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A Preliminary Study to Determine the Radon Exhalation Rates of Soil Samples at Different Depths

Farklı Derinliklerdeki Toprak Örneklerinin Radon Salınım Hızlarının Belirlenmesine Yönelik Ön Çalışma

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Abstract

The radon area exhalation rate and mass exhalation rate of the soil samples collected from five different depths from soil surface with 20 cm intervals of a construction site at Kütahya, Turkey was determined by using the E-Perm detectors. Accumulator method is used to determine the radon concentration of the soil samples. Radon concentration of the soil samples were measured between 34 ± 4.4 Bq.m^{-3} to 86 ± 6.8 Bq.m^{-3} with an average value of 48.6 ± 5.1 Bq.m^{-3} for five days analysis time. Radon area exhalation rate of the soil samples were calculated between 163.7 ± 21.3 and 413.1 ± 32.6 mBq.m^{-2}.h^{-1} with an average of 233.2 ± 24.4 mBq.m^{-2} .h^{-1}. Radon mass exhalation rate of the soil samples were found between 32.9 ± 4.3 to 83 ± 6.6 mBq.kg^{-1}.h^{-1} with a mean value of 46.9 ± 4.9 mBq.kg^{-1}.h^{-1}. The radon contribution to the indoor air from soil samples is estimated as 6.5 Bq.m^{-3}.

Keywords: *Radon area exhalation rate, Radon mass exhalation rate, Indoor radon contribution estimation*

Öz

Kütahya, Türkiye'de bir inşaat alanında toprak yüzeyinden itibaren 20 cm aralıklarla beş farklı derinlikten toplanan toprak örneklerinin radon alan salınım hızı ve kütle salınım hızları E-Perm algıçları kullanılarak belirlenmiştir. Toprak örneklerinin radon konsantrasyonu akümülatör yöntemi kullanılarak belirlenmiştir. Toprak örneklerinin radon konsantrasyonu beş günlük analiz süresinde 34 ± 4,4 Bq.m^{-3} ile 86 ± 6,8 Bq.m^{-3} aralığında, ortalama 48,6 ± 5,1 Bq.m^{-3} olarak ölçülmüştür. Toprak örneklerinin radon alan salınım hızları 233,2 ± 24,4 mBq.m^{-2}.h^{-1} ortalama ile 163,7 ± 21,3 ve 413,1 ± 32,6 mBq.m^{-2}.h^{-1} aralığında hesaplanmıştır. Toprak örneklerinin radon kütle salınım hızları 32,9 ± 4,3 ve 83 ± 6,6 mBq.kg^{-1}.h^{-1} aralığında ortalama 46,9 ± 4,9 mBq.kg^{-1}.h^{-1} olarak bulunmuştur. Kapalı ortama örnekleme yapılan toprak örneklerinden 6,5 Bq.m^{-3}'lük radon katkısı geldiği tahmin edilmektedir.

Anahtar Kelimeler: *Radon alan salınım hızı, raadon kütle salınım hızı, kapalı ortama radon katkısı tahmini*

1. Introduction

Radon (Rn-222) is naturally occurring radioactive gas originating from the decay of the U-238 series found everywhere in the nature with a half-life of 3.8 days. Radon is one of the well known alpha particle emitters. Alpha particles have short range and can be stopped by the skin but in case of respiration in high activities, radon may become dangerous to human health and can cause deaseases such as lung cancer. Thus monitoring of radon gas becomes important in environments such as indoor places, water sources, mines etc [1].

Radon can transported from porous materials to atmosphere by emanation, diffusion, advection and exhalation, thus contributes to environment radon concentration, especially to indoor radon concentration [1, 2, 3]. Radon exhalation can be defined as the escape of radon gas from porous materials into the atmosphere [1]. The radon gas released from the unit surface area per unit time is called as radon area (surface) exhalation rate [4].

Radon exhalation rate strongly depends on Ra-226 concentration of materials and its distribution, also depends on several factors such as the geological properties of region of interest, emanation power, porosity of material, soil temperature, soil water saturation, atmospheric pressure [5, 6]. The main sources of indoor radon concentration are the building materials and soil, making it necessary to determine the radon exhalation rate of these materials, as it is an important parameter in terms of health [2, 7- 11].

