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Gradient chamber for measuring the exhalation rate of radon and thoron from surfaces

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A novel instrument (gradient chamber) and method for measuring the exhalation rates of radon (^{222}Rn) and thoron (^{220}Rn) from surfaces (e.g., soil, rocks, building materials) have been developed and tested. Determining radon and thoron exhalation rates is essential both for protecting human health from radon/thoron exposure and for providing valuable data relevant to climate change research. The chamber is cylindrical with open bases: the upper base is covered with a porous material that blocks atmospheric turbulence while remaining permeable to radon and thoron, and the lower base is placed in direct contact with the test surface. Within the chamber, radon and thoron activity concentrations form a diffusion profile, measured by detectors for alpha particles, installed at different heights inside the chamber walls. Each detector comprises a Kodak Pathe LR-115/II alpha track detector covered by activated carbon fabric (ACC-5092-10), which efficiently adsorbs radon and thoron. The detectors are compact (≈ 1 cm) and locally sensitive, enabling high-resolution profiling of radon and thoron distributions. Exhalation rates are determined from the diffusion gradient using Fick's law. Results from theoretical modeling and pilot field experiments confirm the high sensitivity of this method, sufficient to quantify even low environmental radon exhalation rates from the earth's surface.

Keywords Radon, Thoron, Exhalation measurement, Gradient chamber, Environment

Radon (^{222}Rn) is a radioactive noble gas and an isotope belonging to the ^{238}U decay series. It is generated by the decay of ^{226}Ra and gives rise to the so-called *radon progeny* (^{218}Po, ^{214}Pb, and ^{214}Bi + ^{214}Po, with isotope ^{214}Po always being in equilibrium with ^{214}Bi). Exposure to radon and its progeny is a recognized radiological risk factor, classified in 1988 by the International Agency for Research on Cancer as a proven carcinogen[1]. It is one of the leading causes of lung cancer, ranked by the World Health Organization[2] as the second leading cause after smoking. Thoron (^{220}Rn), an isotope of the ^{232}Th decay series, also poses a potential health risk. Exposure to thoron and its progeny (^{216}Po, ^{212}Pb, ^{212}Bi + ^{212}Po/^{208}Tl) is generally considered of secondary importance compared to radon exposure, although certain regions have been identified where thoron concentrations may be significant[3].

The major sources of radon and thoron in both indoor and outdoor environments are their exhalation from the ground surface (soil) and from building materials. Consequently, the direct determination of the *free radon/thoron exhalation rate*—defined as the activity released per unit surface area per unit time under natural, undisturbed conditions—is critically important for several applications, including:

- **Delineation of radon priority areas (RPAs)**[4]. According to the European Council Directive EURATOM 59/2013[5], Member States are required to establish reference levels for the annual average ^{222}Rn activity concentration in dwellings and workplaces not exceeding 300 Bq m^{-3}, and to identify RPAs where a significant proportion of dwellings exceed this reference level.
- **Monitoring of greenhouse gas emissions**[6,7], which provides important insights into climate change processes[8,9].
- **Geophysical applications**, where radon exhalation is used for fault identification[10] and seismic research[11], and is considered a potential earthquake precursor[12].
- **Environmental management** in areas affected by uranium mining and milling[13,14].
- **Radiation protection**, particularly regarding building materials[15–17], which represent the second most important indoor source of radon, after exhalation from the ground.

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Despite its importance, the assessment of radon (and thoron) exhalation rates from various surfaces (e.g., soil, building materials) remains one of the major challenges in radon/thoron measurement and metrology. It should be noted that thoron exhalation measurements are still in their infancy, with much less data available compared to radon. In the present work, the primary focus is on radon.

The earliest methods for determining radon exhalation rate involved monitoring the accumulation of these gases within a confined volume placed over the test surface[18,19]. Other approaches measured the activity of radon accumulated in the enclosed volume after a given time interval[20]. For material samples (e.g., building materials), the sample is sealed in a container, and the accumulation of radon or thoron within this closed environment is tracked over time[21,22]. However, other studies[23,24] have shown that such accumulation methods alter the radon concentration profile near the surface of the material due to the back diffusion, leading to significant underestimation of the true free exhalation rate. Experimental confirmation of this effect has been reported[25]. As demonstrated in[23], the profile reshaping occurs rapidly and can lead to substantial underestimation.

Another method relies on constructing a radon concentration profile down to 60–100 cm below the surface and fitting it to a diffusion model, to estimate radon exhalation-rate[26,27]. This technique requires soil gas sampling at various depths, embedding radon-adsorbing samples, or burying detectors to measure radon activity at depth[27]. It assumes homogeneous distribution of the radon source (^{226}Ra) and a uniform diffusion coefficient in the soil. In practice, however, both the radon source distribution and diffusion properties are often highly variable[25,27,28]. These variations lead to deviations from the theoretical diffusion profile and introduce systematic errors. In addition, inserting probes or detectors into the soil disturbs the natural vertical concentration profile. As a result, in intercomparisons considerable discrepancies, sometimes exceeding an order of magnitude, are observed between results from different laboratories and methods[29–31], highlighting the lack of sufficiently accurate and standardized radon exhalation rate measurement techniques.

Although the above-mentioned methods measure only radon exhalation, the growing interest in thoron in recent years highlights the need for methods and instruments capable of accurately measuring both radon and thoron exhalation rates from surfaces into the air, with high sensitivity and reliability. To address thoron, a ventilated accumulation chamber has been proposed[32]. In this instrument, the accumulation volume is naturally ventilated to enable the independent determination of radon and thoron exhalation. Air ventilation is achieved through two rectangular openings, covered by filters, perforated at different heights on the chamber walls[32]. While radon is homogeneously distributed throughout the ventilated volume, thoron concentration decreases with increasing distance from the ground[32]. To determine the thoron exhalation rate, signals from thoron detectors placed at different heights above the ground are fitted to estimate the thoron concentration at the ground surface. This concentration is then used to calculate the thoron exhalation rate[32].

In recent years, compact and highly sensitive radon detectors with *local sensitivity* have been developed[33,34]. These include devices that combine a highly efficient radon adsorbent—activated carbon fabric—with a solid-state nuclear track detector (SSNTD)[33,35,36]. A specific fabric that maintains high adsorption capacity even under high humidity conditions has been identified (ACC-5092-10, Kynol Europa GmbH, Germany)[37,38]. Due to the high adsorption capacity of the adsorbent material, radon is concentrated within the volume by several orders of magnitude compared to its concentration in ambient air. As a result, even low environmental radon concentrations can generate a measurable signal in the detectors. Sensitivity assessments have shown that the minimum detectable radon activity concentration with such detectors can surpass that of conventional state-of-the art radon detectors by one to two orders of magnitude[34,39]. Owing to their small size (~ 1 cm or less), these detectors possess *local sensitivity*, enabling studies of fine-scale spatial variations in radon concentration—for example, near the soil–air interface[25] or in identifying unusual indoor radon ingress pathways[40].

