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Controlled Laboratory Experiments on Radon Diffusion Coefficient

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ABSTRACT

Radon which occurs in uranium decay series is a noble gas and it moves from the deeper layers of the earth crust to the atmosphere. Formation of the radon gas in the Earth's crust and its migration to the atmosphere are quite a complex process due to the external factors like tectonic movements, meteorological factors etc. In this process, both the advection and diffusion are effective mechanisms for radon transport. But, the most effective mechanism is diffusion. In this study, radon diffusion of soils taken from Küçük Menderes Basin was investigated by controlled laboratory experiments and the radon diffusion coefficient of these soils was calculated. Soil gas radon measurements were performed by LR 115 Type 2 solid state nuclear track detectors.

Keywords: Radon, Diffusion, Soil gas

1. INTRODUCTION

Radon has been studied for a long time [1] because of its harmful health effects (lung cancer) and its concentration variation related to the geodynamic events (earthquakes and volcanic tremors etc.). It is a colorless, odorless, naturally occurring, chemically inert and radioactive gas [2, 3]. It is originated from the deep layers of earth crust and moves to the atmosphere by diffusion and convection or advection [4, 5, 6]. It is recognized as one of the most important environmental indicator or precursor for being a noble gas and a member of radioactive decay series [7].

The most important isotopes are radon (^{222}Rn), thoron (^{220}Rn) and actinon (^{219}Rn) which are the members of the uranium, thorium and actinium decay series, respectively. The abundance of uranium in the environment and relatively long half-life of ^{222}Rn ($t_{1/2}$=3.82 d) that the reason for the radon refers to ^{222}Rn [8]. The other radon isotopes are relatively lower half-life compared to ^{222}Rn (^{220}Rn; $t_{1/2}$=55 s, ^{219}Rn; $t_{1/2}$=3.96 s). The second important properties of radon (^{222}Rn) is its decay products are chemically active and relatively short lives radionuclides the following four decay products have a half-life of less than 30 min [1, 9, 10].

Radon transport is a fairly complex process and it should be examined to understand radon concentrations variations. In this study, radon diffusion coefficient of soil samples collected from Küçük Menderes Basin is studied.

1.1. Theoretical background

The main sources of radon are the rock and soil where radium decays to radon and then it escapes

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from the solid material to the soil pores. As it is a natural gas, radon migrates to the atmosphere by diffusion and advection mechanisms. However, it is interpreted that the third mechanism as convection, can occur when a sufficient thermal gradient is available within the soil, depending on many local parameters such as viscosity, porosity and permeability [11]. At the end of these process, radon reaches to the open air/indoor air and at least 80 % of the radon into the atmosphere comes from the top few meters of the surface soil [12].

Transport of radon strongly connected the several factors such as decay rate of radon, gases and liquid in the pores, pore volume, atmospheric pressure etc. [13, 14]. Radon moves to the atmosphere by diffusion and advection mechanisms. Diffusion is the simplest and usually the dominant mechanism which is defined by Fick's law. It can be described as a random molecular motion which relates a concentration gradient to a flux. According to the first theory of the Fick law, in steady state, the radon flux (J) is directed towards the region where the concentration is greatest in the region. To simplify the definition, the motion of radon can be considered in one-dimension equation [15];

$$J = -D\frac{\partial c}{\partial z} \qquad (1)$$

Here, J is the radon flux (Bq m^{-2} s^{-1}), D is the radon diffusion coefficient (m^2/s) and $\partial C/\partial z$ is the concentration gradient (Bq/m^3). Similarly, the second theory of the Fick law describes time-dependent concentration change as:

$$\frac{dC}{dt} = D\,\frac{d^2C}{dz^2} \qquad (2)$$

where C is the radon concentration (Bq/m^3), t is the elapsed time, and z is the depth from the soil surface.

$$\frac{\partial C(z,t)}{\partial t} = D\frac{\partial^2 C(z,t)}{\partial z^2} - \lambda X(z,t) + S(\Phi) \qquad (3)$$

