 - e^{-\lambda_e t})$$

The linear fitting solution is [**?**]:

$$\ln \left(1 - \frac{C(t) \lambda_e V}{ES_a} \right) = -\lambda_e t$$

In subsequent calculations, fitted radon exhalation rates E_{f-exp} and E_{f-lin} were obtained by curve fitting (Eq. 4) and linear fitting (Eq. 5), respectively, with criterion $R^2 > 0.9$. The R^2 calculation involves residual sum of squares (RSS) and total sum of squares (TSS):

$$R^2 = 1 - \frac{\text{RSS}}{\text{TSS}} = 1 - \frac{\sum_{i=1}^{n} (y_i - f_i)^2}{\sum_{i=1}^{n} (y_i - \bar{y})^2}$$

where n is the total number of data points, y_i is the actual data point, f_i is the value obtained from the fitted curve, and $\bar{y}$ is the mean value of all data points.

3.1 Measuring Device and Geometric Model

The three-dimensional geometry for the accumulation chamber (AC) model had internal dimensions of 30 cm × 30 cm × 10 cm with a wall thickness of 4 mm, inserted into uranium mill tailings (filled to a height of 50 cm in a container of 100 cm × 100 cm × 60 cm). Two vent tubes (Ø6 mm) were designed on the chamber diagonal, positioned 3 cm from each hole to the inner wall. In addition to the AC model, a reference (R) model was established—a container of 100 cm × 100 cm × 60 cm—to determine the reference radon exhalation rate by the closed-loop method for uranium mill tailings (filled to 50 cm height) sealed in the container with two holes (Ø6 mm, 50 cm apart) on the top cover.

Considering disturbance of the porous medium caused by jet flow from the inlet vent tube, two vent tube schemes were designed: open-ended and half-open. The first type features a vent tube with an open end. The second was sealed at the bottom end of the vent tube with four holes (Ø2 mm, 2 cm apart, with normal directions at 90° between two adjacent holes) opened on the tube wall to avoid direct impact of the jet on the porous medium. The chamber had a thickness of 4 mm, and its volume was subtracted from that of the porous medium upon insertion.

The geometry was discretized into polyhedral and hexahedral elements using Fluent Meshing to reduce memory and computing time. The mesh was refined in regions where rapid changes in radon concentration and air flow were expected (e.g., the gas–solid interface and local openings of the vent tube), as shown in Fig. 4 [Figure 4: see original paper].

3.2 Governing Equations

Gas flow, regarded as incompressible, in the chamber and porous medium follows conservation laws for mass and momentum. The mass conservation equation is [**?**]:

$$\nabla \cdot \mathbf{v} = 0$$

where $\mathbf{v}$ is the physical velocity vector (m s^{-1}).

The momentum conservation equation is [**?**]:

$$\rho_a \frac{\partial \mathbf{v}}{\partial t} + \rho_a \nabla \cdot (\mathbf{v}\mathbf{v}) = -\nabla p + \rho_a \mathbf{g} + \nabla \cdot \tau + \mathbf{F}$$

where ρ_a is air density (kg m^{-3}), p is static pressure (Pa), $\mathbf{g}$ is gravitational vector (m s^{-2}), and $\mathbf{F}$ is the source term of external body forces (N m^{-3})

describing porous medium viscosity, present only in the porous zone. Laminar flow is described as:

$$\mathbf{F} = -\frac{\mu}{K}\mathbf{v}$$

where K is permeability (m^2) and μ is dynamic viscosity (Pa s).

The viscous stress tensor (τ) is [**?**]:

$$\tau = \mu_t \left[\nabla \mathbf{v} + (\nabla \mathbf{v})^T\right]$$

where μ_t is turbulent viscosity (Pa s), defined by $\mu_t = C_\mu \rho_a k^2/\varepsilon$ (C_μ is a constant, k is turbulent kinetic energy (m^2 s^{-2}), and ε is turbulent dissipation rate (m^2 s^{-3})).