This new generation of radon detectors has opened the way to developing a conceptually new method for measuring radon and thoron exhalation from surfaces. In this work, a novel instrument, the *gradient chamber*, is described, and a method for measuring the free exhalation rates of radon and thoron using this device is developed. To test the feasibility of the method, pilot field experiments were conducted. The preliminary data demonstrate a level of sensitivity sufficient for measuring radon exhalation rates across the full range encountered in natural environments[41]. The proposed method may be less affected by the limitations of previously used techniques, as the gradient chamber volume is open (Fig. 1) and radon cannot reach the concentrations observed in confined accumulation volumes.

Materials and methods

Concept

The method employs conceptually new instrument, the *gradient chamber* (patent pending[42]), schematically illustrated in Fig. 1. This chamber is designed for measuring the exhalation rates of radon and thoron from surfaces[42]. It is equipped with the new-generation detectors described above, which exhibit high local sensitivity and can be selectively responsive to either radon or thoron[42].

The lower base of the chamber is open to ensure direct contact with the tested surface. The upper base is covered with a porous, low-resistance material (e.g., an aerosol filter or cloth) that allows radon and thoron to diffuse through while damping the effects of atmospheric turbulence within the chamber volume. As a result, a stable diffusion profile of radon and thoron activity concentrations is established along the chamber's height. When the device is used outdoors, a protective rain cap covers the porous layer to prevent it from becoming wet, without impeding air circulation.

The chamber houses vertically aligned detectors mounted on its inner wall. These detectors are "composite" and incorporate an alpha-particle detector whose sensitive surface is coated with a highly efficient radon adsorbent that also serves as a radiator. In the present experiments, the adsorbent consisted of a strip of activated carbon fabric (type ACC-5092-10; see Fig. 1). The detectors register alpha particles emitted by the adsorbed radon and thoron and their respective decay products, being protected from the deposition of radon/thoron progeny in air

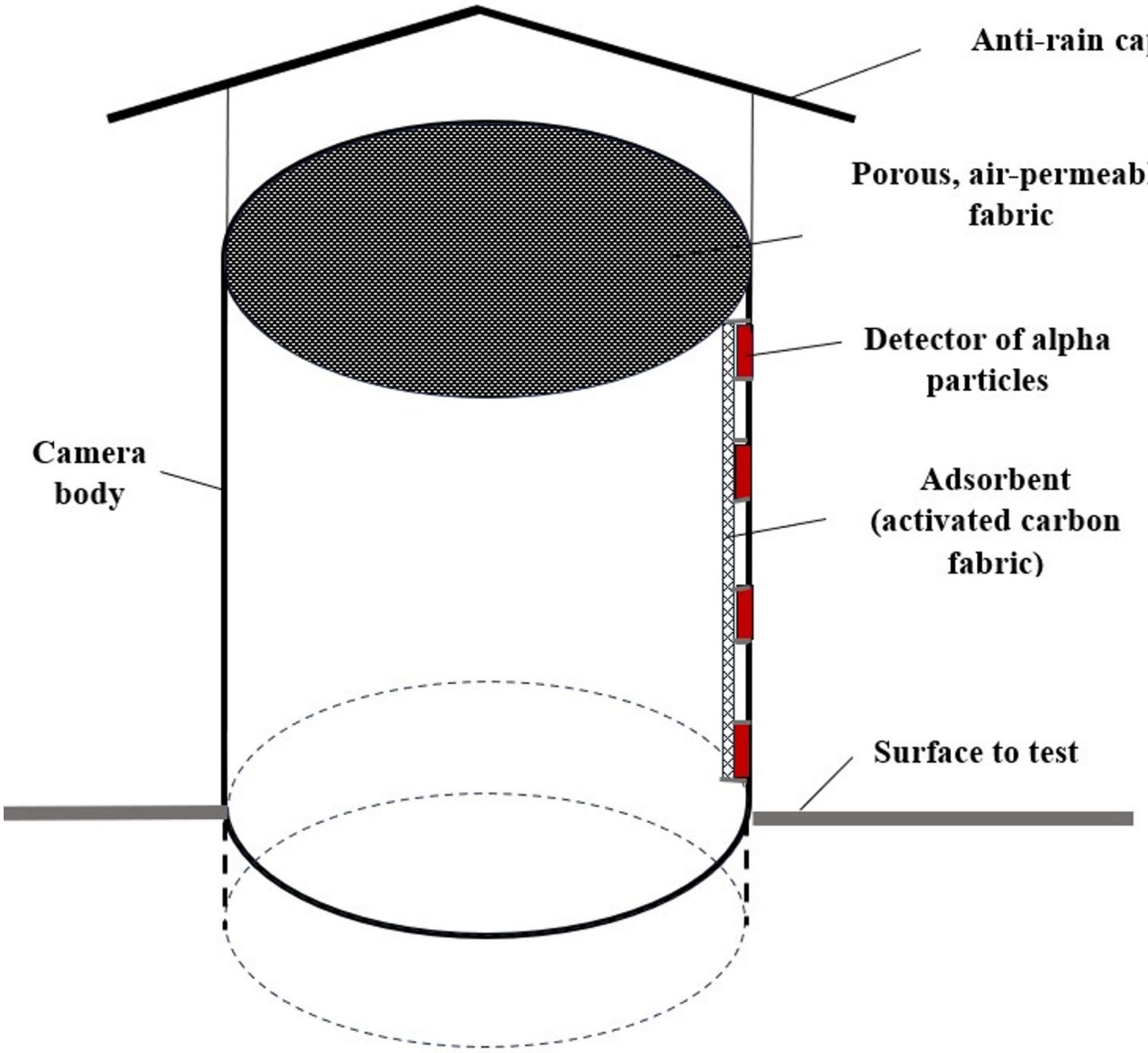


Fig. 1. Design of the gradient chamber. When used on a loose surface (soil), the open base is inserted 2–3 cm deep. On a hard surface, the open base is placed directly on the surface, and its perimeter is sealed appropriately (with silicone or similar material).

on their surface by covering them by adsorbent layer that is thicker than the range of alpha particles of radon/thoron progeny deposited on the outer, looking to the chamber volume, surface of the adsorbent.