The terms in Eq. (3) represents diffusion, radioactive decay of radon, radon production rate, respectively. Radon production rate is given by [16] as:

$$S(\Phi) = \frac{f.\lambda.\rho.C_{Ra}}{\varepsilon} \qquad (4)$$

where f is the emanation coefficient (dimensionless), ρ is the bulk density (kg m^{-3}), C_{Ra} is the radium concentration (Bq kg^{-1}) and ε is the porosity. To solve Eq. (3) it is assumed that radium concentration of soil (C_{Ra}) is constant (steady state method) and time-dependent radon production does not change (dC/dt=0). Therefore, a new form of Eq. (3) is;

$$D\frac{d^2C}{dz^2} - \lambda C = 0 \qquad (5)$$

and consecutive steps to solve the diffusion equation is given below according to [17, 18]:

$$D\frac{d^2C}{dz^2} - \lambda C = 0 \; (6)$$

$$D\frac{d^2}{dz^2} = \lambda C \qquad (7)$$

$$D\frac{d^2C}{C} = \lambda dz^2 \quad (8)$$

$$D\int d\left(\frac{dC}{C}\right) = \lambda \int z dz \quad (9)$$

$$D\left(\frac{dC}{C} + m\right) = \lambda\frac{z^2}{2} + a \; (10)$$

The "m" and "a" terms in Eq. (10) are assumed to be a constant and are ignored.

$$D\, lnC = \lambda\frac{z^2}{2} \quad (11)$$

$$D\; 2lnC = \lambda z^2 \quad (12)$$

$$D = \lambda\frac{z^2}{2ln} \qquad (13)$$

$$D = \lambda\frac{z^2}{lnC^2} \qquad (14)$$

$$\mathbf{D} = \lambda\left(\frac{(X_2-X_1)^2}{\ln\left(\frac{C_1}{C_2}\right)^2}\right) \qquad (15)$$

2. EXPERIMENTAL METHODS

2.1. Sampling

In this work, soil samples were collected from the Bayındır district and its environment. The study area was placed on the Küçük Menderes basin where the dominant soil type is Non-Calcic Brown (Typic Haploxeralf). In this area, the soil was excavated to a depth of 50 cm and then soil samples were taken homogeneously. These samples transported to the laboratory, their wet and dry weight was recorded, and then samples sieved. Finally, each soil sample was placed in the experimental setup.

2.2. The Experimental Setup

The experimental setup of this study is installed based on the system was used in [19]. It consists of a cylindrical container, radium reach soil and a radon detector (Fig. 1). The collected soil sample is placed on the bottom of the cylindrical containers and the radon detector is located on the top. The soil in the container is charged as radon source and it is also media where radon motion is fairly low than open air. To show the differences among the soils related their radium content, five parallel setups was prepared. The first cylindrical container was filled with a soil sample and separated as background. The other four containers were supplemented with a liquid Radium ($RaCl_2$) standard solution at different activities (42, 422, 590 and 1688 pCi). In these containers, the soil was mixed thoroughly to homogenize samples. LR 115 Type 2 detectors were applied to measure alpha particles emitted from radon gas. A square piece of detector was fixed at the top of the container. Detectors were etched in 2.5 N NaOH solutions at 60 °C for 120 min and then, tracks were counted by using an optical microscope (LEICA DM 750), photographed by using a digital camera (LEICA ICC50) and LEICA Application Suite (LAS EZ) coupled to a PC. To reported result as activity unit calibration study was performed according to [20]. The detectors were changed within the new ones after a period of 4 weeks to complete to reach radioactive equilibrium between ^{226}Ra and its short-lived decay products. Measurements were repeated triple times to minimize standard error.