The governing equation for radon migration is:

$$\frac{\partial C}{\partial t} + \nabla \cdot (C\mathbf{v}) = \nabla \cdot (D \nabla C) + S_{Rn}$$

where D is the radon diffusion coefficient (m^2 s^{-1}), $\mathbf{v}$ corresponds to physical convection velocity, and S_{Rn} is the source term (Bq m^{-3} s^{-1}). In a porous medium, S_{Rn} is determined by source and decay terms ($\alpha - \lambda_{Rn} C\eta$), and $D = D_m$. In the accumulation chamber, porosity is set to 1; S_{Rn} is determined by the decay term ($-\lambda_{Rn} C$) and $D = D_m + D_t$ (D_t is turbulent diffusion coefficient, m^2 s^{-1}, defined by $D_t = \mu_t/(\rho_a Sc_t)$, where turbulent Schmidt number $Sc_t = 0.7$ [**?**]; D_m is molecular diffusion coefficient of radon in air, 1.05$×10^{-5}$ m^2 s^{-1}).

3.3 Parameters for Simulation

Simulated scenarios involved reference (R) and accumulation chamber (AC) models. In the AC model, effects of vent tube structure and porous medium permeability on radon exhalation were analyzed. Additionally, effects of insertion depth (H1/H3/H5 corresponding to 1/3/5 cm), flow rate (0.5/1/2 L min^{-1}), and effective diffusion coefficient (D1/D2/D3/D4 corresponding to 2.77$×10^{{-7}/7.77×10}{-7}/2.77×10^{{-6}/7.77×10}{-6}$ m^2 s^{-1}) were tested. The effective diffusion coefficient of radon in a porous medium is defined as [**?**]:

$$D_e = D_m \frac{\eta}{\tau}$$

where τ is the tortuosity factor, 0.66 (dimensionless) [**?**, **?**]. In the R model, the radon exhalation rate obtained from the reference container is used to evaluate the rate measured using the AC model. The R model design improves simulation reliability, considering that reference radon exhalation rate is commonly used in laboratory tests to evaluate the radon exhalation rate obtained from the AC model.

The free radon production rate (α) is defined by [?]:

$$\alpha = \rho_s A_{Ra} E_{Rn} \lambda_{Rn}$$

where ρ_s is porous medium density (kg m^{-3}), A_{Ra} is radium activity concentration (Bq kg^{-1}), and E_{Rn} is radon emanation coefficient (dimensionless). Radium activity concentration in uranium mill tailings commonly ranges from less than 5 kBq kg^{-1} to as high as 10 kBq kg^{-1} [?], with emanation coefficient ranging from 0.1 to 0.35 [?], dry-bulk density of 1800 kg m^{-3}, and average porosity of 0.4 measured using the drainage method for laboratory samples.

Laminar flow in a porous medium follows Darcy's law, and permeability—an intrinsic property of a porous medium—is defined by the Kozeny-Carman empirical equation [?]:

$$K = \frac{D_p^2 \eta^3}{180(1-\eta)^2}$$

where D_p is average grain size (mm) and K is permeability (m^2). After sieving tailings sand in the laboratory, D_p was determined to be 0.5 mm and K calculated as 2.47×10^{-10} m^2. Physical parameters for the simulations are listed in Table 1 .

Table 1 Physical parameters used in the simulation

Physical parameters	Value
Free radon production rate (Bq m^{-3} s^{-1})	3 [?, ?]
Porosity	0.4 [?]
Effective diffusion coefficient for radon (m^2 s^{-1})	$2.77\times10^{-7}/7.77\times10^{-7}/2.77\times10^{-6}/7.77\times10^{-6}$ [?]
Permeability (m^2)	$1\times10^{-9}/1\times10^{-10}/1\times10^{-11}$ [?]
Flow rate (L min^{-1})	0.5/1/2
Insertion depth (cm)	1/3/5

The finite volume method (FVM) was adopted for numerical calculations. The k-ε turbulence model was applied to the chamber zone due to confined jet flow from the four openings on the vent tube surface, whereas laminar flow was modeled in the porous media. Empirical constants in the k-ε turbulence model equations [?] are: $C_\mu = 0.09$, $C_{1\varepsilon} = 1.44$, $C_{2\varepsilon} = 1.92$, $\sigma_k = 1.00$, and $\sigma_\varepsilon = 1.30$.

To simplify the model, the following assumptions were made: container leakage was not considered, the porous medium was homogeneous, air was treated as an incompressible gas, and gravity and temperature effects were ignored.

3.4 Initial Value and Boundary Settings

The radon concentration distribution in the porous medium under steady state was calculated before simulating radon accumulation during measurement, with radon concentration in air set to 0 Bq m^{-3}. Transient mode was adopted for measurement, with initial radon concentration at 0 s in the chamber set at 0 Bq m^{-3}. Detailed boundary settings are listed in Table 2 .

