Nuclear Engineering and Technology 51 (2019) 2013–2017

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

journal homepage: www.elsevier.com/locate/net

Original Article

Analysis of radon depth profile in soil air after a rainfall by using diffusion model

Seongjin Maeng [a, b], Seung Yeon Han [a], Sang Hoon Lee [a, *]

[a] School of Architectural, Civil, Environmental and Energy Engineering, Kyungpook National University, South Korea
[b] Radiation Science Research Institute, Kyungpook National University, South Korea

ARTICLE INFO

Article history:
Received 28 August 2018
Received in revised form
18 June 2019
Accepted 19 June 2019
Available online 21 June 2019

Keywords:
Radon
Soil
Rainfall
Diffusion model

ABSTRACT

The radon concentrations in soil air were measured before and after a rainfall. ^{226}Ra concentration, porosity, moisture content and temperature in soil were measured at Kyungpook National University in Daegu. As the results of measurement and analysis, the arithmetic mean of measured ^{222}Rn concentration increased from 12100 ± 500 Bq/m^3 to 16200 ± 600 Bq/m^3 after the rainfall. And the measured ^{226}Ra concentration was 61.4 ± 5.7 Bq/kg and the measured porosity was 0.5 in soil. The estimated values of ^{226}Ra concentration and porosity using diffusion model of ^{222}Rn in soil were 60.3 Bq/kg and 0.509, respectively. The estimated values were similar to the measured values. ^{222}Rn concentration in soil increased with depth and moisture content. The estimations were obtained through fitting based on the diffusion model of ^{222}Rn using the measurement values. The measured depth profiles of ^{222}Rn were similar to the calculated depth profiles of ^{222}Rn in soil. We hope that the results of this study will be useful for environmental radiation analysis.

1. Introduction

Radon (^{222}Rn) and Thoron (^{220}Rn) are Naturally Occurring Radioactive Material (NORM), which exist as inert gas at standard ambient temperature and pressure (SATP) and occurs mainly from soil, the wall of building which contain Uranium (^{238}U) and Thorium (^{232}Th) decay series. ^{222}Rn is the progeny of radium (^{226}Ra), one of the uranium decay series and has the half-life of 3.8 days which is alpha-emitting isotope. ^{220}Rn is the progeny of ^{224}Ra and has the half-life of 55.6 s also emits alpha rays. ^{222}Rn and ^{220}Rn are classified as carcinogens that cause lung cancer because the progeny of ^{222}Rn and ^{220}Rn that have moved to the lungs remain there and are constantly damaging [1]. So each country provides guideline on radon and regulates by indoor air condition [2–4].

Studies on environmental radioactivity and radon's behavior in soil have been carried out variously at domestic and overseas. The correlation research of ^{226}Ra and ^{222}Rn activities of same rock was studied by Hyomin et al. (2008) [5]. The radon behavior study in groundwater was conducted by analyzing the correlation between uranium and radon in groundwater [6]. In addition, to analyze outdoor radon, the study of ^{222}Rn exhalation rate from soil was conducted [7]. Since the move of ^{222}Rn in soil can influence the exhalation rate on the soil surface and indoor radon, researches on the diffusion model of ^{222}Rn in soil have been advanced [8–11].

In this study, to analyze ^{222}Rn concentration in soil before and after a rainfall, ^{222}Rn depth profile in soil and physical variables of soil were measured. ^{222}Rn depth profile of the diffusion model was calculated by applying the measured properties of soil. And ^{226}Ra concentration, porosity, moisture content in soil were estimated and compared with measured results.

2. Materials and methods

There are two types of instrument for measuring radon, passive detector and active detector. The passive detector that measures ^{222}Rn by exposing to the natural diffusion of gases requires relatively long measurement period, about a week to three months depending on cases. However, the active detector uses a pump to transport external gases into a detector chamber and measures ^{222}Rn or daughter nuclides (^{218}Po, ^{214}Po) by a semiconductor detector, Lucas Cell or an ionization chamber [12]. This study used RAD7 to measure ^{222}Rn concentration in soil. This active detector has advantages such as measuring ^{222}Rn and ^{220}Rn separately and having short measuring time. And a solid state detector which is

---

* Corresponding author.
E-mail address: lee@knu.ac.kr (S.H. Lee).

https://doi.org/10.1016/j.net.2019.06.018

made up a semiconductor to detect an alpha radiation was used at the instrument. The parameters of the instrument were shown in Table 1.