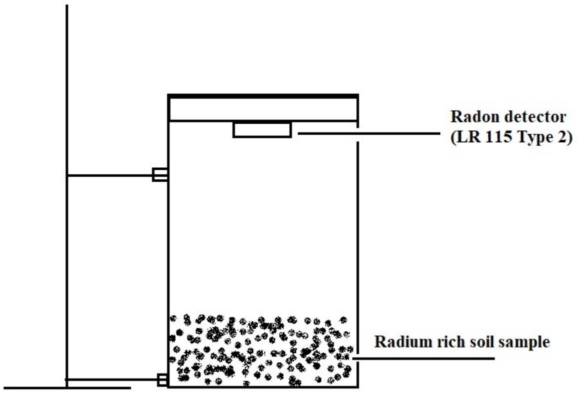

Fig. 1. System of radon diffusion through soil

3. RESULTS AND DISCUSSIONS

In this work, soil samples were collected form Küçük Menderes basin for measuring and calculating radon activity concentrations, radon diffusion coefficients and radon exhalation rates. Soil samples were taken from two different points which are defined as the non-calcic brown soil group. According to added radium activity level, the cylindrical container labelled as A (background), B (+42 pCi), C (+422 pCi), D (+590 pCi) and E (+1688 pCi), respectively. The measured radon activity concentration values and calculated parameters were shown in Table 1.

It has been found that soil gas radon activity concentrations were changed between 1.31 and 4.53 kBq m^{-3}. Also, there are not any significant differences between the two locations of soils. It is observed that radon activity levels strongly correlate with added radium activities. Radon diffusion coefficients of soils were calculated from the Eq. (15) and the diffusion coefficients vary from 1.3×10^{-8} and 9.3×10^{-7} m^2 s^{-1}.

Table 1. Radon activity concentration, diffusion coefficient and radon exhalation rates calculated from the soil of Küçük Menderes Basin

Sample	Radon Activity Concentration (kBq m^{-3})	Diffusion Coefficient (m^2s^{-1})	Diffusion Length (m)	Exhalation Rate ($Bqm^{-2}h$)
1A	1.31±0.35	$4.47x10^{-7}$	0.46	2.08
1B	2.12±0.48	$5.50x10^{-8}$	0.16	3.37
1C	4.21±0.82	$1.48x10^{-8}$	0.08	6.70
1D	3.22±0.53	$2.25x10^{-8}$	0.10	5.12
1E	4.53±0.79	$1.34x10^{-8}$	0.08	7.20
2A	1.03±0.26	$9.30x10^{-8}$	0.67	1.63
2B	1.60±0.67	$7.85x10^{-8}$	0.19	2.54
2C	1.66±0.54	$7.00x10^{-8}$	0.18	2.64
2D	3.02±0.76	$1.91x10^{-8}$	0.10	4.80
2E	3.96±0.15	$1.30x10^{-8}$	0.08	6.30

The studies related the radon diffusion coefficient shows that open-air radon diffusion coefficient is the upper bound as 1.2 $x10^{-5}$ m^2 s^{-1} and typically radon diffusion coefficient for soil with low moisture content is $x10^{-6}$ m^2 s^{-1} [1]. On the other hand, it is reported that the radon diffusion coefficient of gravel, sand and soil, as 7.59 $x10^{-6}$, 4.41 $x10^{-6}$ and 2.29 $x10^{-7}$ m^2 s^{-1} [19]. In another study was conducted in Russia, a new method is adopted for estimating radon diffusion coefficient and it was found that D varies from 1.4 to 24.4 x 10^{-7} m^2 s^{-1} [21]. Obtained results indicate that radon diffusion coefficients of soil collected from Küçük Menderes basin is lower than the values reported by the literature. It is possible to have several reasons for this difference. One reason for low coefficients can be the type of the soil samples. Non-Calcic Brown soil generally includes fine-grained particles and tight structure can prevent radon and/or other gasses pass. Diffusion length is varied from 7.9 to 66.5 cm while radon exhalation rates calculated between 1.63 and 7.20 Bq m^{-2} h.

4. CONCLUSIONS

In this study, soil gas radon diffusion coefficients were calculated by controlled laboratory experiments. Küçük Menderes Basin soils were collected and analyzed for this work. This study is the first study to be performed in this territory of the region in terms of radon diffusion. Results show that radon diffusion coefficients are relatively low. Also, radon diffusion length and exhalation rates were calculated. This study includes only Non-Calcic Brown group soils and it is planned to extend works on other soil types exists in this region.

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