Accumulator method is widely used to determine the radon concentration of solid samples, thus radon area exhalation rate and mass exhalation rate can be determined. When the sample is placed in a sealed chamber to allow radon growth inside, the sealed chamber acts as an accumulator. Some precautions are to be taken to prevent the underestimation of the radon concentration inside the accumulator which increases with time, especially in areas close to the sample. There will be a difference between the radon concentration of the ambient air of the accumulator and the radon concentration at the surface of sample. The difference depends on the accumulator geometry and on the detector area that covers the sample. Radon atoms also diffuse back to the sample. This results in the decrease of equilibrium radon concentration inside the accumulator and causes the underestimation of radon exhalation rate [11-15]. Such a phenomenon is called the radon back diffusion effect. If the volume of accumulator's ambient air is more than 10 times of the sample volume, radon back diffusion effect is lower than 10% and can be ignored [4, 13]. It is also noted by Kotrappa and Stieff that the effect of radon back diffusion increases for the measurement (analysis) periods over 5 days [4].

Soil radon exhalation rate data is limited in Turkey: it is only performed for several locations by several authors by using track detectors (CR-39 and LR-115 type II Solid State Nuclear Track Detectors-SSNTD) and by gamma spectroscopy methods as given in the Table 1 [5, 16-18].

Tablo 1. Türkiye'de toprak örneklerinin Radon alan salınım hızı ve kütle salınım hızı.

Table 1. Radon area and mass exhalation rate of soil samples around the Turkey.

Sampling Site	**Area Exhalation Rate ($mBq.m^{-2}.h^{-1}$)**	**Mass Exhalation Rate ($mBq.kg^{-1}.h^{-1}$)**
Sakarya, Turkey [5]	73 - 5180	35.76 - 253.15
Adapazarı, Turkey [16]	1035.18-5333.39	50.35 - 259.41
North and East Anatolian fault zone in Turkey [17]	13.3 - 400.7	-
Karabük, Turkey [18]	-	29.88 - 137.16

A preliminary study was conducted to determine the radon area exhalation rate and mass exhalation rate of the soil and the radon contribution to a new building in a construction site located in Kütahya, Turkey. Longitude and latitude of the studied area are 39.412996 and 29.981550, respectively. Soil samples were collected in May, 2022 from five different depths of the construction site. Radon concentrations of soil samples were determined using the E-PERM method. Later, radon area exhalation rate and mass exhalation rate were calculated, indoor radon contribution from soil to indoor air of the building is estimated.

2. Material and Method

At the previously metioned construction site at the city center of Kutahya Province, Turkey, an area of 50 cm x 50 cm on the soil surface was cleared of grass, roots, stones and pebbles. Five soil samples were collected from different depths starting from 20 cm below the surface with 20 cm intervals up to 100 cm soil depth. Soil samples each of mass approximately 1 kg were gathered into the polyethylene bags. Later, samples were brought to the Nuclear Physics Research Laboratory at Dumlupinar University, Kütahya. Samples were dried in oven to remove moisture and sieved with a maximum grain size of 2 mm. Bulk density of soil samples varies from 1.56 to 2.24 g.cm^{-3}. Sample weights are used to determine the net volume of the samples.

E-PERM radon detectors were used to measure the radon concentration of soil samples to determine the radon area exhalation rate and mass exhalation rate. E-PERM is a commercially available passive method and it is generally used to determine the indoor and water radon concentration as well as the Ra-226 concentration. Only radon (Rn-222) can be determined with appropriate setup with this method, other isotopes of radon like thoron (Rn-220) can not be determined without modifying method [19]. Accumulator method is used to determine the radon concentration of the soil samples.

A glass jar, which contains an E-PERM radon-in-water kit, is used as an accumulator. E-PERM radon-in-water kit glass jars have their own rubber sealing o-ring to prevent the leakage between the ambient air of the jar and the atmosphere.

E-PERM S-chamber and short term electret configuration which corresponds to E-PERM SST-configuration were used to determine radon concentration inside the jars. This configuration is appropriate for 2 to 7 days short-term radon measurements. Electrets are made of Teflon and they are electrically charged. Their voltages are measured by special voltage reader.

100 grams of each soil sample was weighed and placed into jars. The soil sample was evenly spread on the bottom of the jar. Initial voltage of the ST-electrets were measured and E-PERM SST-configuration is placed to the cover of the jar as seen from schematic of experimental setup given in Figure 1. The cover of glass jars carefully sealed to prevent radon escape. Measurements were performed for 5 days to measure the radon concentration of the soil samples inside the jars.

The air volume of the jar has some radon concentration at the time of the sealing which will effects our measurement results. An empty jar is used to determine the ambient (background) radon concentration as advised by Kotrappa and Stieff [4]. All measurements were performed simultaneously.