A dust filter of RAD7 was used to remove the general dust from soil gases. An inlet filter was used to prevent fine dust particles and all solid type radon daughters in sample gases from entering into the detector chamber. Since the measurement efficiency of this detector is sensitive to sample humidity in the detector chamber, sample gas was set to pass through a desiccant so that the soil gas humidity could be kept 4% or less.

In this study, RAD7 was set as shown in Fig. 1 to measure the ^{222}Rn in soil. Clay was used as shielding materials to prevent soil gas from spreading to air through the gap between soil and the soil gas probe. The measured soil depths were 15, 30, 45, 60, 75 and 105 cm by using a soil gas probe which can measure up to 120 cm depth. And the measurement setup was designed to allow the soil gases to pass through the dust filter, the laboratory drying unit and the inlet filter in order. The measurement cases were classified as wet case and dry case. Wet case was measured within 24 h after the rainfall, while dry case was measured at least 72 h after the end of the rainfall [13]. Temperature and moisture contents were measured at 10-min interval by a temperature smart sensor (Accuracy: $<\pm$ 0.2 °C from 0° to 50 °C) and TDR soil moisture sensor (Range: 0 ~ 100%, Accuracy: ± 0.03 m^3/m^3 typical in mineral soils) at the depth of 5 cm below the soil surface. In addition, to prevent the changes of temperature and environmental conditions of the measurement systems due to sunlight, a temporary covering was installed above the equipment and soil surface. The depth measurements were performed at one place (Kyungpook National University), where the surface is fairly flat and homogenous. Each measurement spot was spaced at least 1 m from previous measurement spot, also, the holes made by measurements were refilled with soil to prevent disturbances. For the wet or dry case, we measured at least twice and used averaged value.

To determine measurement time, a preliminary measurement was taken at each depth at 5-min intervals before main experiment. The results were shown in Fig. 2. The measurement time was determined by using the measured ^{222}Rn of depth 105 cm. By using exponential fitting, the equilibrium of ^{222}Rn concentration was estimated as 26500 Bq/m^3. The measured results of 5, 10, 15 min-measurements reached about 49% (12900 Bq/m^3), 82% (21800 Bq/m^3) and 99% (26200 Bq/m^3) of the equilibrium value. The other measured results were similar, except that the results of two depths (15 and 30 cm) which could be diluted with air. But the measurement results of all depths show similar trend. Based on these analysis results and the half-life of ^{218}Po (3.05 min), ^{222}Rn concentration in the inner chamber of RAD7 was assumed to be in equilibrium after 15 min (5 half-life). Therefore 15-min-measurement time interval was chosen. In main measurement, a pump was activated for 10 min to intensifying soil gases into an inner chamber before each depth measurements and measurements were performed twice at 15-min intervals for each depth. And then, the second interval result was used for soil depth profile analysis.

Table 1
The parameters of measurement instrument setup.

Parameter	Value	Unit
RAD7 Pump flow rate	0.6	Liter/min
Desiccant ($CaSO_4$)	8	mesh
RAD7 Detecting chamber volume	0.7	Liter
Inner diameter of soil gas probe	6.9	mm
Diameter of rod inserts of soil gas probe	4.6	mm