Table 2 Boundary settings in the simulation

Boundary	Type	Settings
Inlet/Outlet	Velocity-Inlet	Inlet: 0.5/1/2 L min^{-1}, normal to boundary; radon concentration equals that from outlet. Outlet: -0.5/1/2 L min^{-1}, normal to boundary; radon concentration gradient equals 0 Bq m^{-4}
Surface of porous medium not covered by chamber	Pressure-Inlet	0 Pa; radon concentration equals 0 Bq m^{-3}
Openings of vent tube inside chamber	Interior	Radon flux equals 0 Bq m^{-2} s^{-1}
Gas–solid interface	Interior	-
Wall of geometry	Wall	-

3.5 Mesh Independence

The three-dimensional geometric model was discretized into coarse, medium, and refined meshes (0.5 M, 1.2 M, and 2.3 M cells, respectively), with radon exhalation rates calculated as 1.4676593, 1.4674347, and 1.4674092 Bq m^{-2} s^{-1}, respectively (Fig. 5 [Figure 5: see original paper]). The decline in radon exhalation rate slowed with increasing cell count. The mesh discretization criterion is to minimize error between numerical and analytical results while maintaining low computational cost. Thus, medium mesh was used in subsequent simulations. Calculations were considered converged when residuals fell below 10^{-10} and the differential value of radon exhalation rate between two consecutive iterations fell below 10^{-7}.

4 Data Processing

As the FVM method was adopted for numerical calculation, volume-averaged radon concentration (C_{v-avg}) in the chamber was obtained using discrete data approximation:

$$C_{v-avg} = \frac{\sum_{i=1}^{n} V_i C_i}{\sum_{i=1}^{n} V_i}$$

where n is the total number of cells inside the chamber, V_i is the volume of cell i (m^3), and C_i is the radon concentration in cell V_i.

The area-averaged radon concentration (C_{s-avg}) at the outlet is:

$$C_{s-avg} = \frac{\sum_{i=1}^{m} A_i C_i}{\sum_{i=1}^{m} A_i}$$

where m is the total number of faces on the outlet surface and A_i is the area of face i (m^2).

In numerical simulation, the transient radon exhalation rate for diffusion was calculated as:

$$E_{n-d} = -\eta D_e \frac{\partial C}{\partial x}\bigg|_{x=0}$$

The transient radon exhalation rate for convection was calculated by:

$$E_{n-c} = \eta v C|_{x=0}$$

where v is the physical velocity of gas convection at the porous medium-air interface (m s^{-1}). The total transient radon exhalation rate is defined by:

$$E_n = E_{n-d} + E_{n-c}$$

The initial radon exhalation rate, corresponding to the radon exhalation rate from the porous medium surface in its natural state, is the value intended to be measured experimentally. In numerical simulation of homogeneous porous medium, the initial radon exhalation rate was equivalent to the value calculated using Eq. 3.

The nonuniformity of radon concentration distribution in the chamber is defined as [?]:

$$\text{NUI} = \frac{C_{v-max} - C_{v-min}}{C_{v-avg}}$$

where C_{v-max} is the maximum radon concentration in the chamber (Bq m^{-3}) and C_{v-min} is the minimum radon concentration (Bq m^{-3}). A lower NUI indicates better uniformity.

5.1 Effect of Vent Tube Structure on Radon Concentration

Prior to experimental simulation, vent tube structures of the chamber were designed and reconstructed. Two schemes were established: vent tube with end opening for Case 1 and vent tube with four side wall openings (Ø2 mm) for Case 2 (both under 1 L min^{-1}, 1$×10^{-9}$ m^2). The growth curves of radon concentrations for these cases are shown in Fig. 6a [Figure 6: see original paper]. Additionally, permeability in Case 2 was changed to 1$×10^{-11}$ m^2 (Case 3 with four openings) to test permeability effects.

Prior to reconstruction (Case 1), C_{s-avg} was larger than C_{v-avg} in the first 100 s, with the gap narrowing and remaining constant thereafter. However, C_{v-avg} was observed to grow faster than C_{s-avg} given that initial radon concentration equaled 0 Bq m^{-3}, implying defects in the vent tube structure in Case 1. Upon modification (Case 2), C_{s-avg} increased more slowly than C_{v-avg} and the discrepancy remained constant. C_{v-avg}, representing the actual trend of radon concentration in the chamber, was used for subsequent simulations to study radon migration.