Within the chamber, the activity concentration of radon and thoron naturally forms a vertical diffusion profile. This allows the average exhalation rate over the exposure period to be determined from the concentration levels measured at different heights above the surface. Importantly, because the chamber volume is not completely enclosed, the radon concentration inside does not substantially exceed that of the surrounding air near the surface and therefore are not expected to significantly alter the natural radon concentration profile in the tested soil or material.

According to Fick's law, the exhalation rate $\in_i$ of radon or thoron from a flat surface into air can be expressed as:

$$\in_i = -D.gradc_{Ai}(x)\big|_{x=0} = -D\left.\frac{dc_{Ai}}{dx}\right|_{x=0} \quad (1)$$

where the subscript $i=1$ refers to radon and $i=2$ to thoron. The quantities c_{A_i} denote the corresponding activity concentrations in the air inside the chamber, expressed as functions of height x above the surface. The parameters $\in_i$ represent the respective exhalation rates, and D is the diffusion coefficient of radon or thoron in air. Since both isotopes diffuse similarly in air, a single value of D applies to both. In the present work, the following value

was used: $D = 0.105\,\mathrm{cm}^2\,s^{-1} = 1.05 \times 10^{-5}\,\mathrm{m}^2\,s^{-1}$ [43]. Unlike the diffusion coefficient of radon in soil or solid materials, which varies considerably depending on composition, porosity, and environmental conditions[25], the diffusion coefficient of radon in air is well established and minimally influenced by meteorological factors.

The equilibrium distribution of radon and thoron within the chamber can be described by the steady-state solution of the diffusion equation, which accounts for radioactive decay:

$$D\frac{d^2c_{Ai}}{dx^2} - \lambda_i c_{Ai} = 0 \tag{2}$$

where λ_i are the respective decay constants. This equation is considered together with two boundary conditions treated as unknown parameters: the average exhalation rate ($\in_i$) from the surface and the average activity concentration at the upper end of the chamber (c_{0i}), which may approximate or equal the ambient activity concentration. These parameters can be determined through mathematical analysis of the signals recorded by the detectors at different heights. Equation (2), together with the specified boundary conditions, was solved analytically. The solution, derived using well-established mathematical methods, provides the activity concentration at a height x above the surface tested as:

$$c_{Ai}(x) = \in_i \left[\frac{\exp\left(\frac{H}{L_{Di}}\right) exp\left(-\frac{x}{L_{Di}}\right) - \exp\left(\frac{-H}{L_{Di}}\right) exp\left(\frac{x}{L_{Di}}\right)}{\lambda_i L_{Di}\left(\exp\left(\frac{H}{L_{Di}}\right) + exp\left(-\frac{H}{L_{Di}}\right)\right)}\right] + c_{0i}\left[\frac{\exp\left(-\frac{x}{L_{Di}}\right) + exp\left(\frac{x}{L_{Di}}\right)}{\exp\left(\frac{H}{L_{Di}}\right) + exp\left(-\frac{H}{L_{Di}}\right)}\right] = \in_i A_{i1}(x) + c_{0i}A_{i2}(x) \tag{3}$$

In this expression, H denotes the height of the chamber measured from the tested surface, and L_{D_i} is the diffusion length in air for each isotope, defined as $L_{D_i} = \sqrt{D/\lambda_i}$, with $L_{D1} = 223\mathrm{cm}$for radon and $L_{D2} = 2.9\mathrm{cm}$for thoron. The terms within the square brackets (labeled here as $A_{i1}$and $A_{i2}$for simplicity) are known numerical expressions, calculated from the relevant parameters.

For chamber heights not exceeding 20 cm, the dependence given in Eq. (3) for radon can be approximated linearly with an accuracy better than 1%. This simplification allows the use of Eq. (3) in a form that facilitates more straightforward mathematical processing of the results:

$$c_{A1}(x) = \in_1 \left(\frac{H}{D} - \frac{x}{D}\right) + c_{01} = -\frac{\in_1}{D}x + \frac{\in_1 H}{D} + c_{01} \tag{4}$$

In this case, the gradient of radon activity concentration along the chamber height remains practically constant and is represented by the coefficient of the variable x in the linear dependence (Eq. 4).

Figures 2 and 3 illustrate sample distributions obtained from model calculations based on Eq. (3) for radon and thoron activity concentrations within a chamber of height $H = 10\mathrm{cm}$. The signal recorded by a detector

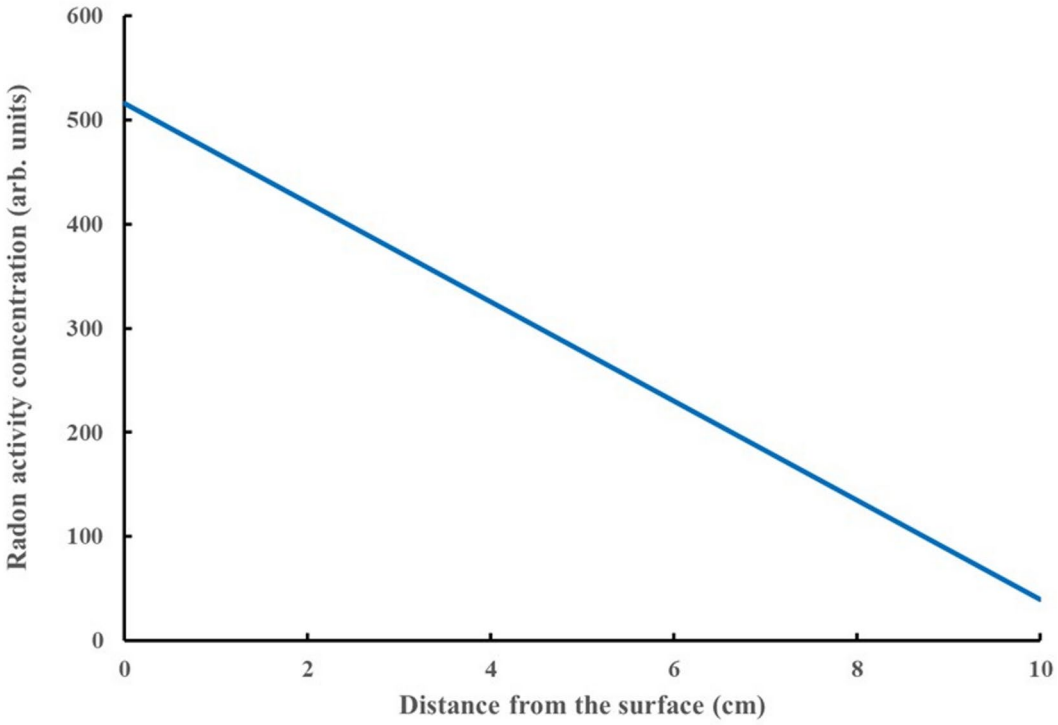


Fig. 2. Model distribution of radon activity concentration along the height of the gradient chamber (H = 10 cm).