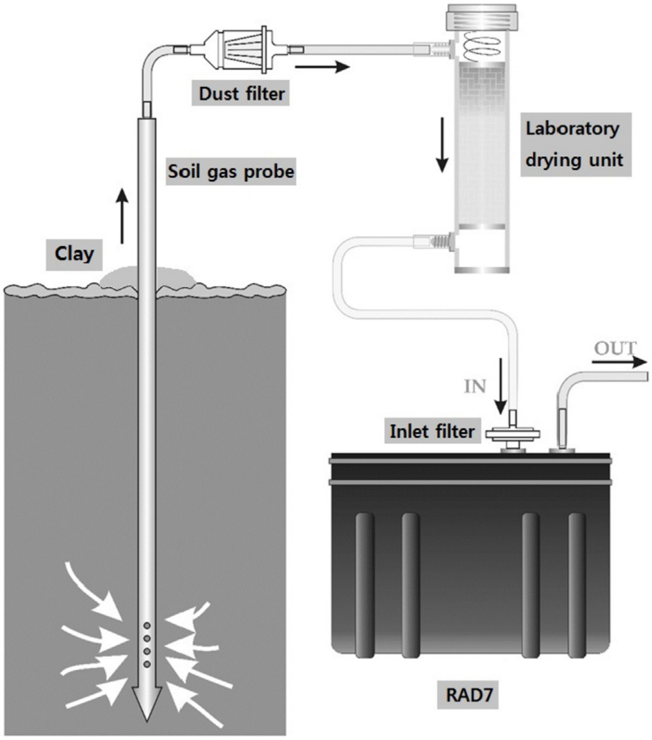

Fig. 1. An open-loop measurement setup for radon in soil gases using RAD7.

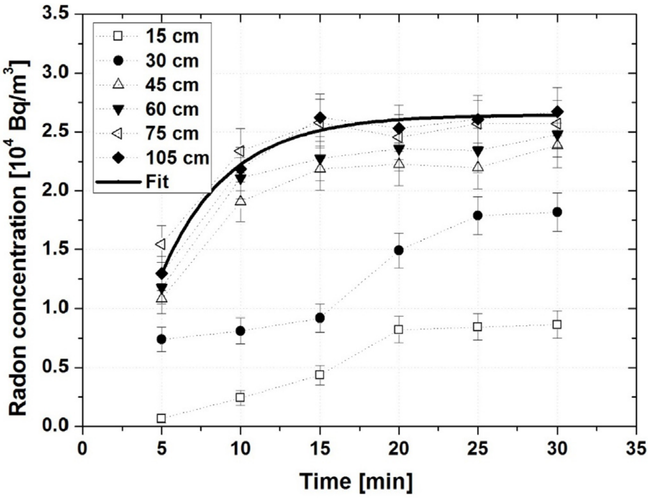

Fig. 2. Measurement results of radon concentration at each depth to determinate a measuring time.

2.1. Radon diffusion model

The diffusion model of Fick's law including the variables, bulk density, porosity, ^{226}Ra concentration in soil, temperature, moisture content, was used as analysis technique. This model consists of three terms, diffusion $\left(D_e\frac{\partial^2 C}{\partial z^2}\right)$, decay $(-\lambda C)$ and generation $\left(\frac{\lambda \cdot \rho_b \cdot A_{Ra} \cdot f}{\varepsilon}\right)$ of ^{222}Rn, as given:

$$\frac{\partial C(t,z)}{\partial t} = D_e\frac{\partial^2 C(z)}{\partial z^2} - \lambda C(z) + \frac{\lambda \cdot \rho_b \cdot A_{Ra} \cdot f}{\varepsilon} \tag{1}$$

where C(z) is ^{222}Rn concentration (Bq/m^3) in soil by depth, z is soil depth (meter), D_e is the diffusion coefficient in soil, λ is the physical decay constant of ^{222}Rn, ρ_b is the bulk density of soil, A_{Ra} is ^{226}Ra concentration (Bq/kg), ε is the porosity in soil, and f is the emanation fraction of soil [10,11].

The diffusion coefficient (D_e) of ^{222}Rn in soil was calculated by Eq. (2).

$$D_e(m,\varepsilon) = D_{a0}\cdot\varepsilon\cdot e^{\left(-6m\varepsilon-6m^{14\varepsilon}\right)} \tag{2}$$

where D_{a0} is the diffusion coefficient ($1.1 \cdot 10^{-5}(T/273)^{3/2}$ m^2/s) of ^{222}Rn in the air, m is the moisture content (fraction) in soil.

An emanation fraction of ^{222}Rn in soil is defined as the rate that generated ^{222}Rn from soil particles divided by pores. The emanation was calculated by Eq. (3).

$$f(m,T) = f_0\cdot\left\{1 + a\cdot\left[1-e^{(-bm)}\right]\right\}\cdot[1 + c\cdot(T-298)] \tag{3}$$

In this study, the arithmetical mean of measured temperature (T, K) were used for the emanation fraction calculation. The texture of measured soil was classified as sand based on U.S.A. regulations and international Guide [14], the constants which depend on the particle size were used, $f_0 = 0.1$, $a = 1.85$, $b = 18.8$, $c = 0.012$, as proposed by A.D. Griffiths [11].