The radon concentration contours for Cases 1 and 2 at 600 s are shown in Fig. 7 [Figure 7: see original paper]. Streamlines in Case 1 show that the jet from the vent tube enters the porous medium and then returns to the chamber, directly affecting radon concentration distribution in the porous medium and leading to inhomogeneity of radon concentration in the chamber. Streamlines in Case 2 remain mainly in the chamber, indicating minimal impact on radon concentration distribution in the porous medium. For Case 2, maximum, minimum, and average radon concentrations in the chamber are 8676.49, 6208.08, and 7089.7 Bq m^{-3}, respectively, while for Case 1 they are 9473.19, 4444.73, and 6575.9 Bq m^{-3}. The NUI values for Cases 2 and 1 are 34% and 76%, respectively, revealing that uniformity of radon concentration distribution in Case 2 improved significantly.

5.2 Permeability Effect on Radon Exhalation Rate

The total transient radon exhalation rate (E_n) on the surface covered by the chamber at 600, 1200, and 1800 s were 1.03988, 0.84632, and 0.69968 Bq m^{-2} s^{-1} for Case 2, and 1.03943, 0.84595, and 0.69937 Bq m^{-2} s^{-1} for Case 3. The approximate similarity of radon exhalation rates, as well as contours and streamlines for Cases 2 and 3, implies that permeability effects on closed-loop measurement of radon exhalation rate can be neglected. Therefore, permeability was set to 1$×10^{-10}$ m^2 in subsequent simulations.

5.3 Reference Radon Exhalation Rate

For assessment of radon exhalation rate under the AC model, reference exhalation rate (E_{ref}) under the R model was calculated. The C_{v-avg} growth curve from 0 to 1.2$×10^{7}$ s was obtained by radon accumulation until saturation in the sealed container (Fig. 6b [Figure 6: see original paper]). The initial (E_a), fitted (E_{f-exp}), and total transient radon exhalation rates (E_n) were calculated, as shown in Fig. 6c [Figure 6: see original paper]. As the transient radon exhalation rate for convection remains approximately 0 Bq m^{-2} s^{-1}, the effect of convection on exhalation rate measurement can be neglected. Thus, the total transient radon exhalation rate was determined by the transient radon exhalation rate for diffusion (E_{n-d}), represented by the overlap of E_n and E_{n-d} in Fig. 6c.

The fitted radon exhalation rate characterizes average exhalation for a given duration. Upon saturation of radon accumulation, E_{f-exp} was approximately equal to E_n, suggesting consistency of the fit, as well as the back-diffusion phenomenon, with E_{f-exp} (0.520835 Bq m^{-2} s^{-1}) being only 35.59% equivalent to E_a (1.463235 Bq m^{-2} s^{-1}).

5.4 Flow Rate and Insertion Depth Effect on Radon Exhalation Rate

Increased insertion depth of the chamber has been experimentally proven to reduce leakage during measurements. This section analyzes effects of insertion depth (H1/H3/H5) and flow rate (0.5/1/2 L min^{-1}) on radon concentration and exhalation rate. Radon distribution in the accumulation chamber at 1800 s is shown in Fig. 8 [Figure 8: see original paper]. At low flow rate (0.5 L min^{-1}), radon concentration distribution shows notable spatial variation, gradually becoming uniform as flow rate increases. Additionally, lateral diffusion is responsible for the average concentration in the chamber for H1 being lower than that for H5, implying that insertion depth effects on measurement cannot be neglected.

Excluding points in the initial 60 s, the average value for every 60 s was calculated as the concentration at the midpoint and further fitted to obtain the exhalation rate. For example, average radon concentration from 61 s to 120 s was calculated as the value at 90 s. Duration for curve fitting included 600, 720, 900, 1020, 1200, 1500, and 1800 s, while for linear fitting it was 240, 360, 480, 600, 720, 840, 960, 1080, 1200, 1320, 1440, and 1560 s.

E_{f-exp} and E_{f-lin} were obtained by fitting points to the curve and linearly fitting equations using different time ranges, as shown in Fig. 9 [Figure 9: see original paper]. Long duration (1800 s) is unfavorable for curve fitting, with discrepancy from E_a enlarged (0.9% to 3.3% increase in deviation for various insertion depths compared with 600 s). In contrast, linear fitting values declined significantly with increasing duration (14.7% to 22.8% increase in deviation for various insertion depths from 240 s to 1800 s).