Şekil 1. E-Perm sistemi ile Radon salınım hızı ölçümü

Figure 1. Radon exhalation rate measurement with E-Perm system

Ratio of ambient air of the accumulator to the volume of the sample is greater than 10 and analysis time is 5 days, thus radon back diffusion effect can be neglected under these test conditions with respect to literature. After 5 days measurement time, E-PERM detectors were removed from the jars and final voltages of electrets determined immediately. Radon concentration of the accumulators ambient can be determined by electrets voltage drop as [20]:

$$RnC = \left[\left(\frac{(IV-FV)-(0.067\times TA)}{CF\times TA}\right) - (BG\times G)\right]\times(Elev\ CF)\times 37 \quad (1)$$

where RnC is the average radon concentration in Bq.m^{-3} units, IV and FV are the initial and final voltage of electrets placed into the accumulators, TA is the analysis time in days, Elev CF is the correction factor of the test site where the E-PERM electrets used. E-PERM detectors are also sensitive to gamma radiation, background gamma radiation of the laboratory, BG, is measured by using Ludlum 44-2 handheld detector, G is the gamma conversion constant. CF is the calibration factor that is the response of the electrets voltage drop to the known activity source that is given by manufacturer for SST E-PERM configuration as:

$$CF = a + \left(b \times \left(\frac{IV + FV}{2}\right)\right) \quad (2)$$

a=1.69776 and b=0.0005742 are calibration constants. E-Perm detectors calibration constant depends on the initial (IV) and final (FV) voltage of electrets as given in Equation 2, calibration factors of electrets used in this study varies between 1.9352 and 2.0225 V/Bq.m^{-3}.d.

2.1. Lower limit of detection (LLD)

Lower limit of detection(LLD) for the E-PERM system is defined as the measurement of the radon concentration with 50% uncertainty. Uncertainty of the measurements can be calculated as follows [21]:

$$E = \begin{pmatrix} (RnC)^2 \times \\ \left((0.05)^2 + \left(\frac{\sqrt{2}}{(IV - FV)}\right)^2\right) \\ +(0.10 \times BG \times G)^2 \end{pmatrix}^{\frac{1}{2}} \quad (3)$$

and percent uncertainty %E is given by:

$$\%E = 100 \times \frac{E}{RnC} \quad (4)$$

The voltage of the E-PERM electrets decreases as they exposed to radiation. This makes necessary to determine the LLD of each electret. It is determined that the lowest and highest LLD for the 5 days measurements are 2 and 7 Bq.m^{-3}, respectively.

2.2. Radon exhalation rate determination

The radon area (surface) exhalation rate, E_A given in the units of mBq.m^{-2}.h^{-1} is defined as the activity transfer rate per unit area at the soil-air interface [22] can be calculated by following equation:

$$E_A = \frac{RnC \times V \times \lambda}{A \times \left[1 - \left(\frac{1 - e^{-\lambda \times TA}}{\lambda \times TA}\right)\right]} \times \frac{1000}{24} \quad (5)$$

where A is the area of the sample in m^2, λ is the decay constant of Rn-222, TA is the analysis time in days and V is the air volume of the jar in m^3 units, (1000/24) is used to convert the units to appropriate ones for comparison with the literature [4, 11].

Mass exhalation rate E_M in mBq.h^{-1}.kg^{-1} units can be calculated by following equation:

$$E_M = \frac{RnC \times V \times \lambda}{m \times \left[1 - \left(\frac{1 - e^{-\lambda \times TA}}{\lambda \times TA}\right)\right]} \times \frac{1000}{24} \quad (6)$$

where, m is the sample mass in kg units.

2.3. Indoor radon concentration estimation

Radon transfer from soil to indoor environment can be calculated by folowing equation [5]:

$$C_{Rn}^{ind} = \frac{E_A \times S_r}{V_r \times \emptyset} \quad (7)$$

C_{Rn}^{ind} is the estimated indoor radon concentration in Bq.m^{-3} units, E_A is the radon area exhalation rate in Bq.m^{-2}.h^{-1} units, S_r is the internal surface area of the room in m^2 units, V is the volume of the room in m^3 units, $\emptyset$ is the air exchange rate in h^{-1} units.