By applying the boundary condition of ^{222}Rn concentration on the soil surface as C(z=0) = 0 to Eq. (1), the depth-dependent radon profile in soil can be derived as,

$$C(z) = C_\infty\cdot\left\{1-e^{\left(-\frac{z}{l_d}\right)}\right\} \tag{4}$$

where C_∞ is ^{222}Rn concentration (Bq/m^3) at deep depth ($z=\infty$) in soil, and l_d is the diffusion length ($(D_e/\lambda)^{1/2}$) of ^{222}Rn in soil.

^{222}Rn exhalation rate of measured results was calculated by using Fick's law as given below:

$$J(z) = -\varepsilon\cdot D_e\cdot\frac{\partial C}{\partial z} \tag{5}$$

where $\left(\frac{dC}{dz}\right)$ is a gradient of ^{222}Rn concentration on soil surface. This study, the gradient was based on the measured ^{222}Rn at soil depths from 15 cm to 45 cm. A theoretical exhalation rate was derived as Eq (6), by Eqs. (4) and (5),

$$J(0) = -\rho_b\cdot A_{Ra}\cdot f\cdot\sqrt{\lambda D_e} \tag{6}$$

where ^{226}Ra concentration and the bulk density of soil were the measured values. ^{222}Rn emanation fraction was calculated values by Eq. (3). D_e was calculated values by Eq. (2).

3. Results

^{226}Ra concentration in soil was 61.4 ± 5.7 Bq/kg, measured by a calibrated high purity germanium detector (HPGe detector), after heating the collected soil at 105 °C for 24 h. The bulk density of soil was 1.333 g/cm^3. Porosity was 0.5. The arithmetical mean of temperatures was 295.15 K (23 °C) during the measurement. According to data from a daily report of Korea Meteorological Administration, the rainfall intensities before the 3rd and the 4th measurements were 6.5 mm, 27.0 mm, respectively. The measurement results of soil parameters are given in Table 2.

As a result, the arithmetical mean of the measured ^{222}Rn concentration in soil before the rainfall was 12100 ± 500 Bq/m^3 and was 16200 ± 600 Bq/m^3 after the rainfall. The trends of the measured ^{222}Rn concentration depth profile were like an exponentially saturating function which is $a(1-e^{-bz})$. As shown in Fig. 3, the ^{222}Rn concentration increased with the depth after the rainfall. The mean of ^{222}Rn concentration of each depth; 15, 30, 45, 60, 75 and 105 cm increased 125%, 51%, 63%, 20%, 20% and 26% after the rainfall. The average growth rate was 51% after the rainfall. And ^{222}Rn exhalation rate calculated by Eq. (5) decreased from 7.0 x 10^{-2} Bq/m^{2}s to 4.5 x 10^{-2} Bq/m^{2}s. The measured moisture contents

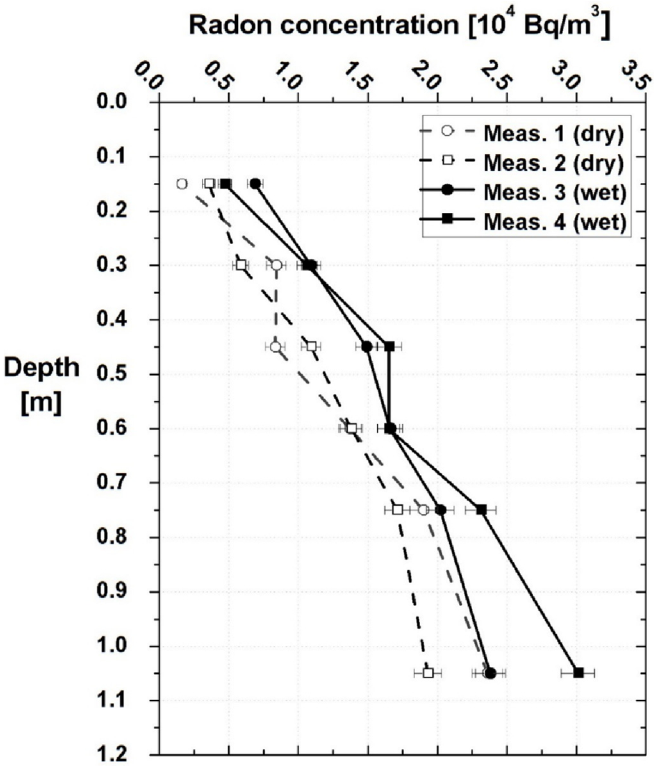

Fig. 3. Concentration of ^{222}Rn as a function of soil depth for dry and wet cases.