Both E_{f-exp} and E_{f-lin} increased with flow rate for a fixed insertion depth, approaching E_a. Throughout the accumulation duration, E_n maintained a decreasing trend and was only 48–59% (for various insertion depths) of E_a at 1800 s, indicating that E_f is an averaged value for a certain duration and cannot represent E_n at a specific moment. Moreover, E_f was more susceptible to back-diffusion for longer fitting durations.

For in-depth analysis of insertion depth effects (1 L min^{-1}), E_{f-exp} and E_{f-lin}, as well as the effective decay constant (λ_e), were calculated (Fig. 10 [Figure 10: see original paper]). λ_e (2.23582$×10^{-6}$ s^{-1}) is comparable to λ_{Rn} during closed-loop measurement in the R model, given the assumption of radon-impermeable walls in the sealed container. A significant decrease of 27.2%–34.2% is observed for λ_e with increased insertion depth, implying that leakage was effectively reduced, consistent with previous experimental observations [**?**].

Furthermore, ratios of E_{ref} and E_f to E_a were calculated (Fig. 10b and 10c). The ratio for curve fitting (E_{f-exp}) is positively correlated with λ_e. Increased insertion depth results in lower λ_e and lower ratios, suggesting an obvious back-diffusion effect. Unlike curve fitting, ratios for linear fitting showed slight growth. Specifically, exhalation rates from curve fitting using 1800 s and linear fitting using 480 s for H1 were 0.89952 and 0.81364 Bq m^{-2} s^{-1}, respectively, while for H5 they were 0.89241 and 0.83551 Bq m^{-2} s^{-1}. The effect of increased insertion depth on reduced leakage is demonstrated by linear fitting (i.e., fitted exhalation rate approaches E_a with increased insertion depth), whereas curve fitting for longer durations is more affected by back-diffusion.

In general, uniform mixing of radon inside the chamber facilitates radon exhalation rate measurement as flow rate increases from 0.5 to 2 L min^{-1}. Although increased insertion depth helps lower leakage rate, the side effect is decreased accuracy of curve fitting due to back-diffusion.

5.5 Validation of the Numerical Model

The reliability and accuracy of the numerical model were validated against previous experiments. Gutiérrez-Álvarez [**?**] deployed two reference boxes (RB1 and RB2) to obtain reference radon exhalation rates. Cylindrical (CX, where X denotes insertion depth in centimeters) and rectangular (VX, where X has the same definition) accumulation chambers were designed to evaluate insertion depth influence. V10, C0, C3, C6, C9 were tested on RB1, while V10, C0, C3, C6, C9, C12, and C14 were tested on RB2.

The effective decay constant measured on RB1 (approximately 4.88$×10^{-6}$ s^{-1}) is 62% greater than that on RB2 (approximately 3$×10^{-6}$ s^{-1}) measured by RAD7, whereas it is 58% greater by Alphaguard, implying RB2's container was better sealed during measurement. The effective decay constant for the accumulation chamber peaked at insertion depth of 0 cm (C0), approximately 1.88$×10^{-4}$ s^{-1} on RB1 and 2.5$×10^{-4}$ s^{-1} on RB2. As insertion depth increased, the effective decay constant declined by 38%–52% to almost 2.9$×10^{-5}$ s^{-1}, indicating lower leakage rate. Variation in effective decay constant for reference boxes (RB1 and RB2) and accumulation chamber fit well with simulated results shown in Fig. 10a, decreasing by 27.2%–34.2% with depth increase from 1 to 5 cm.

Furthermore, ratios of curve-fitted radon exhalation rate to reference values for C0, C3, and C6 on RB2 were chosen for comparison with H1, H3, and H5 because RB2's airtightness was better than RB1's. Ratios for C0, C3, and C6 were 1.65, 1.22, and 1.1 measured by Alphaguard, and 1.18, 1.04, and 1.02 by RAD7, respectively. An evident downward trend was observed for both instruments, agreeing well with trends of ratios for H1, H3, and H5 by exponential fitting in Fig. 10b.

Although reference radon exhalation rate (equivalent to the value obtained from the R model in this study) was used as an intrinsic value for comparison, potential discrepancy caused by back-diffusion should be noted. Inaccurate reference values can potentially render comparisons unreliable.