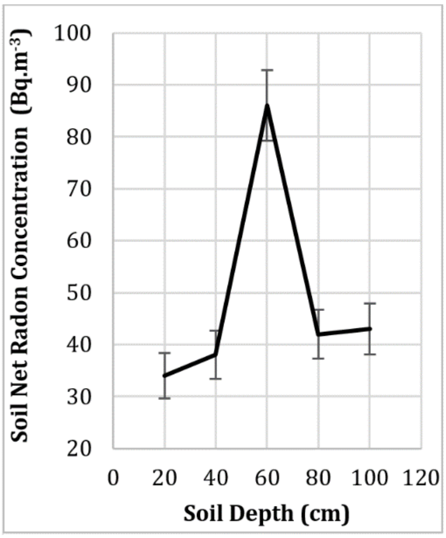
3. Results and Discussion

Gross (background + net) radon concentrations and background radon concentration were measured and calculated as given in Table 2. Background radon concentration is measured as 18 ± 1.9 Bq.m^{-3}. Gross radon concentration of soil samples varies between 52 ± 4 Bq.m^{-3} to 104 ± 6.5 Bq.m^{-3}. Uncertainty errors of the radon measurements varies between 6.3 to 10.6 % where the highest uncertainty belongs to the background measurements and all uncertainty error percentages are lower than the 50% which shows us that all results are higher than the LLD. Net radon concentrations were calculated by subtracting background radon concentration from gross radon concentration and found between 34 ± 4.4 Bq.m^{-3} to 86 ± 6.8 Bq.m^{-3} with an average value of 48.6 ± 5.1 Bq.m^{-3}. Lowest radon concentration was found to be at the sampling point of 20 cm below the surface where it is the closest point to the surface. Highest radon concentration was found to be at 60 cm depth. The measurement results shows us that radon concentration increases with increasing depth up to 60 cm sampling depth and then radon concentration drops to value where it does not show significant change as shown in Figure 2. No correlation was found between radon concentration and depth. Soil radon concentration strongly depends on the soil radium (Ra-226) concentration which indicates that soil radium concentration shows significant changes in short distance.

Tablo 2. Toprak örneklerinin Radon konsantrasyonu.

Table 2. Radon concentration of soil samples.

Soil Depth (cm)	Gross Radon concentration (Bq.m^{-3})	Background Radon concentration (Bq.m^{-3})	Net Radon concentration (Bq.m^{-3})
20	52 ± 4.0	18 ± 1.9	34 ± 4.4
40	56 ± 4.2	18 ± 1.9	38 ± 4.6
60	104 ± 6.5	18 ± 1.9	86 ± 6.8
80	60 ± 4.3	18 ± 1.9	42 ± 4.7
100	61 ± 4.5	18 ± 1.9	43 ± 4.9



Şekil 2. Toprak örneklerinin net radon konsantrasyonunun derinlikle değişimi

Figure 2. Variation of net radon concentration of soil samples with depth

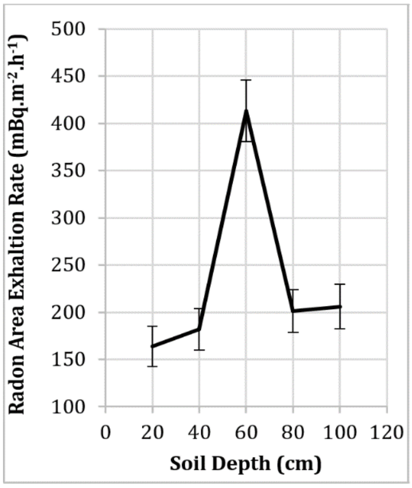
Results of radon area exhalation rate and mass exhalation rate of soil samples are given in Table 3 and also presented in Figure 3 and Figure 4. Radon area exhalation rate for soil samples varies between 163.7 ± 21.3 and 413.1 ± 32.6 mBq.m^{-2}.h^{-1} with an average value of 233.2 ± 24.4 mBq.m^{-2}.h^{-1}. Mass exhalation rate of soil samples varies between 32.9 ± 4.3 and 83 ± 6.6 mBq.h^{-1}.kg^{-1} with an average value of 46.9 ± 4.9 mBq.h^{-1}.kg^{-1}. The radon area exhalation rate and mass exhalation rate variation of the samples are similar as the variation in radon concentration, as expected. Both of them are found to be lowest at the 20 cm depth where it is closest to the surface and highest at the 60 cm depth.

Tablo 3. Örneklerin Radon Alan (yüzey) salınım hızı ve Kütle salınım hızı.

Table 3. Radon Area (surface) exhalation rate and Mass exhalation rate of samples.