Table 2
Measurement results of the soil parameters and the radon concentrations in the soil.

	Date	Moisture content [m^3/m^3]	Temp. [K]	Mean of ^{222}Rn [Bq/m^3]	dC/dz [Bq/m^4]
Meas. 1 (dry)	2017.09.15	0.023	298.5	12400 ± 400	25800 ± 4900
Meas. 2 (dry)	2017.09.18	0.033	295.5	11700 ± 400	23100 ± 5300
Avg. of dry cases	-	0.028	297.0	12100 ± 500	24500 ± 7200
Meas. 3 (wet)	2017.10.07	0.28	296.7	15500 ± 400	26700 ± 6400
Meas. 4 (wet)	2017.10.13	0.26	294.0	16900 ± 400	39300 ± 6400
Avg. of wet cases	-	0.27	295.3	16200 ± 600	33000 ± 9100

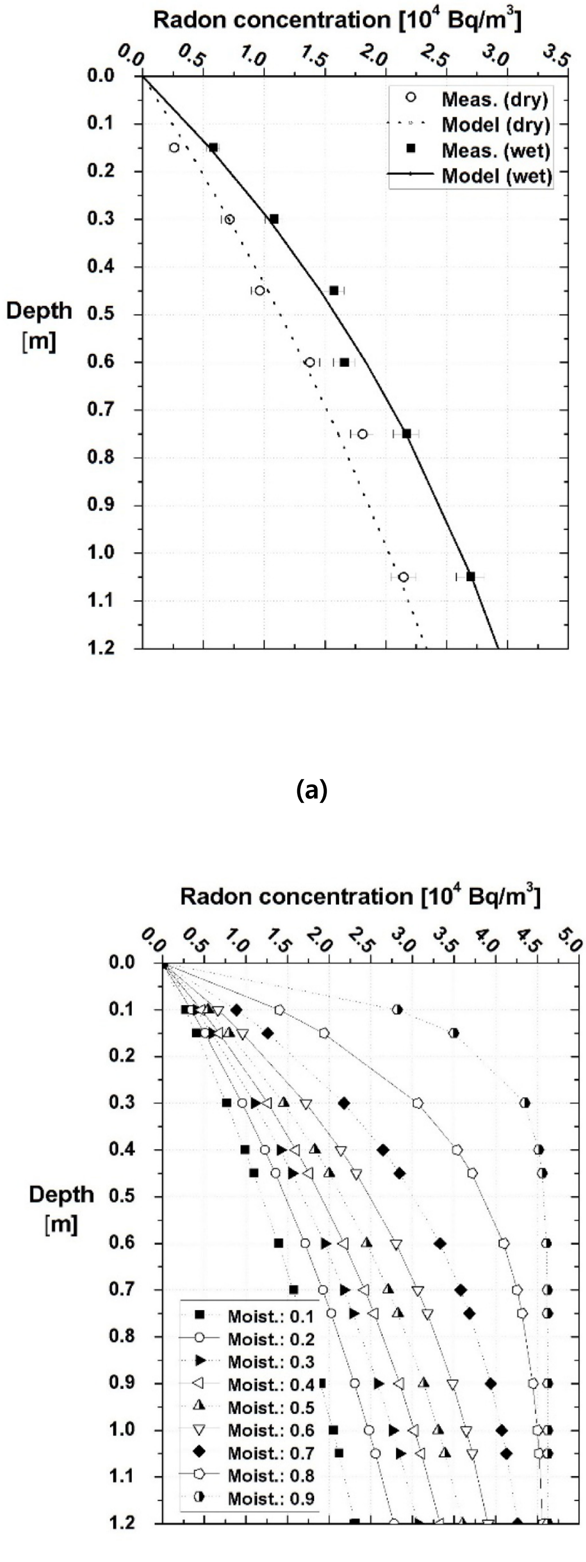

Fig. 4. The radon depth profiles for the measured cases (a) and the calculated radon depth profile based on the ^{222}Rn diffusion model with different moisture contents (b).

were 0.028 m^3/m^3 (dry case), 0.27 m^3/m^3 (wet case), possibly caused by water diffusion in soil [14].