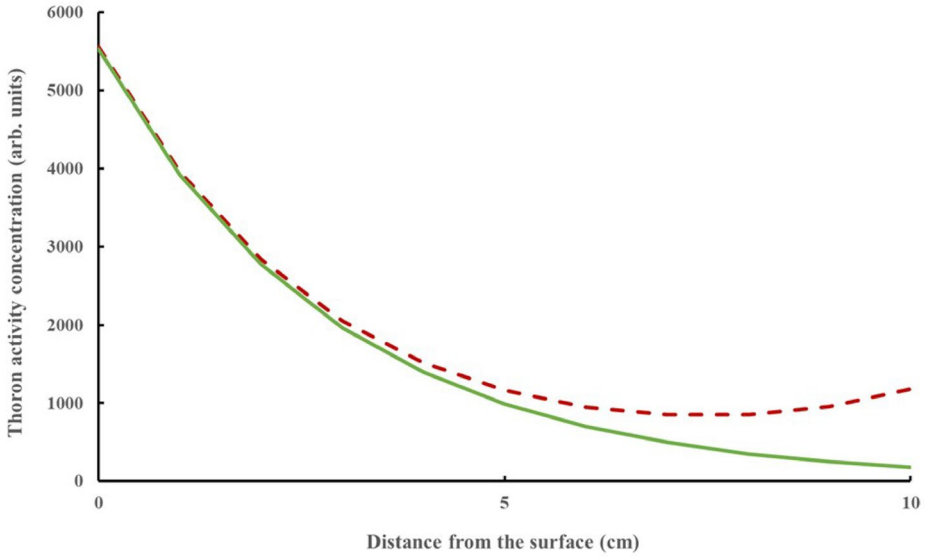


Fig. 3. Model distribution of thoron activity concentration along the height of the gradient chamber ($H = 10$ cm). The solid line is when the ambient thoron concentration is low, while the dashed one is for the case of high ambient thoron concentration.

positioned at height x above the investigated surface, corresponding to the i-th isotope, can be expressed as: $S_i(x) = CF_i\, c_{A_i}(x)\, T$, where $T$is the exposure duration during which the detectors accumulate the signal, c_{A_i} is given by Eq. (3) (or Eq. (4) for radon), and CF_i is the calibration factor for the i-th isotope. The calibration factor, specific to radon or thoron, represents the ratio of the accumulated detector signal over the exposure time to the product of the isotope's activity concentration in air and the exposure duration.

When the detectors simultaneously register signals from both radon and thoron, the signal recorded by a detector positioned at height x_i above the tested surface can be expressed as:

$$S(x) = S_1(x) + S_2(x) = CF_1 T c_{A1}(x) + CF_2 T c_{A2}(x) = \in_1 CF_1 TA_{11}(x) + c_{01} CF_1 TA_{12}(x) + \in_2 CF_2 TA_{21}(x) + c_{02} CF_2 TA_{22}(x), \quad (5)$$

where the coefficients A_{jk} $(j,k = 1,2)$ are defined according to Eq. (3). This configuration allows the determination of the four unknowns: $\in_1, \in_2, c_{01}, c_{02}$, provided that signals from detectors positioned at a minimum of four different heights are recorded. These values are obtained as solutions to a system of four linear equations with four unknowns, constructed by substituting the numerical values of the known coefficients and chamber dimensions into Eq. (3) (or Eq. (4) for radon) for each of the four detectors at their respective heights x_i. When more than four detectors are employed, the solutions can be refined using standard regression analysis techniques.

If detectors selectively sensitive to either radon or thoron are used, the signal from a radon-specific detector positioned at height x above the investigated surface can be expressed as:

$$S(x) = S_1(x) = CF_1 T c_{A1}(x) = \in_1 CF_1 TA_{11} + c_{01} CF_1 TA_{12} \quad (6)$$

and from one sensitive only to thoron:

$$S(x) = S_2(x) = CF_2 T c_{A2}(x) = \in_2 CF_2 TA_{21} + c_{02} CF_2 TA_{22} \quad (7)$$

When two detectors of each type and positioned at different heights are used (two sensitive to radon and two sensitive to thoron), the desired quantities can be determined by solving two independent systems of equations, each consisting of two linear equations with two unknowns. These equations are derived from Eqs. (6) and (7) and can be solved using standard methods. If more than two detectors are used for one or both isotopes, regression analysis techniques can be applied to refine the results. When only one isotope—radon or thoron—is of interest, two detectors selectively sensitive to that isotope may suffice.

On Figs. 2 and 3 it might be seen that the vertical distributions of radon and thoron in chamber differ significantly. While that of radon is almost perfectly linear, that of thoron is markedly non-linear. In the illustrated case at height 5 cm radon concentration decreases twice, while that of thoron by a factor of 6. Thus statistically significant deviation above the linearity of experimental points at small heights is considered as indication of thoron presence and data processing should account also for thoron.

Results

Practical implementation

The detectors respond to the local radon and thoron concentrations at their respective positions. Each such detector comprises a small alpha-particle detector (1–2 cm^2) coated with a highly effective adsorbent, ACC-5092-10, which maintains high radon adsorption capacity even when saturated with water[38]. Radon activity concentrations in such efficient adsorbents are thousands of times higher than in the surrounding air[35], accounting for the high sensitivity of these detectors and enabling accurate measurements of low radon and thoron levels, even outdoors[34].

The adsorbent layer is thicker than the range of the highest-energy alpha particles associated with radon and thoron, ensuring that the alpha detector registers only alpha particles emitted by the adsorbed radon/thoron and their progeny. In our experiments, the ACC-5092-10 fabric (~ 24 mg cm^{-2}) satisfied this condition. Because the adsorbed radon and thoron activity is proportional to their local concentrations in the air adjacent to the adsorbent, the detector signal directly reflects the local radon/thoron concentrations.

