ESTIMATION OF RADON EMANATION POWER: A COMPARISON OF DIFFERENT METHODS

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ABSTRACT

Radon emanation power was estimated by applying three different methods. The first was based on measuring radon exhalation rates by closed-loop accumulation method employing RAD7 device. Radon leakage rate was determined by applying two models of fitting the experimental data. Specific activities of ^{226}Ra in soil samples were measured by coaxial HPGe detector (GEM30-70, ORTEC). The second method was indirect gamma-ray spectrometry method which included two separate measurements of counts under the photopeaks of 351.9 keV (^{214}Pb) and 609.3 keV (^{214}Bi). The influence of sample moisture content on radon emanation was demonstrated by both methods. Radon emanation power of the sample with the highest radon exhalation rate was also estimated by two-month exposure of two radon diffusion chambers equipped with CR-39 detectors. A good agreement among the results was obtained; coefficient of variation was below 10% for the samples employed in the study. Assuming zero volumetric fraction of ^{218}Po in air provided more consistent results.

Keywords: radon, emanation power, exhalation rate, RAD7, gamma-spectrometry

1. INTRODUCTION

Exposure to high radon concentrations is associated with excess lung cancer mortality. Radon contributes up to 40% of ionizing radiation dose received by humans (WHO, 2009) and it is assumed to be responsible for about 21,000 lung cancer deaths every year (EPA 2012). Radon ^{222}Rn gas is generated by the decay of ^{226}Ra within solid grains. Most of the radon atoms produced in a material usually remain trapped inside the grains. However, a fraction of radon escapes into the pore spaces among the grains and eventually diffuses into the atmosphere. The ratio of the number of radon atoms that enter the pore spaces over the total number of atoms generated is called radon emanation power (in literature it is sometimes called radon emanation coefficient or radon emanation fraction). Radon activity released per unit mass (or unit surface area) per unit time is called radon exhalation rate. Radon exhalation rate is sometimes also referred to as radon flux.

The main sources of indoor radon are soil underneath the buildings, building materials, and groundwater supplies (Abdo et al. 2020; Ivanova et al. 2017). Soil is the dominant source of radon in most buildings and according to Nazaroff (1992), the average flux density of radon from undisturbed soil into the atmosphere was reported to be 0.015 – 0.048 Bq m^{-2} s^{-1}. Measuring the content of parent ^{226}Ra in different mediums is commonly not sufficient to predict their potential to increase indoor radon levels and the associated health risks. Radon emanation and exhalation depend on the abundance and spatial distribution of radium atoms within the grains, but also on the type of the material and other properties such as grain size, porosity, and humidity. Increasing moisture content of the material enhances radon emanation and exhalation by increasing the probability of capturing radon atoms in the pore spaces (Barillon et al. 2005; Bossew 2003; Janik et al. 2015; Sakoda et al. 2011). Previous studies have also shown that increasing the size of solid grains usually leads to decrease in radon emanation fraction, particularly for grain sizes below 1μm (Sakoda et al. 2011). The dependence of radon exhalation rate on sample porosity has also been explored; some authors reported steady increase of radon exhalation with increasing porosity of the medium (Hassan et al. 2009), while a recent study of Pyngrope et al. (2022) reported no convincing trend of variation i.e. it initially increased, saturated till some values of porosity and then it either increased or decreased. Accordingly, emission of radon from each source is a complex feature influenced by many different factors. Therefore, measuring radon emanation and exhalation from different materials found in human environment is of great importance for radon exposure prediction and radiation protection. Different methods for measuring these variables have been investigated and applied (Abo-Elmagd 2014; Gutiérrez-Álvarez et al. 2020; López-Coto et al. 2009a; Noverques et al. 2019; Yang et al. 2017).

A number of studies compared different methods of measuring radon exhalation and radon emanation fraction. Müllerová et al. (2018) measured radon exhalation of soil using AlphaGUARD and scintillation chamber of Lucas type; a good agreement between these two methods was reported. López-Coto et al. (2009b) compared active and passive measuring techniques and obtained a good agreement for soil and large discrepancies for phosphogympsum samples probably due to uncontrolled radon leakages in the passive technique. Generally, mesurements of radon emanation and radon exhalation rate depend on the characteristics of experimental system (such as leakage rate, back-diffusion rate etc.) and particular conditions used in the measurements. The methods based on closed can techniques also depend on volumetric fractions of radon short-lived progeny i.e. on the fractions of ^{218}Po and ^{214}Po that decay in the air volume before the deposition, affecting the measuring efficiency. Methods for estimation of radon emanation power based on gamma-spectrometric measurements are also widely used (Hancerliogullari et al. 2019; Sakoda et al. 2010; Turhan et al. 2018).