To compare the calculated depth profile of ^{222}Rn with the measured depth profile of ^{222}Rn, the depth profile of the model was calculated by substituting the measured parameters; ^{226}Ra concentration, the moisture content and others. In order to compare the estimated soil properties with the measured properties, ^{226}Ra concentration, moisture content and porosity in soil were estimated using the measured depth profile by applying the least squares method of the ^{222}Rn diffusion model. The estimated value of ^{226}Ra concentration, 60.3 Bq/kg, was similar with the measured ^{226}Ra concentration (61.4 ± 5.7 Bq/kg). The estimated moisture contents, 0.083 (dry case) and 0.283 (wet case) and the estimated porosity (0.509) were similar with the measured data. And the estimated moisture contents are comparable to the values of U. Karstens (2015), which are in the range of 0.098–0.345 [10].

For calculating the ^{222}Rn depth profile by using the diffusion model, the estimated moisture contents were used because the moisture contents measured at 5 cm below the soil surface don't represent the actual soil moisture contents. Therefore, the measured values were used as the parameters for calcuation except the moisture content. And the calculated ^{222}Rn depth profiles were shown in Fig. 4 (a) with the measurement results of each case. The correlation coefficients of the results were 0.994 (dry case) and 0.992 (wet case). The calculated ^{222}Rn depth profiles by using the diffusion model on moisture contents, from 0.1 to 0.9, were shown in Fig. 4 (b). The saturation of radon depth profile has a strong dependence on the soil moisture. Generally, the higher soil moisture increases the emanation fraction, and decreases the diffusion coefficient. The changes in these two physical parameters combine to give early saturated depth profile (saturation reached at shallow depth) for high moisture for example $m = 0.9$ given in Fig. 4 (b). In model calculation result, the saturated radon concentrations from the diffusion model were 46200 Bq/m^3 when the soil moisture is over 0.3. And the saturation depths range from 0.3 m (soil moisture $m = 0.9$) to 7 m (soil moisture $m = 0.3$).

To estimate the exhalation rate of ^{222}Rn, the exhalation rates of the measured cases based on the diffusion model were calculated using Eqs. (5) and (6), on the assumption that the porosity, the temperature and the other measured variables are constants. The calculated exhalation rates of the model and the estimated exhalation rates with measured data are shown in Fig. 5. The estimated

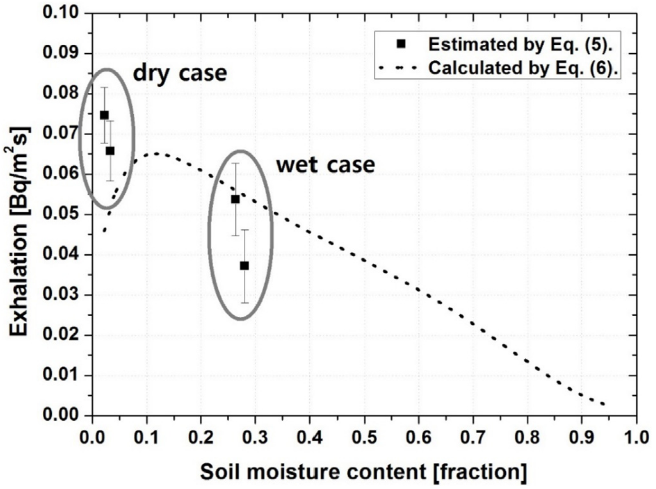

Fig. 5. The estimated exhalation rates based on measured radon gradient of the soil surface and the calculated exhalation rates of the diffusion model on soil surface.

exhalation rates of ^{222}Rn were 7.58 x 10^{-2}, 6.47 x 10^{-2} Bq/m^{2}s for dry cases and 5.51 x 10^{-2}, 3.61 x 10^{-2} Bq/m^{2}s for wet cases, which are similar with the measurement range of the radon exhalation rate, 0.77 x 10^{-2} Bq/m^{2}s to 7.32 x 10^{-2} Bq/m^{2}s as proposed by Weihai Zhuo [9]. According to the diffusion model, considering the variation of radon concentration due to the amount of radium, the temperature, and the moisture content, these results are reasonable values. And the exhalation rates calculated by Eq. (6) were also similar with results of A. D. Griffiths research [11].