The chamber used in this study had a height of 12 cm, well below the 223 cm diffusion length of radon in air, allowing equilibrium to be established within approximately one hour. The chamber's lower base was open to the surface and sealed by driving it 2 cm into the soil, giving an effective height of $H = 10$ cm. The soil around the chamber edges was compacted to ensure a tight seal, such that radon and thoron could enter the chamber only via exhalation from the surface.

The SSNTDs employed in present experiments were Kodak Pathé LR-115/II coupled with ACC-5092-10 fabric. Each detector was individually calibrated at various temperatures and humidity levels as described elsewhere[33,34,38] using the facility detailed in[44]. Experimental testing established that these detectors are significantly more sensitive to radon than to thoron[40]. During data processing, calibration factors adjusted for the average temperature and humidity during exposure were applied.

Ten solid-state nuclear track detectors (Kodak Pathé LR-115/II, 1.5 cm × 1 cm) were installed on the chamber walls (Fig. 1). The first detector was positioned such that, after the chamber base was driven 2 cm into the soil, its center was 0.5 cm above the surface. Subsequent detectors were centered at heights of 1.5 cm, 2.5 cm, and so on, up to the tenth detector at 9.5 cm, directly beneath the fiber aerosol filter at the upper base (H = 10 cm)[42]. After exposure, the SSNTDs were removed and subjected to chemical etching using 10% NaOH at 60 °C for 100 min, followed by a 30-minute rinse in agitated water and a 2-minute rinse in still 50% ethanol. After drying, alpha tracks were counted visually under a microscope. The detector signal, expressed as net track density (tracks per unit area, background subtracted), was determined from these measurements.

Experiment 1

The first test was conducted on a soil surface in the city of Sofia, Bulgaria. The chamber was deployed on soil in the field and left for an exposure period of 283.6 h. The exposure was made wintertime (second half of January) under wet and unstable weather conditions characterized by wind and atmospheric turbulence. The average temperature during this exposure was 0 °C. After exposure, the detectors were processed, and signals from each detector were determined. Within experimental uncertainties, the relationship between signal and distance from the soil surface was found to be linear (Fig. 4), with no measurements at small heights significantly above the line that could be attributed to thoron. This allowed the data to be processed considering radon only, for which the linear dependence was confirmed with high accuracy. Average radon activity concentrations were then calculated for the different detector heights (left vertical axis in Fig. 4).

Standard linear regression analysis was performed using TableCurve 2D, version 5.0 software to determine the slope of the line approximating the relationship between radon activity concentration and the height of the detector center. This slope represents the vertical gradient of radon activity concentration, which value and its uncertainty were obtained by standard linear regression analysis and found to be $\mathbf{586 \pm 31\ Bq\ m^{-4}}$. By multiplying the absolute value of this slope by the diffusion coefficient of radon in air (see Eq. (4)), the average radon exhalation rate from the soil over the exposure period was calculated as $\mathbf{6.15 \pm 0.33\ mBq\ m^{-2} \cdot s^{-1}}$, equivalent to $\mathbf{22.2 \pm 1.2\ Bq\ m^{-2} \cdot h^{-1}}$.

According to the European Atlas of Natural Radiation[41], this value corresponds to a relatively low soil exhalation rate. This result underscores the high sensitivity of the proposed gradient chamber, which can accurately measure radon exhalation across the full range of environmentally relevant values.

Experiment 2

The second test was conducted using the same gradient chamber as in Experiment 1, but in terrain heavily affected by the uranium milling industry. Large areas in this region are significantly contaminated with ^{226}Ra[28], resulting in very high radon exhalation from the soil[27]. The test was made during springtime (end of April) in sunny and stable weather, without wind and at temperature 24 °C. The exposure duration was 3 h. After exposure, the gradient chamber was left for one day at low outdoor radon concentrations (< 20 Bq m^{-3}) to allow the adsorbed radon to desorb/decay and its progeny to decay. Such precautions are not necessary, when the exposure time is long (days/weeks).

In this experiment, a parallel measurement was carried out using the accumulation method (Fig. 5) with a radon/thoron monitor (AlphaGUARD PQ 2000 PRO Rn/Tn, Berthin GmbH, Germany), equipped with the standard accumulation chamber supplied with the monitoring system. The system of Fig. 5 is of total circuit volume V = 0.035 m^3 and area of the open base of the chamber that contacts with the ground S_b=0.21 m^2. The instrument was operated in the $^{222}Rn/^{220}Rn$ 10 min flow mode, with air continuously circulated through the accumulation chamber, pump, and detector (Fig. 5). No ^{220}Rn exhalation from the test surface was observed, as the monitor did not register any measurable thoron activity during the measurement period.