This study was conducted to compare the results of different methods commonly used for estimating radon emanation power, as well as to explore the parameters involved in the estimation (such as leakage rate, initial measuring conditions and ^{218}Po volumetric fraction). Two methods considered here are based on estimating radon exhalation rates by active (RAD7) and passive (CR-39) radon measurements while the third method included gamma-ray spectrometry measurements. The study also tends to point out the similarities and differences among three commonly used modifications of active measuring technique, in order to explore the advantages and flaws of their application.

2. MATERIALS AND METHODS

2.1 Gamma spectrometry

Surface samples (1-10 cm in depth) of undisturbed soil were taken from six different, randomly selected locations applying IAEA (2004) template method. Sampling locations were spatially very distant from each other, ensuring different physico-chemical properties of sampled soils. Samples were cleaned from stones, grass and plant roots, air dried for two weeks, ground in a glass mortar and sieved through 2 mm mesh. Samples (0.5-0.6 kg in mass) were further packed in 450 mL plastic Marinelli beakers, filling them almost completely to avoid additional errors (Carconi et al. 2012; Mauring and Gäfvert 2013). The beakers were wrapped with adhesive, airtight tape to prevent leakage of radon during the period of achieving the equilibrium (around a month). Gamma spectrometry was

performed using coaxial HPGe detector (model GEM30-70 ORTEC) with 30% relative efficiency. The detector resolution is 1.65 keV at FWHM of 1.33 MeV gamma-ray photopeak from ^{60}Co. In order to reduce the background, the detector was placed in 10 cm lead shield coated with 1 cm thick copper layer. Calibration was performed using a calibration source type MBSS 2 (a Marinelli mixture of eleven radionuclides provided by the Czech Metrological Institute). Sample densities were similar to that of the calibration source and self-attenuation in samples was assumed to be negligible due to the fact that only lines above 200 keV were considered (Millsap and Landsberger 2015). Specific activity of ^{226}Ra was estimated by averaging the specific activities of its progeny ^{214}Pb and ^{214}Bi obtained using the photopeaks at 351.9 keV and 609.3 keV, respectively. The counting time for each soil sample was 10800 s.

2.2 Calculation of radon emanation power (EP) by gamma-ray spectrometry method

The difference between total amount of ^{222}Rn generated by some material (^{226}Ra activity of 4-week sealed samples) and the ^{222}Rn trapped in the material (^{214}Pb and ^{214}Bi activity of the ventilated sample) yielded the amount of ^{222}Rn that escaped from the sample (Chowdhury et al. 2002). Therefore, a simple gamma-ray spectrometry method for estimation of radon emanation power was proposed, including two measurements of net counts at gamma-ray photopeaks of ^{214}Pb (351.9 keV) and ^{214}Bi (609.3 keV) (Hancerliogullari et al. 2019; Sakoda et al. 2010; Turhan et al. 2018). The following formula was used to calculate the emanation power (*EP*):

$$EP = \frac{N_{eq} - N_0}{N_{eq}} \quad (1)$$

where N_{eq} and N_0 are the averages of the net count rates of two photopeaks (351.9 keV and 609.3 keV) in the equilibrium (4-week sealed) and initial (ventilated) conditions, respectively.

In this study, net count rates were recorded when the equilibrium between ^{226}Ra and ^{222}Rn was achieved in samples (closed Marinelli beakers). After that, the beakers were opened, and the same samples were measured once again. N_{eq} and N_0 (corresponding to the first and the second measurement, respectively) were obtained by averaging net count rates at the energies of 351.9 keV and 609.3 keV.

2.3 Measuring of radon exhalation rates by RAD7

In order to estimate radon exhalation rates, samples were placed in a glass petri dish (18 cm in diameter, 3 cm in depth) and enclosed in Plexiglas chamber (~ 30 L in volume) connected to RAD7 measuring device (Durridge Company). A schematic view of the experimental setup was shown in Fig. 1 (Stajic and Nikezic 2015). Radon concentration in the chamber air was continuously measured during 10-14 days (in 1-hour cycles) for each sample. As the result, radon build-up curves were obtained. Fitting the experimental data was performed using three models presented below.