4. Conclusions

In this study, we measured the soil depth profiles of ^{222}Rn before and after rainfall at Kyungpook National University and calculated ^{222}Rn profile by using the diffusion model in soil. The arithmetical mean of measured ^{222}Rn concentrations increased from 12100 ± 500 Bq/m^3 to 16200 ± 600 Bq/m^3 after the rainfall. The measured porosity was 0.5 in soil. The measured moisture content of soil surface was 0.028 (dry case), 0.272 (wet case) after the rainfall. The ^{226}Ra concentration measured from HPGe gamma spectroscopy was 61.4 ± 5.7 Bq/kg.

And the estimated exhalation rate of ^{222}Rn decreased after the rainfall due to the moisture contents increase as studied by Nazaroff [15]. The estimated ^{226}Ra concentration was 60.3 Bq/kg. The moisture contents were 0.083 of dry case and 0.283 of wet case. And the estimated porosity of soil was 0.509. The estimates were obtained through fitting based on the diffusion model using the measurements. The estimated values were similar with the measured values of soil properties.

This study allows us to estimate soil properties of some location by using the ^{222}Rn diffusion model with the measured ^{222}Rn depth profile. The authors will continue radon depth profile study of soil air and apply for other locations for advanced studies. We hope that this study would be useful in the environmental radiation research field.

Acknowledgments

This work was supported by K-CLOUD project “Measurement and analysis of radon exhalation rates from soil”, KOREA HYDRO & NUCLEAR POWER CO., LTD (2017-TECH-08).

References

[1] World Health Organization, Who Handbook on Indoor Radon: A Public Health Perspective, 2009.

[2] Health Canada, Guide for Radon Measurements in Residential Dwellings (Homes), 2017.

[3] A. EOA, Citizen’s Guide to Radon: the Guide to Protecting Yourself and Your Family from Radon, 2005.

[4] Korea Environment Corporation, Environment Technology, Indoor Radon Management, 2016.

[5] Hyomin Lee, et al., Distribution of some environmental radionuclides in rocks and soils of guemjeong-gu area in busan, Korea, Jour. Petrol. Soc. Korea 17 (3) (2008) 179–190.

[6] Chan Ho Jeong, et al., Occurrences of uranium and radon-222 from ground waters in various geological environment in the hoengseong area, J. Eng. Geol. 25 (No.4) (2015) 557–576.

[7] Weihai Zhuo, Masahide Furukawa, Qiuju Guo, Yoon Shin Kim, Soil radon flux and outdoor radon concentrations in East Asia, Int. Congr. Ser. 1276 (2005) 285–286.

[8] Vanessa Haverd, Matthias Cuntz, Soil-Litter-Iso: a one-dimensional model for coupled transport of heat, water and stable isotopes in soil with a litter layer and root extraction, J. Hydrol. 388 (2010) 438–455.

[9] Weihai Zhuo, I.D.A. Takao, Masahidc Furukawa, Modeling radon flux density from the earth’s surface, J. Nucl. Sci. Technol. 43 (No.4l) (2006) 479–482.

[10] U. Karstens, C. Schwingshackl, D. Schmithuesn, I. Levin, A process-based 222radon flux map for Europe and its comparison to long-term observations, Atmos. Chem. Phys. 12 (2015) 12845–12864.

[11] A.D. Griffiths, W. Zahorowski, A. Element, S. Werczynski, A map of radon flux at the Australian land surface, Atoms. Chem. Phys. 10 (2010) 8969–8982.

[12] Mark Baskaran, Radon, A Tracer for Geological, Geophysical and Geochemical Studies, Springer Geochemistry, 2016, pp. 37–40.

[13] Kyoochul Ha, Yongcheol Kim, Sung-Yun Kim, Monitoring of soil water content and infiltration rate by rainfall in a water curtain cultivation area, J. Geol. Soc. Korea 52 (No. 3) (2016) 221–236.

[14] United States Department of Agriculture, Soil survey manual: soil science division staff, in: Agriculture Handbook No. 18, 2017, p. 121.

[15] William W. Nazaroff, Radon transport from soil to air, Rev. Geophys. 30 (1992) 137–160.