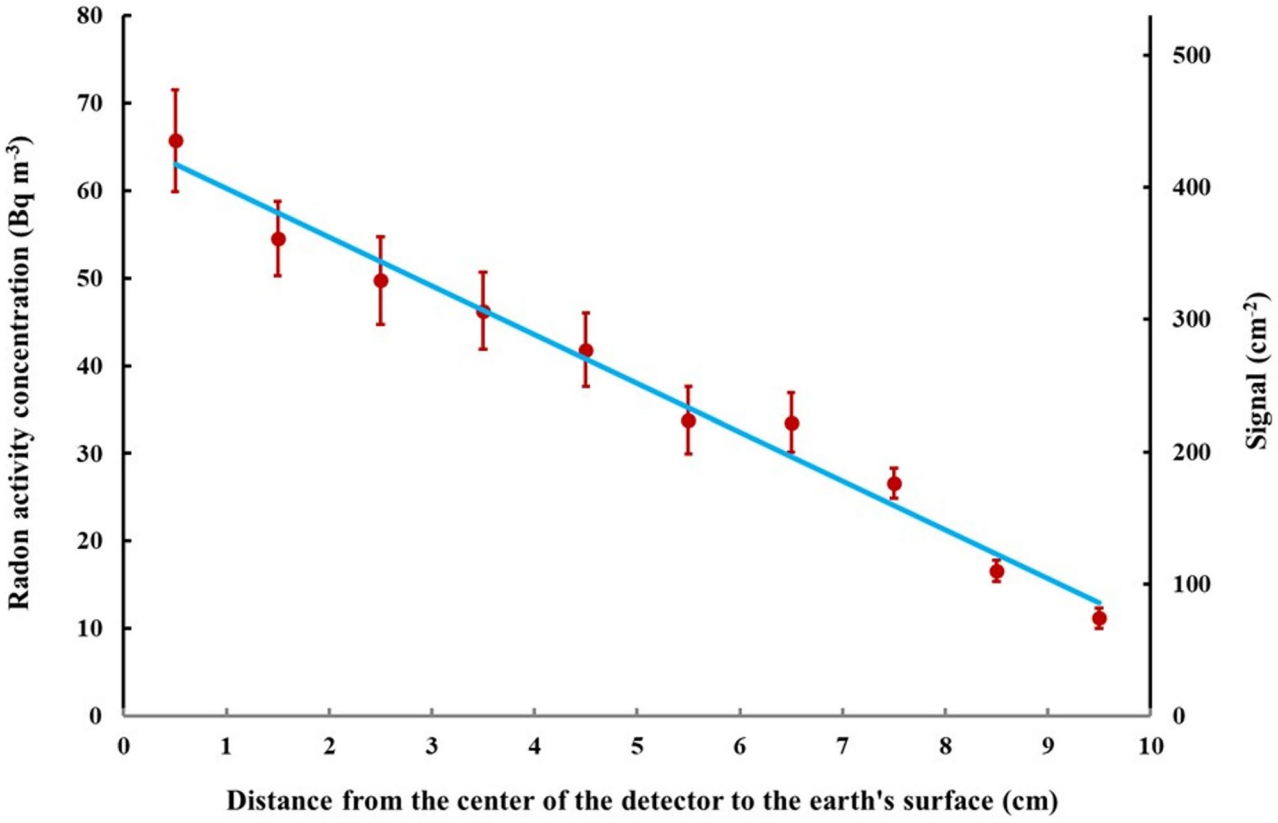


Fig. 4. Experimental results obtained in experiment 1. Error bars indicate one standard deviation, considering counting statistics and detectors' calibration uncertainty. The line is the fit of the experimental points, according to Eqs. 3 and 4. No deviation from linearity can be seen.

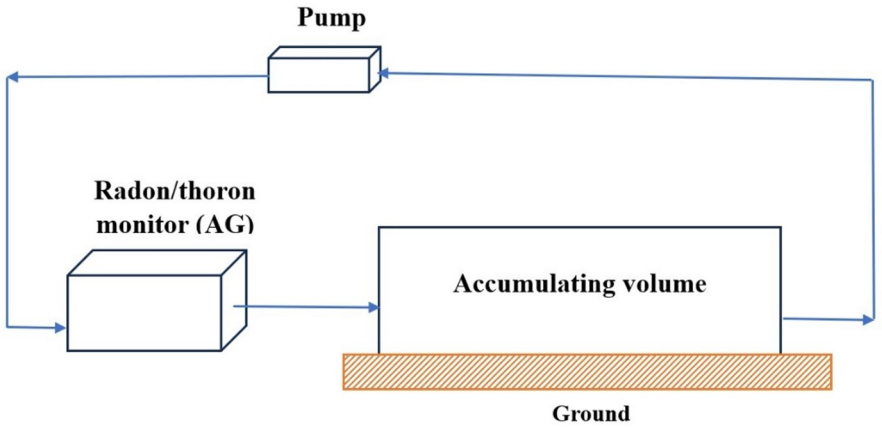


Fig. 5. The scheme of parallel measurement by the accumulation method in experiment 2.

The increase in ^{222}Rn activity concentration within the accumulation volume was monitored for 3 h after placing the accumulation box over the tested surface. The exhalation rate was then calculated from the initial growth rate of radon activity concentration, using the established method described elsewhere[21]. ($\in$ = initial growth rate multiplied by V/S_b).

Simultaneously, the gradient chamber was positioned approximately 1 m from the accumulation box, and both devices were exposed for 3 h. The experimental results obtained with the gradient chamber are shown in Fig. 6. As observed, similar to Experiment 1, the dependence of the detector signal on height is nearly perfectly linear, with no indication of nonlinearity that might suggest the presence of thoron (see Fig. 3). The slope of the line, corresponding to the vertical gradient of radon activity concentration, was determined using the same procedure as in Experiment 1.

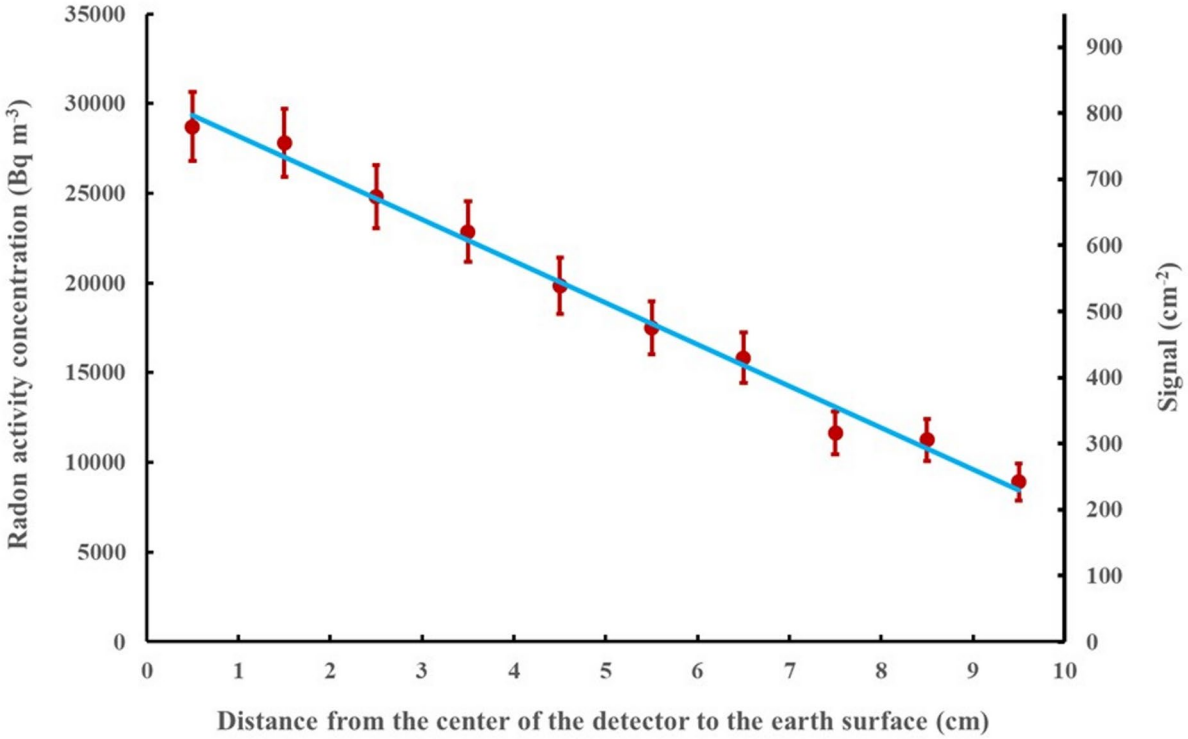


Fig. 6. Experimental results obtained in experiment 2. Error bars indicate one standard deviation, considering counting statistics and detectors' calibration uncertainty. The line is the fit of the experimental points, according to Eqs. (3, 4). No deviation from linearity can be seen.