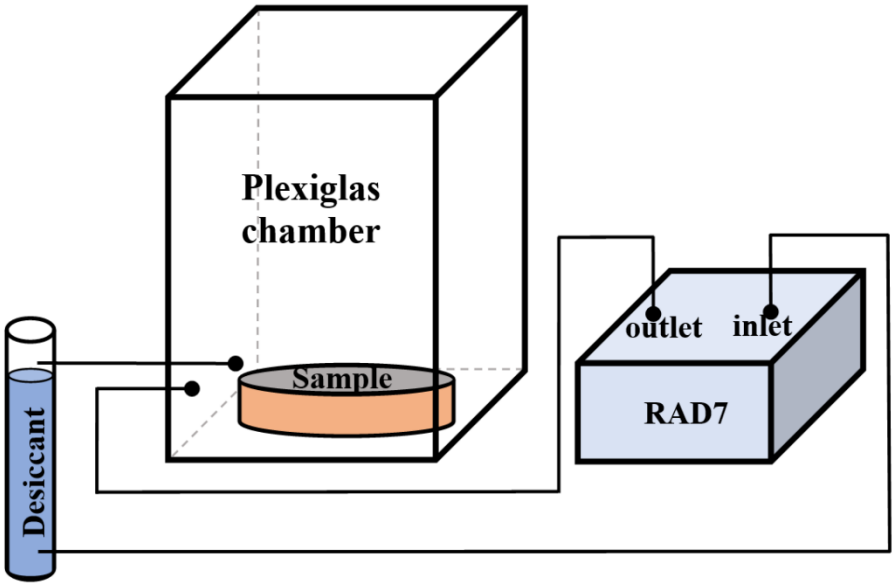

Figure 1. A schematic view of the experimental setup employed for active measurements.

2.3.1 Model 1

The accumulation of radon in the chamber is controlled by three processes: radon exhalation from the sample, radioactive decay and radon leakage from the chamber. Assuming that the measuring system itself does not exhale radon, the growth of radon concentration in the chamber, $C(t)$ can be presented using the following equation (Stajic and Nikezic 2015):

$$\frac{dC(t)}{dt} = \frac{Em}{V} - \lambda C(t) - \lambda_L (C(t) - C_{ext}) \quad (2)$$

where:

E – radon mass exhalation rate;

m – mass of the sample;

V – free chamber volume;

C_{ext} – radon concentration measured outside the chamber (currently treated as a constant although it is important to note that considering temporal variation of radon outside the chamber, $C_{ext}(t)$, would be more accurate and even necessary in some cases);

λ – radon decay constant (~ 0.00755 h^{-1});

λ_L – radon leakage rate.

Obviously, the previous equation does not include back-diffusion processes which are commonly negligible when a sample occupies less than 10% of a chamber volume (Poffijn et al. 1984; Ujić et al. 2008). If the initial radon concentration in the chamber is $C_0 = C\ (t = 0)$, the solution of Eq. 2 can be written as:

$$C(t) = \frac{Em+\lambda_L V C_{ext}}{V(\lambda+\lambda_L)}\left(1 - e^{-(\lambda+\lambda_L)t}\right) + C_0 e^{-(\lambda+\lambda_L)t} \quad (3)$$

Similar forms of the previous equation can be found in many papers (Abo-Elmagd 2014; Gutiérrez-Álvarez et al. 2020; López-Coto et al. 2009a; Müllerová et al. 2018; Yang et al. 2017). Slight differences between the formulas are due to the fact that most authors usually assume that initial radon concentration in the chamber is equal to 0 and the external concentration (outside the chamber) is not considered. However, Eqs. 2 and 3 include both parameters, assuming that it was more applicable for the real laboratory conditions.

Radon leakage rate λ_L can be obtained by observing the decrease of radon concentration in the "empty" chamber (chamber without samples). In the course of the study, radon was accumulated in the chamber using a sample of uranium ore, and then left to decay naturally (Fig. 2).