In summary, the simulation not only validates variation in experimental data but also further reveals the positive correlation between back-diffusion phenomenon and increased insertion depth.

5.6 Diffusion Coefficient Effect on Radon Exhalation Rate

Given the inevitable back-diffusion phenomenon mentioned in Section 5.4 (i.e., curve-fitted radon exhalation rate declines with increased insertion depth and lower effective decay constant), the effect of radon diffusion coefficient for the porous medium on E_a, E_f, and E_n was additionally analyzed.

According to Eq. 3, radon exhalation rate is positively correlated with effective diffusion coefficient, explaining why E_a (1.2081 Bq m^{-2} s^{-1}) for D1 scenario (lowest effective diffusion coefficient among four scenarios) is apparently lower than for other scenarios (81.3%–87.7% of the latter in Fig. 11 [Figure 11: see original paper]). Lower radon exhalation requires longer time for radon concentration to reach saturation, evidenced by lower λ_e for D1 (Fig. 12a [Figure 12: see original paper]).

Increasing effective diffusion coefficient causes E_f or E_n to deviate from E_a, suggesting that effective diffusion coefficient is an important factor in the back-diffusion phenomenon.

Ratios of E_f to E_a using various fit durations for the four effective diffusion coefficient scenarios are shown in Fig. 12b and 12c. Ratios decrease with increasing effective diffusion coefficient, from 0.98 to 0.84 for curve fitting (E_{f-exp}, 1800 s) and from 0.96 to 0.67 for linear fitting (E_{f-lin}, 480 s). Deviations between curve (1800 s) and linear (480 s) fitting were 2.08%, 3.88%, 9.55%, and 20.68% for D1, D2, D3, and D4, respectively, making linear fitting a time-saving approximate method for calculating exhalation rate in media with lower effective diffusion coefficients.

6 Conclusion

A CFD-based technique was employed to model three-dimensional geometry and simulate various scenarios for measuring radon exhalation rate beyond traditional methods. The transient radon exhalation rate for diffusion maintained a decreasing trend during the initial 1800 s of measurement due to back-diffusion caused by accumulated radon, whereas convection contributed little to radon exhalation. Results revealed that radon migration patterns are subject to vent tube structure, effective radon diffusion coefficient, insertion depth, and flow rate. Their effects on radon exhalation rate were analyzed, and discrepancies between initial, fitted, and transient exhalation rates were quantified.

The conclusions are as follows:

The improved vent tube structure (tube with side wall openings) in the accumulation chamber was favorable for stable radon concentration distribution in the porous medium, avoiding disturbance from the jet of a tube with end opening. Increased circulation flow rate (from 0.5 to 2 L min^{-1}) further improved uniformity of radon concentration in the chamber (NUI from 76% to 34%), beneficial for accurate radon concentration measurement. Additionally, porous medium permeability had little effect on closed-loop measurement of radon exhalation rate.

The effect of effective diffusion coefficient on radon exhalation rate measurement was notable. Deviation of fitted radon exhalation rate (E_f) from initial value (E_a) was small (max. 2.14%) for relatively low effective diffusion coefficient (D1), but increased for D4 (max. 15.96%), making measurement susceptible to back-diffusion. A lower effective diffusion coefficient indicates lower radon diffusion capability in the porous medium, limiting radon's ability to accumulate inside the chamber at high concentrations and diffuse back into the porous medium. Therefore, the closed-loop method is more suitable for measuring radon exhalation rate on surfaces of compact porous media with low effective radon diffusion coefficients.

Although increased insertion depth (from 1 cm to 5 cm) contributed to decreased effective decay constant (decreases of 23.17% and 19.55% for 600 s and 1800 s curve fitting at 1 L min^{-1}), back-diffusion could not be avoided. This is evident from deviation of exhalation rate using curve fitting (E_{f-exp}) from initial value (E_a), which increased by approximately 7% (1800 s), whereas deviation in linear fitting (E_{f-lin}) decreased by approximately 12% (480 s). Deeper insertion depth reduces leakage rate and, to some extent, increases equilibrium radon concentration inside the chamber, which in turn suppresses radon exhalation from the medium surface. Thus, linear fitting allows radon exhalation rate calculation at lower time cost and reduces back-diffusion effects.