The results with the gradient chamber were as follows:

- **Gradient (line slope, obtained, together with its uncertainty by linear regression as in Experiment 1)**: 271.6 ± 10.2 kBq m^{-4}.
- **Exhalation rate**: 10.27 ± 0.39 kBq m^{-2} h^{-1} = 2.85 ± 0.11 Bq m^{-2} s^{-1}.

The ingrowth of ^{222}Rn obtained using the parallel accumulation method is shown in Fig. 7. The radon exhalation rate determined by this method was **6.02 ± 0.22 kBq $m^{-2} \cdot h^{-1}$ = 1.67 ± 0.06 Bq $m^{-2} \cdot s^{-1}$**. This value is about 60% of the value obtained by the gradient chamber, the difference being statistically significant.

In contrast to experiment 1, the measured value of exhalation rate in experiment 2 appears to be very high.

Discussion

The novel approach for determining free radon and thoron exhalation rates from surfaces has demonstrated potential for practical use. The method employs a gradient chamber, whose upper open base is covered with a porous material (filter) to protect it from atmospheric turbulence. The effectiveness of this protection was demonstrated in Experiment 1, conducted under highly windy conditions. Any air turbulence entering the chamber would disrupt the diffusion profile of radon within. The fact that the observed distribution perfectly matches the theoretical model of purely diffusional radon transport provides evidence that radon transport is governed solely by diffusion, with no turbulent contribution. The method appears capable of covering the full range of exhalation rates reported for Europe[41,45].

Comparison with parallel measurements by the accumulation method using AlphaGUARD monitor and its accumulation box revealed that there is a statistically significant difference between exhalation rate measured by the accumulation method and that obtained with the gradient chamber, the "accumulation method value" being about 60% of that provided by gradient chamber. Two possible reasons for such discrepancy might be identified. The first reason is that reshaping radon concentration profile by a confined accumulation volume over the surface leads to reducing the observed exhalation. The second reason might be due to natural variability in exhalation rates at the two measurement locations. Even at a distance of only 1 m, our experience shows that soil gas radon levels and, consequently, radon exhalation rates can vary significantly. Our data is insufficient to distinguish between these two causes—both may play a role. Additional field comparisons are planned to determine whether the accumulation method systematically reports lower exhalation rates than the gradient chamber and to evaluate the source of any discrepancies.

Measurements in the accumulation box did not detect thoron, and similarly, the gradient chamber showed no measurable thoron exhalation at this site. However, interest in thoron has been increasing in recent years. This interest lagged behind that for radon, possibly because thoron is considered of secondary radiological

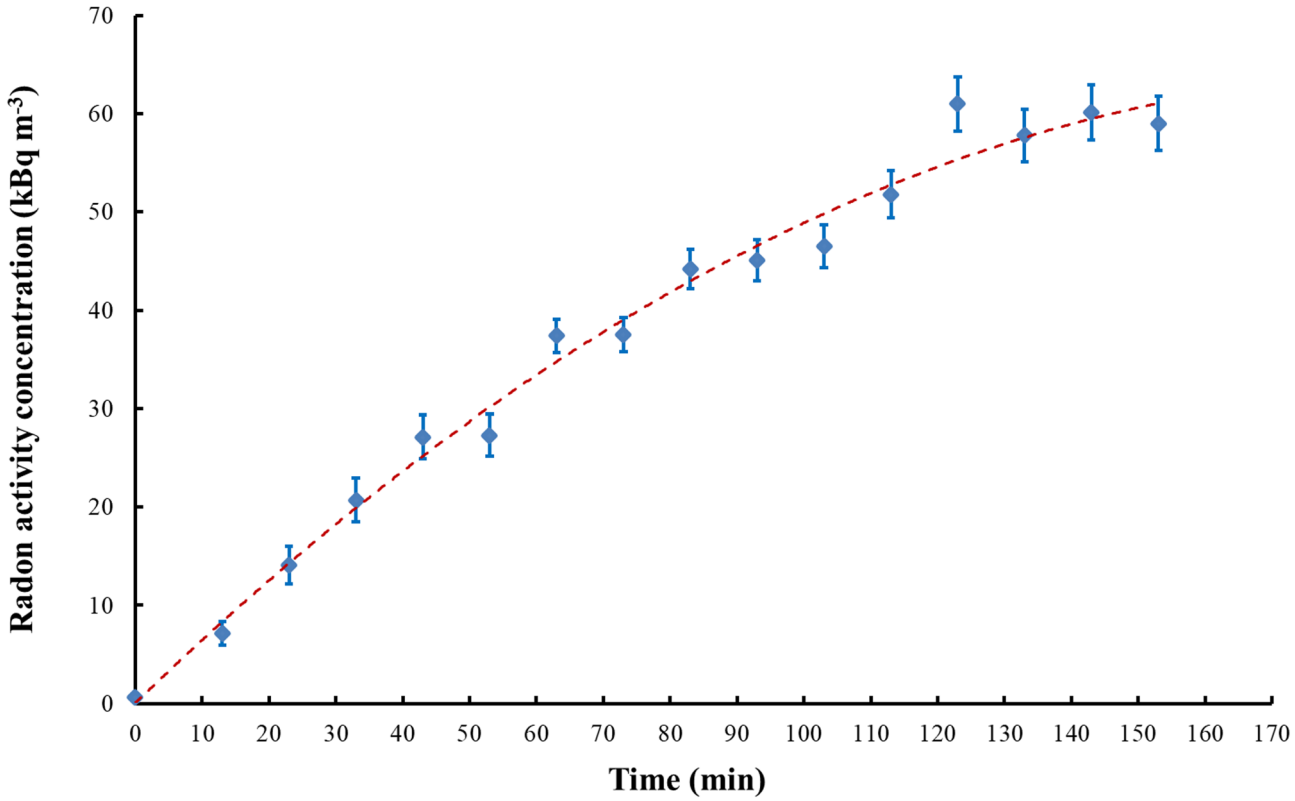


Fig. 7. The ingrowth of ^{222}Rn in the accumulation volume in the parallel measurement in experiment 2.

significance compared with radon. Research and measurements are also driven by legislative requirements, which exist for radon in the European Union and many other countries, but not for thoron.