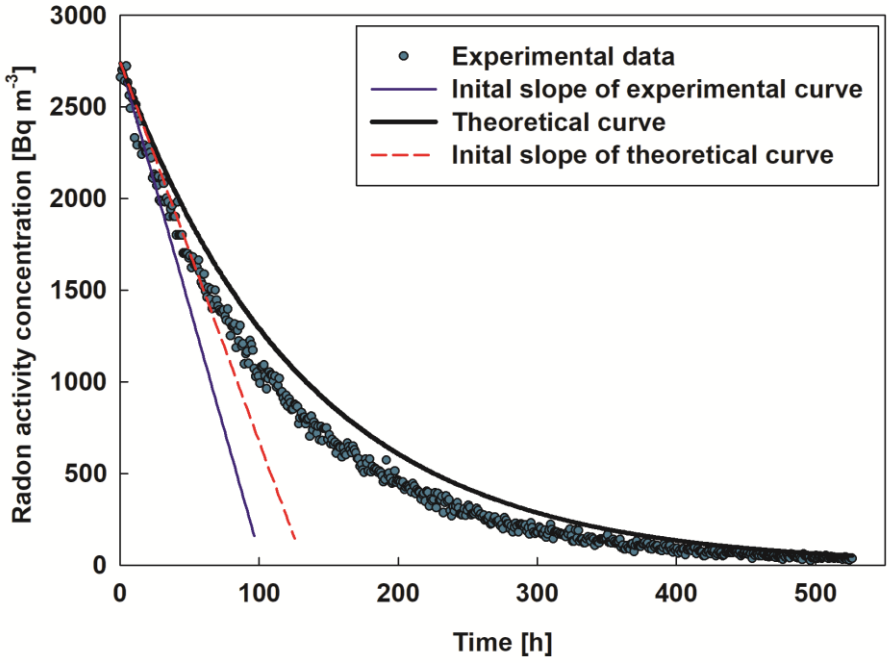

Figure 2. Decrease of radon concentration in the empty chamber: experimental observation and the theoretical curve presenting simple radon decay

In this case, the decrease of radon concentration in the chamber is caused only by the decay and leakage and therefore can be expressed as follows (based on Eq. 3):

$$C_{D+L}(t) = \left[C_0 - \frac{\lambda_L C_{ext}}{\lambda+\lambda_L}\right] e^{-(\lambda+\lambda_L)t} + \frac{\lambda_L C_{ext}}{\lambda+\lambda_L} \quad (4)$$

In the current model (*Model 1*), radon leakage rate was estimated by the method based on the initial slopes of radon decreasing curves (Abo-Elmagd, 2014; Chao et al. 1997). If the initial slope of radon decreasing curve is denoted by K_{L+D}, then:

$$K_{D+L} = \left(\frac{dC_{D+L}(t)}{dt}\right)_{t\to 0} = \lambda_L(C_{ext} - C_0) - \lambda C_0 \quad (5)$$

On the other hand, in the ideal case when there is absolutely no leaking of radon from the chamber (λ_L = 0), radon decay curve would be defined by a simple equation:

$$C_D(t) = C_0 e^{-\lambda t} \quad (6)$$

The initial slope of the simple decay curve is then:

$$K_D = \left(\frac{dC_D(t)}{dt}\right)_{t\to 0} = -\lambda C_0 \quad (7)$$

The difference between two slopes defined by Eq. 5 and Eq. 7 is:

$$K_D - K_{D+L} = \lambda_L (C_0 - C_{ext}) \quad (8)$$

Therefore, radon leakage rate can be calculated using the difference between the initial slopes of the experimental and theoretical radon decreasing curves:

$$\lambda_L = \frac{K_D - K_{D+L}}{C_0 - C_{ext}} \quad (9)$$

Fig. 2 presents the decrease of radon concentration in the chamber monitored for about 20 days (in 1-hour cycles). The initial slope of the curve (K_{D+L}), as well as the initial radon concentration (C_0) were obtained by linear regression of data recorded during the first few hours of measurements (Fig. 1). Figure 1 also presents the theoretical curve obtained for ideal chamber conditions with no leaking allowed (Eq. 6). Average radon concentration measured outside the chamber was C_{ext}= 28.5 ± 1.4 Bq m-3. Finally, radon leakage rate was obtained as λ_L = (0.0022 ± 0.0011) h^{-1}. Radon leakage was estimated in two separate experiments and the results were quite similar. Therefore, it was assumed that radon leakage would be almost the same after each closing of the chamber. Using this value of leakage rate, radon build-up curve obtained for each sample was fitted to Eq. 3 in order to obtain radon exhalation rate as a fitting parameter.

2.3.2 Model 2

This model is also based on fitting the experimental data to Eq. 3. Accordingly, leakage rate λ_L, can simply be obtained by fitting the data of radon decrease in the empty chamber (Fig. 2) to the following equation (assuming E = 0):

$$C(t) = \frac{\lambda_L V C_{ext}}{V(\lambda+\lambda_L)}\left(1 - e^{-(\lambda+\lambda_L)t}\right) + C_0 e^{-(\lambda+\lambda_L)t} \quad (10)$$

In this case, the leakage constant was obtained as a fitting parameter: λ_L = (0.00168 ± 0.00004) h^{-1}. This method provides more accurate determination of λ_L, but it is more time-consuming than the previous one.