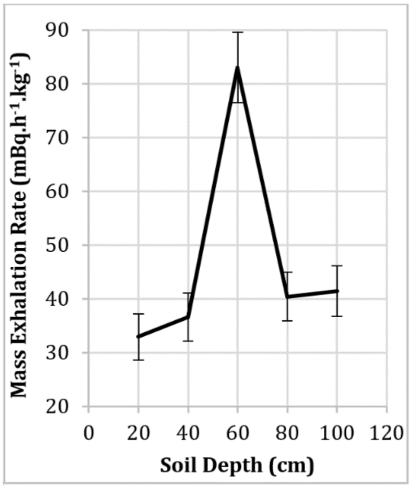
Soil Depth (cm)	Radon Area Exhalation Rate (mBq.m^{-2}.h^{-1})	Mass Exhalation Rate (mBq.h^{-1}.kg^{-1})
20	163.7 ± 21.3	32.9 ± 4.3
40	181.9 ± 36.6	36.6 ± 4.4
60	413.1 ± 32.6	83.0 ± 6.6
80	201.1 ± 22.6	40.4 ± 4.6
100	206.1 ± 23.4	41.4 ± 4.7

A comparison is performed between the area and mass exhalation rate data found for the soil samples for the Turkey in the literature given in Table 1 with soil samples. The lowest and highest area radon exhalation rate data for soil samples given in the literature for Turkey changes from 13.3 to 5333.39 mBq.m^{-2}.h^{-1} over wide range. Our data is limited and found within the range of literature values. The mass exhalation rate literature values of soil samples for Turkey change from 29.88 to 259.41 mBq.kg^{-1}.h^{-1}. Our results are found to be in the range of the literature data for Turkey.



Şekil 3. Radon alan salınım hızının derinlikle değişimi

Figure 3. Area exhalation rate variation with depth



Şekil 4. Kütle salınım hızının derinlikle değişimi

Figure 4. Mass exhalation rate variation with depth

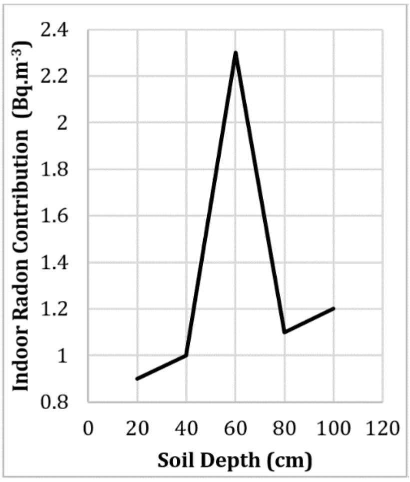
Indoor radon contribution from soil is also estimated. S_R/V_R ratio is calculated as 1.61 m^{-1} for 4x5x2.8 m^3 room dimensions and air exchanger rate is taken as 0.29 h^{-1} from the literature [23]. Results of indoor radon contribution from soil is given in Table 4 and also presented in Figure 5.

Indoor radon contribution from soil samples varies between 0.9 and 2.3 $Bq.m^{-3}$. The highest contribution to the indoor radon concentration from soil samples is found at 60 cm soil depth which corresponds to 2.3 $Bq.m^{-3}$ indoor radon concentration. Total of 6.5 $Bq.m^{-3}$ radon contribution to indoor air from these 5 sampling points is estimated. The estimation of the indoor radon concentration which is the result of the radon transfer from soil to indoor depends on several factors such as room dimensions and air exchange rate as well as the soil radon area exhalation rate. Such estimation may differ with respect to these factors and also depends on other effects like the sample's moisture content.

Tablo 4. Farklı derinliklerdeki toprak örneklerinin kapalı ortam radon konsantrasyonuna katkıları.

Table 4. Indoor radon concentration contributions of soil samples from different depths.

Soil Depth (cm)	**Indoor Radon Contribution ($Bq.m^{-3}$)**
20	0.9
40	1.0
60	2.3
80	1.1
100	1.2



Şekil 5. Farklı derinliklerdeki toprağın kapalı ortam radon konsantrasyonuna katkıları

Figure 5. Indoor radon contribution of soil from different soil depths

4. Conclusions

Radon concentrations of 5 soil samples collected from a construction site at Kütahya, Turkey were measured using E-PERM method. Radon area exhalation rate and radon mass exhalation rate of the soil samples were calculated. Indoor radon contribution from the soil samples were estimated by using the radon exhalation rate values of the samples. Estimated indoor value is lower than the action level for the Turkey [24].

5. Sonuçlar

Kütahya'daki bir inşaat alanından toplanan 5 toprak örneğinin radon konsantrasyonları E-PERM yöntemi kullanılarak ölçülmüştür. Toprak örneklerinin radon alan salınım hızı ve radon kütle salınım hızı hesaplanmıştır. Toprak örneklerinden kaynaklanan kapalı ortam radon katkısı, örneklerin radon alan salınım hızı değerleri kullanılarak tahmin edildi. Tahmin edilen kapalı ortam radon konsantrasyonu, Türkiye'de belirlenen eylem seviyesinden düşüktür [24].

6. Ethics committee approval and Conflict of interest declaration

Ethics committee approval is not required for the prepared article.

The author decleares that prepared article cannot have a conflict of interest with any person/institution.

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