It would be interesting to compare the gradient chamber with the method of ventilated accumulation volumes[32]. Both methods allow simultaneous measurement of radon and thoron exhalation rates, but they operate differently. In the gradient chamber, diffusion profiles of both radon and thoron concentrations are established and used to determine exhalation rates. In the method described in[32], the chamber volume is naturally ventilated, and the independently determined air exchange rate is a required parameter for calculating radon exhalation, with radon being homogeneously distributed within the accumulation volume[32]. Thoron concentrations, however, decrease with increasing distance from the ground, and data from thoron detectors placed at different heights are fitted to estimate the thoron concentration at the ground surface, which is then used to calculate the thoron exhalation rate, with the air exchange rate also serving as a required parameter. The accumulation volume in this method is relatively large ($0.8 \times 0.8 \times 0.85$ m) and is embedded 15 cm into the ground, whereas gradient chambers can be very compact, with diameters and heights on the order of centimeters and embedded only 2–3 cm into the ground. This makes gradient chambers more convenient for screening multiple points over a large area.

The experiments reported here were conducted with passive track detectors. However, small active alpha-particle detectors could be easily integrated into this method. Using surface-barrier detectors or PIN-photodiodes could additionally allow spectral analysis, enabling differentiation between radon and thoron signals. In such a case, the number of detectors required for implementation could be reduced to two. Active detectors would also allow time-resolved monitoring of radon/thoron exhalation with sub-hour resolution. Given that equilibrium in a chamber of height ~ 10 cm is reached in less than an hour, the method has the potential for high temporal resolution when active detectors are employed.

Although the tests reported here were performed on soil, the gradient chamber can also be applied to solid surfaces such as building materials, floors, or walls. In these cases, the contact line of the chamber's open base should be sealed with silicone or another suitable sealant. Such measurements may be nondestructive, as sample extraction is not required.

At this stage, variations in the radon diffusion coefficient in air due to environmental conditions were not considered, as data on radon diffusion at different temperatures and pressures are largely absent in the scientific and technical literature. It is known that gas diffusion at low concentrations depends on environmental conditions, particularly temperature and pressure, following approximately $D \propto T^{3/2}/P$, where T is absolute temperature and P is pressure[46]. Indirect estimates based on this relationship suggest that if the temperature changes from 0 to 30 °C, the diffusion coefficient may vary by ~ 17%. Such variation is non-negligible and highlights the need for experimental studies to determine radon diffusion coefficients in air under varying temperatures, improving the accuracy of current estimates.

Certain limitations of the method can be anticipated. For example, whether the use of a porous, air-permeable fabric to cover the upper base of the chamber (Fig. 1) provides sufficient protection against external air turbulence. If exterior wind penetrates the chamber, it could disrupt the vertical diffusive profile of radon concentrations. Notably, Experiment 1 was conducted under windy conditions with high air turbulence for most of the time. Nevertheless, the observed vertical radon concentration profile closely matches the purely diffusive model of Eq. (4), demonstrating effective anti-turbulence protection of the inner volume. Another consideration arises for measurements under rainy conditions. The anti-rain cap protects the ground beneath the chamber, keeping it drier than surrounding soil. High soil moisture can influence exhalation, particularly for thoron. However, rainwater may migrate horizontally, and when the chamber's base area is small (the vertical distribution of radon/thoron concentrations is independent of base area, see Eq. (3)), the surface soil beneath the chamber may still experience increased water content.

The influence of the gradient chamber on radon distribution in soil has not been experimentally tested but is expected to be weaker than that of closed accumulation volumes used in conventional methods. Since the chamber's upper boundary is not hermetically sealed, radon concentrations inside remain lower than in confined accumulation volumes. Future studies should evaluate potential alterations in the radon profile in the upper soil layer beneath the chamber. One possible mitigation is to use chambers with smaller heights (H), which produce lower radon concentrations near the surface than taller chambers (see Eq. (4) at $x = 0$).

Methods using ventilated accumulation volumes[32] may disturb soil radon distribution less than methods with confined volumes. However, the method[32] reports an air exchange rate of $Q = 0.3\ \mathrm{h}^{-1}$, corresponding to a mean radon "residence time" of ~ 3.3 h ($1/Q$), which is comparable to the accumulation times used in confined volumes in this and other studies[27]. Therefore, radon concentrations in both approaches may be similar under equivalent conditions.

Overall, these considerations emphasize the need for further experimental studies on radon distribution in soil beneath confined, ventilated, and gradient chambers to clarify the actual situation and evaluate potential biases in exhalation-rate estimates.

Clear advantage of the gradient chamber is its simple, compact and cost-effective design, particularly when used with passive detectors. This allows for the deployment of multiple chambers over a grid to map radon exhalation across large areas and to investigate spatial variations in exhalation even at fine scales - a task that has not yet been systematically addressed. This opens a new direction in radon research, which will require substantial effort in the coming years.

Conclusions

A novel method and instrument, the *gradient chamber*[42], for measuring radon and thoron exhalation rates is described. The theoretical framework for data processing has been developed and presented. Pilot experimental tests were conducted, and results were compared with the traditional accumulation method. Relatively good agreement was observed, particularly considering the inherent underestimation typical of accumulation methods[23,24]. The gradient chamber method might be less affected by the limitations of previous techniques. In particular, the inherent bias towards underestimations of the accumulation methods may partly explain moderate discrepancy between the two methods; in this study, where the accumulation method measured approximately 60% of the exhalation rate recorded by the gradient chamber.

The method can also be adapted to measure thoron exhalation, although no measurable thoron was detected at the tested sites. It is applicable to both soft surfaces (e.g., soil) and solid surfaces (e.g., rocks, building materials). Further studies using both passive and active detectors are planned to extend the method's capabilities, including spectral analysis and time-resolved measurements.

Data availability

The datasets used and/or analysed during the current study available from the corresponding author on reasonable request.

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Author contributions

D.P. did the research, analyzed the results and wrote the manuscript.

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Declarations

Competing interests
D.P. is the author of the following patent pending invention: Pressyanov, D. Chamber for measuring the exhalation rate of radon and thoron from surfaces. Patent application to the Bulgarian Patent Office Nr. 113878 A (assignee: Sofia university "St Kliment Ohridski", priority 04 April 2024).

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