2.3.3 Model 3

After determining radon leakage rate by some of the methods described above, radon exhalation rate can also be estimated using the initial slope of radon build-up curves obtained by RAD7 measurements (Abo-Elmagd 2014; Chao et al. 1997). The initial slope *S* can be derived from Eq. 3 as follows:

$$S = \left(\frac{dC(t)}{dt}\right)_{t\to 0} = \left(\frac{Em+\lambda_L V C_{ext}}{V(\lambda+\lambda_L)} - C_0\right)(\lambda+\lambda_L) \quad (11)$$

Therefore, radon exhalation rate can be expressed as:

$$E = \frac{V}{m}[S + C_0(\lambda+\lambda_L) - \lambda_L C_{ext}] \quad (12)$$

This method is based on the fact that the first portion of radon growth curve is nearly a straight line (as shown by Fig. 3). According to Chao et al. (1997), the initial slope S can be obtained by visual inspection or by considering the data of the first few hours modified by linear regression technique. In the present study, linear fitting of the first 20 data points was performed (Fig. 3).

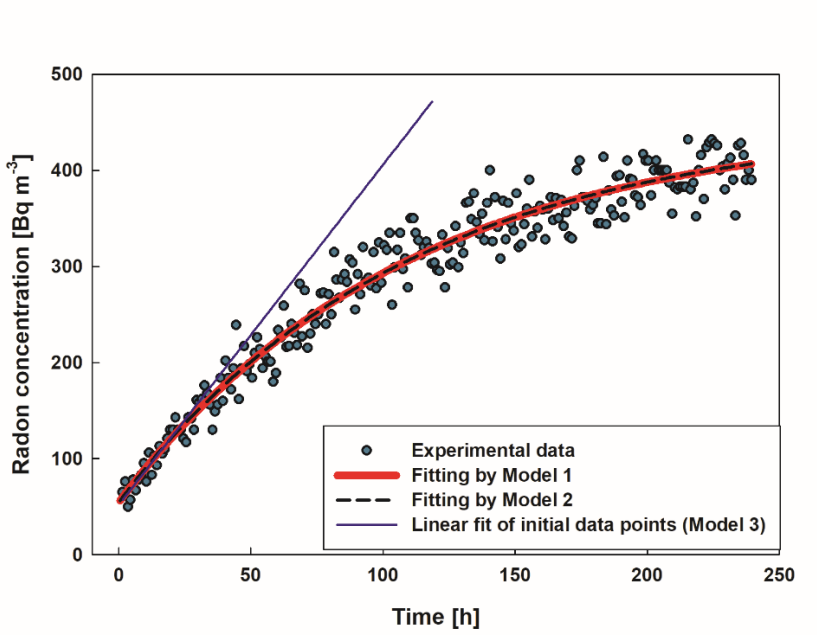

Figure 3. Application of three models based on RAD7 measurements (sample 3).

2.4 Radon exhalation rate estimation based on CR-39 detectors

The highest radon exhalation rate measured by previous method was also checked by a method based on passive radon measurements. The sample (denoted as Sample 1 WET in Table 1) was enclosed in the Plexiglas box along with two conical radon diffusion chambers:

Chamber 1 (lower radius: R_1 = 2.6 cm, upper radius: R_2 = 3.3 cm, and height: H = 10 cm)

Chamber 2 (lower radius: R_1 = 2.4 cm, upper radius: R_2 = 3.5 cm, and height: H = 8.2 cm).

The experimental setup is illustrated in Fig. 4. Each chamber was equipped with one CR-39 detector (2x2 cm^2, TASTRAK®, 1 mm in thickness). Chambers were placed at some distance from the sample to eliminate thoron contribution, and filter paper was used to prevent radon (and thoron) progeny from entering the chambers. The exposure lasted for 70 days. After that, CR-39 detectors were etched in a water bath, at the temperature of (70±1) ^{0}C, for 6 h in 6.25 N solution of NaOH. Alpha-particle tracks on detectors were observed and counted using optical microscope. Track densities were estimated by randomly selecting about 50 fields of view for each detector.

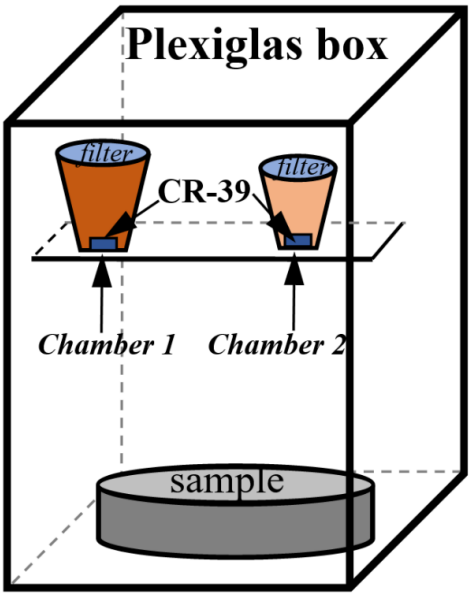

Figure 4. A scheme of the experimental setup used for CR-39 measurements.