In summary, consistency of numerically simulated results with previous experimental data demonstrates that modeling the closed-loop measurement system of radon exhalation rate by CFD-based numerical simulation is verifiable and reliable. The results are expected to provide theoretical guidance for optimizing closed-loop measurement methods (e.g., circulation flow rate, insertion depth, and applicability of porous media).

Nomenclature and Units

Symbol	Description	Units
A_{Ra}	Radium activity concentration	Bq kg^{-1}
C	Radon concentration in air	Bq m^{-3}
C_a	Radon concentration in atmosphere	Bq m^{-3}
C_{s-avg}	Area-weighted average radon concentration at flow-out tube outlet surface	Bq m^{-3}
C_{v-avg}	Volume-weighted average radon concentration inside accumulation chamber	Bq m^{-3}
C_{v-max}	Maximum radon concentration value inside accumulation chamber	Bq m^{-3}
C_{v-min}	Minimum radon concentration value inside accumulation chamber	Bq m^{-3}
C_0	Initial radon concentration	Bq m^{-3}
D	Radon diffusion coefficient in porous medium	m^2 s^{-1}
D_e	Effective radon diffusion coefficient in porous medium	m^2 s^{-1}
D_m	Molecular diffusion coefficient of radon in air (1.05$×10^{-5}$ m^2 s^{-1})	m^2 s^{-1}
D_p	Average grain size	mm
D_t	Turbulent diffusion coefficient	m^2 s^{-1}
D_{eff}	Effective diffusion coefficient of porous medium for simulation	m^2 s^{-1}
E	Radon exhalation rate	Bq m^{-2} s^{-1}
E_a	Initial radon exhalation rate (equivalent to analytical solution in diffusion)	Bq m^{-2} s^{-1}

Symbol	Description	Units
E_f	Fitted radon exhalation rate	Bq m^{-2} s^{-1}
E_{f-exp}	Radon exhalation rate for exponential fitting	Bq m^{-2} s^{-1}
E_{f-lin}	Radon exhalation rate for linear fitting	Bq m^{-2} s^{-1}
E_n	Transient radon exhalation rate ($E_{n-d} + E_{n-c}$)	Bq m^{-2} s^{-1}
E_{n-d}	Transient radon exhalation rate for diffusion	Bq m^{-2} s^{-1}
E_{n-c}	Transient radon exhalation rate for convection	Bq m^{-2} s^{-1}
E_{ref}	Reference radon exhalation rate	Bq m^{-2} s^{-1}
E_{Rn}	Radon emanation coefficient of porous medium	dimensionless
FVM	Finite Volume Method (discretization method for numerically solving PDEs)	-
$\mathbf{g}$	Gravitational acceleration	m s^{-2}
H	Insertion depth of accumulation chamber into porous medium	cm
k	Turbulent kinetic energy	m^2 s^{-2}
K	Permeability	m^2
NUI	None Uniformity Index	dimensionless
R	Reference model for calculating reference radon exhalation rate	-
RB1/RB2	Scenarios for measuring reference radon exhalation rate (35 kg radium-containing medium, 7 cm height on RB1; 70 kg, 14 cm on RB2)	-
S_a	Surface area where radon exhales	m^2
S_{Rn}	Source term in radon migration governing equation	Bq m^{-3} s^{-1}
S_{out}	Area of flow-out tube outlet surface	m^2
Sc_t	Turbulent Schmidt number	dimensionless
t	Time	s
V	Volume of accumulation chamber	m^3
$\mathbf{v}$	Physical velocity of gas in porous medium	m s^{-1}
x	Vertical depth of porous medium	m
α	Free radon production rate	Bq m^{-3} s^{-1}
ε	Turbulent dissipation rate	m^2 s^{-3}
η	Porosity	%

Symbol	Description	Units
λ_{Rn}	Radon decay constant (2.1$×10^{-6}$ s^{-1})	s^{-1}
λ_b	Back-diffusion rate	s^{-1}
λ_e	Effective decay constant ($\lambda_{Rn} + \lambda_b + \lambda_l$)	s^{-1}
λ_l	Leakage rate for radon in accumulation chamber	s^{-1}
μ	Dynamic viscosity	Pa s
μ_t	Turbulent viscosity	Pa s
ρ_s	Porous medium density	kg m^{-3}
ρ_a	Air density	kg m^{-3}
τ	Tortuosity factor	dimensionless

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Note: Figure translations are in progress. See original paper for figures.