The growth of track density (ρ) on solid state nuclear track detectors (SSNTD) exposed to variable radon concentration is defined by (Ujić et al. 2008):

$$\frac{d\rho(t)}{dt} = kC(t) \tag{13}$$

where k represents corresponding calibration coefficient of radon diffusion chamber. Therefore, total track density produced during the exposure time T can be expressed by the equation:

$$\rho = k \int_0^T C(t)dt \quad (14)$$

Applying the function of radon concentration growth *C (t)* expressed by Eq. 3 into the previous equation and integrating it over the exposure time (T = 70 days) gives the expression for ρ:

$$\rho = k\left(\frac{Em+\lambda_L V C_{ext}}{V(\lambda+\lambda_L)^2}\left(e^{-(\lambda+\lambda_L)T} + T(\lambda+\lambda_L) - 1\right) - \frac{C_0}{(\lambda+\lambda_L)}\left(e^{-(\lambda+\lambda_L)T} - 1\right)\right) \quad (15)$$

Therefore, radon exhalation rate obtained by CR-39 technique can be expressed through track density as follows:

$$E_{CR39} = \frac{\rho V(\lambda+\lambda_L) + C_0 kV\left(e^{-(\lambda+\lambda_L)T} - 1\right)}{\frac{mk}{\lambda+\lambda_L}\left(e^{-(\lambda+\lambda_L)T} + T(\lambda+\lambda_L) - 1\right)} - \frac{\lambda_L V C_{ext}}{m} \quad (16)$$

where all the parameters have already been defined above.

Calibration coefficients of two chambers were calculated using Fortran 90 computer program CR39_Sensitivity, previously developed by Nikezic et al. (2014). The program estimates the response of CR-39 detectors to alpha particles emitted by radon and its progeny in radon diffusion chambers. It takes into account the fractions of ^{218}Po and ^{214}Po that decay in the chamber volume (before deposition on chamber surfaces). ^{214}Po is assumed to decay as fully deposited, while different volumetric fractions of ^{218}Po (f_1) can be found in literature. According to experimental study of Koo et al. (2003), ^{218}Po volumetric fraction is about 0.4. However, some theoretical considerations suggests that most of ^{218}Po atoms produced in the chamber volume deposite before the decay i.e. $f_1 \rightarrow 0$ (McLaughlin and Fitzgerald 1994; Stajic et al. 2021). Both of these values were applied in the calculation. Besides, the program also requires bulk etch rate to be specified. It was determined in the experiment by gravimetric method, measuring detector masses before and after the etching. The value of (1.21 ± 0.08) μm h^{-1} was obtained and used in the program. V function reported by Durrani and Bull (1987) was chosen for calculation since it has been previously shown to give a good match to experimental data (Nikezic et al. 2014; Stajic et al. 2021). Finally, the values of calibration coefficients obtained for *Chambers 1* and *2* were 0.0180 and 0.0178, respectively (for f_1 = 0.4), and 0.0162 and 0.0159, respectively (for f_1 = 0).

2.5 Calculation of radon emanation power from radon exhalation rates

In the case when the dimensions of the sample are relatively small in comparison to radon diffusion length (~ 2.3 m in air, 1.5 m in coarse, dry sands, and 0.2 to 0.5 m in fine grained moist soils) (IAEA 2013; Ujić et al. 2008), it can be assumed that almost all radon atoms that emanate from grains will eventually be exhaled from the sample. The activity of radon exhaling from unit mass sample in unit time can be calculated by multiplying the number of radon atoms created in the sample (equal to ^{226}Ra activity concentration) by radon emanation power and radon

decay constant i.e., $E=EP \cdot A_{Ra} \cdot \lambda$ (Chowdhury et al. 2002; Turhan et al. 2018). Therefore, radon emanation power was determined using the following equation:

$$EP = \frac{E}{\lambda A_{Ra}} \quad (17)$$

where E is the mass exhalation rate (in Bq kg^{-1} h^{-1}) obtained by fitting experimental curves; λ is radon decay constant (in h^{-1}) and A_{Ra} is the specific activity of ^{226}Ra (in Bq kg^{-1}) in samples.

3. RESULTS

Radon exhalation rates (*E1*, *E2*, and *E3*) estimated by three models of fitting the data obtained by RAD7, as well as the corresponding radon emanation powers (*EP1*, *EP2*, and *EP3*) are presented in Table 1. Figure 3 illustrates the application of three models (for sample 3). It is obvious that fitting lines obtained by Models 1 and 2 match completely. The last column of Table 1 presents radon emanation power EP_γ estimated by gamma-ray spectrometry method. The uncertainties of gamma-ray spectrometry method, Model 3 (RAD7) and CR-39 method were obtained by combining uncertainties of all independent quantities, based on Eqs. 1, 12 and 16. The uncertainties in Models 1 and 2 were estimated by varying the parameters involved in Eq. 3 within their own uncertainty ranges.

Table 1. Radium specific activity (A_{Ra}), radon exhalation rates (*E1*, *E2* and *E3* obtained by Model 1, 2 and 3, respectively) and radon emanation powers (*EP1*, *EP2*, *EP3* and EP_γ) of soil samples.

Sample	A_{Ra} [Bq kg^{-1}]	*E1*	*E2*	*E3*	*EP1*	*EP2*	*EP3*	EP_γ
		[mBq kg^{-1} h^{-1}]			[%]			
1 WET	195±4	600±50	567±9	580 ± 40	41±4	38±2	40 ± 4	38±2
1 DRY	227±4	260±20	240±7	210 ± 40	15±2	14±1	13 ± 3	16±3
2	111±4	320±30	299±8	330 ± 40	38±4	36±3	39 ± 6	35±5
3	55±4	208±13	194±5	170 ± 30	50±5	47±3	41 ± 7	45±5
4	75±4	174±9	160±7	160 ± 30	30±4	28±2	29 ± 7	29±4

5	48±2	91±8	81±4	80 ± 20	25±3	22±2	23 ± 7	23±5
6	58±2	170±13	156±5	160 ± 30	39±4	36±2	36 ± 7	38±4

Figure 5 illustrates the effect of sample humidity on radon emanation and exhalation, demonstrated by two measuring methods applied before and after the drying. The left panel presents the growth of radon concentration in the Plexiglass chamber, monitored by RAD7 device. Obviously, radon concentration in the chamber reaches significantly higher levels in the case of "wet" sample.

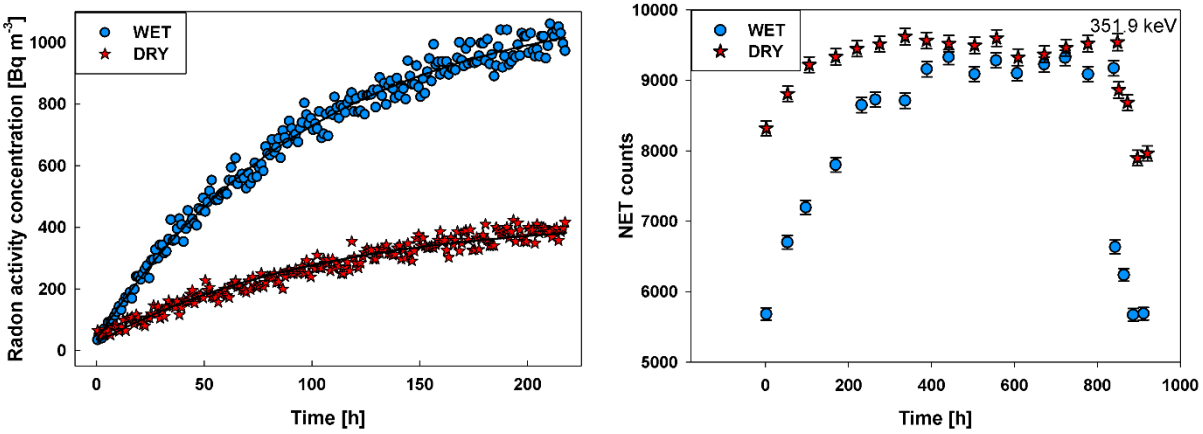

Figure 5. Sample No.1 WET and DRY: The growth of radon concentration inside the chamber, measured by RAD7 (left panel) and changes of net counts recorded in the gamma-ray line of 351.9 keV before and after opening Marinelli beakers (right panel).

Table 2 presents radon exhalation rate and radon emanation power of sample 1 WET, estimated by 70-day exposure of solid state nuclear track detectors (CR-39). Two radon diffusion chambers employed in the experiment gave quite similar results. Besides, ^{218}Po volumetric fraction of 0.4 provided lower results in comparison to total deposition (f_1=0). Compared to data in Table 1, the assumption f_1=0 seems to provide a slightly better agreement with the results obtained by other methods.

Table 2. Sample 1 WET radon exhalation rate (E_{CR39}) and radon emanation power (EP_{CR39}) estimated by CR-39 detectors for two chambers and two volumetric fractions of ^{218}Po.

	Chamber 1		**Chamber 2**	
	f_1**=0**	f_1**=0.4**	f_1**=0**	f_1**=0.4**
E_{CR39} **[mBq kg^{-1} h^{-1}]**	590±70	530±60	570±70	520±60
EP_{CR39} **[%]**	40±6	36±5	39±6	35±5

4. DISCUSSION

According to Table 1 a good agreement among the results was obtained; coefficient of variation ranged between 3% and 9%. It is important to mention that special care was taken to ensure a good sealing of Marinelli beakers used in gamma-ray spectrometry method. In order to avoid possible errors induced by radon leakage, using aluminum containers instead of plastic ones is highly recommended (Lee et al. 2018).

According to Porstendorfer (1994), typical values of radon emanation power of soil range between 1% and 50%. Values within this range were also measured by Greeman and Rose (1996) (5.5 – 32%), Bossew (2003) (6 – 47 %), and Milenkovic et al. (2015) (17 – 29%). A review article by Sakoda at al. (2011) reported mean radon emanation power of 20% based on 1025 soil samples. Radon emanation power obtained in the present study was higher than 20% in all original samples. However, it is important to mention that the samples were just air dried and there was probably some moisture remained in the pore spaces among soil grains. Sample 1 DRY was obtained by drying sample 1 WET in the oven until constant weight. Drying resulted in decreasing the sample mass by 15% as well as in reducing radon emanation power by more than 50% (Table 1). It is well known that radon emanation fraction increases with increasing moisture content due to the fact that stopping power of water is much greater than that of air (Bossew 2003; Sakoda et al. 2011). The recoil range of radon atoms in water is 688 times less than in air (IAEA 2013; Phong Thu et al. 2020). Therefore, water present in a pore space of porous material reduces the probability of recoiled radon atoms to penetrate the pore and to get embedded into some of the adjacent grains (Nazaroff 1992; Sakoda et al. 2011). Although it is more common to measure radon emanation and radon exhalation in completely dry samples, one should investigate emanation from material in environmental conditions, as they are more realistic ones.

The right panel of Figure 5 presents the changes in net counts at the energy of 351.9 keV, obtained by gamma spectrometry measurements of wet and dry samples. The measurements were performed every 2-3 days during one-month equilibration period and every 24 h after opening the beakers. Each point in the graph (right panel) is the result of 3 h gamma-ray acquisition. The measurements started right after sealing the samples in Marinelli beakers, while the last four data were recorded after opening the beakers. The initial net count rates are considerably higher in dry sample due to the fact that more radon atoms remain trapped in the solid grains, resulting in higher content of radon progeny in the sample. After the sealing, radon escape from beakers is prevented, leading to increase of gamma count rate in both, wet and dry samples. Evidently, counting rates reached the equilibrium level in less than 20 days. It took 2-3 days after unsealing the sample, for the final count rate to become equal to the initial one.

5. CONCLUSION

There is a quite good agreement among the results of different methods applied in the study. The method employing solid state nuclear track detectors requires long-time exposure to obtain reliable track counting statistics, particularly in the case of samples with low radon exhalation rates. Besides, the accuracy of these methods highly depends on determination of the system properties such as radon leakage rate (in Model 2, leakages are considered within the measurement itself, requiring no previous knowledge of them). Methods for estimating radon leakage rate and radon exhalation rate using the initial slopes of radon concentration curves (Models 1 and 3) are rather fast and convenient but tend to increase the uncertainty of the measurements. Gamma-ray spectrometry method is quite simple, but it takes at least a month to achieve secular radioactive equilibrium between ^{226}Ra and its decay products. Besides, a tight sealing of Marinelli beakers is required to prevent radon leakage which might result in underestimation of radon emanation power.

STATEMENTS AND DECLARATIONS

Funding: This work was supported by The Ministry of Science, Technological Development and Innovation of the Republic of Serbia (Agreement No. 451-03-47/2023-01/200378 and 451-03-47/2023-01/200123).

Conflict of Interest Statement: The authors declare that there is no conflict of interest.

Data Availability Statement: Data available on request from the authors.

Ethics Approval: Not applicable

Consent to participate: Not applicable

Consent to publish: Not applicable

Acknowledgement

The work was supported by The Ministry of Science, Technological Development and Innovation of the Republic of Serbia (451-03-47/2023-01/200378, 451-03-47/2023-01/200123). We would like to thank Prof. Dr. Şeref Turhan (from Department of Physics, Faculty of Science and Letters, Kastamonu University) for a valuable help and a friendly discussion on the topic.

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