Determination of Radium and Radon Exhalation Rate as a Function of Soil Depth of Duhok Province - Iraq

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1. Introduction

Radon ($^{222}$Rn) is a naturally occurring radioactive inert gas produced during the decay process of radium, when alpha particles start to be emitted, turning the radium into radon. The rate of radon gas that escapes from the soil into the atmosphere is called radon exhalation rate. In this study, radon, radium, and both radon surface and mass exhalation rates were measured for 40 samples of soil at four sampling depths (10, 20, 30, and 40 cm) in three districts of Duhok province. For the radon measurement, alpha-sensitive RAD7 detector was used. While radium concentration measured by well type NaI(Tl) detector. Analysis, shows radon surface exhalation rate vary from 24 ± 10.7 to 57 ± 2.5 Bq.m$^{-2}$.h$^{-1}$ with an average value of 38.7 ± 8.9 Bq.m$^{-2}$.h$^{-1}$. While mass exhalation rate vary from 1.2 ± 0.9 to 6.7 ± 1.6 Bq.kg$^{-1}$.h$^{-1}$ with an average of 4.2 Bq.kg$^{-1}$.h$^{-1}$. Furthermore, the results showed that radon exhalation rate and radon concentrations in soil have direct proportion to soil depths. Overall, radon concentration, radium content and both surface and mass exhalation rate in all sample points present a good correlation and less than global mean average recommendation [30].

2. Material and methods

2.1. Study area

This study has been done on province of Duhok, located in the north-west of Iraq. It has borderline with Syria from the west, Turkey from the north, Nineveh from the south, and Erbil city from the east. The study area is divided officially into four districts. The altitudes are quite different, ranging from 300 to more than 1300 meter above the sea level with three different northern, northeastern and western types of wind. The geographical locations of the study points are shown and tabulated in the Figure 1.
The geological composition formed of red beds of silt, limestone, and hard clay. Conglomerate and regain is existed within the low folded area. The composition of the soil throughout investigation area is mostly characterized by thick sedimentary cover, broad synclines and well-marked folds of asymmetrical antclines, very low permeability, and moisture of sediment of depth from (surface to 10 cm) of 6–7% (Abdulah & Ramadhan, 2011).

2.2. Sample collection and preparation

A total of 40 soil samples were carefully gathered in 10 locations of Duhok governorate. Four measurement points were taken in Duhok district, three in Sumel and three points in Amadi district. At each sample point soil was taken in depths 10 cm, 20 cm, 30 cm, and 40 cm. Measurements were taken in July (dry season). In each point an area of about 50 cm x 50 cm was remarked and accurately cleared of any sediment or debris depending on a standard methodology (Abdulah & Ramadhan, 2011).

It was found that radon exhalation rate has a strong dependence on the moisture and temperature of the soil (Soares et al., 2020). Radon exhalation rate had a tendency to increase when the soil moisture content below 8%, While it decrease steadily when the soil moisture content exceeded 8% (Maeng et al., 2019; Soares et al., 2020). The theory is as follows: Water works to promote radon exhalation up to a particular moisture level. Water keeps more radon if the water content exceeds a certain level, and consequently, exhalation is then restrained (Hosoda et al., 2008). Much research has been conducted concerning the relationship between the soil radon exhalation rates and the soil moisture content (Maeng et al., 2019) and effect of rainfall (Masahiro et al., 2007). In many studies radon exhalation rate in summer season is higher than any other seasons because of the variation in soil moisture, which evidently affect the seasonal variation in soil air radon concentration (Hosoda et al., 2010; Maeng et al., 2019; O.B. Modibo et al., 2011). Therefore, this research adopts dry soil to eliminate the effect of moisture content and rainfall. Thus, the collected samples of soil dried at room temperature for four days then dried in an oven at 110°C for one hour. Then the dried samples were sieved through 250 mm mesh to obtain a homogeneous powder (Amin et al., 2018).

In preparation to measure $^{222}\text{Rn}$, a mass of 125 g from each of the soil samples were stored in a cup of cylindrical plastic tube. Each plastic tube contain of two valves, the dimension and the valves of the plastic containers shown in Figure 2. These plastic tubes stored for one month to acquire secular equilibrium between $^{222}\text{Rn}$ and its progeny (Ismail & Jaafar, 2010).

While to estimate concentration of $^{226}\text{Ra}$ in soil, smaller tubes of about 13 cm$^3$ (Figure 3) were used. These tubes were filled with soil and also stored for one month to acquire secular equilibrium between $^{226}\text{Ra}$ and its progeny.

2.3. Radon activity measurement

In this work the solid-state detector RAD7 was used to measure $^{222}\text{Rn}$ concentration in soil. The benefit of this detector is the measurement of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ concentration separately and having short measurement time. The measurement set up is shown in Figure 4; it consists of a cylindrical plastic tube, RAD7 Professional Electronic Radon Detector and a vinyl tube that contained desiccant (CaSO$_4$). When plastic tube connected to the closed loop, both valves of the plastic tube are opened. The accumulated radon gas will pass from the plastic tube to the desiccant, after that to the inlet filter of RAD7. The air is leaving from the outlet of RAD7. Inside RAD7 air is decayed and detects alpha particle
that emitted from polonium isotopes. Then alpha technique is used by RAD7 which convert alpha particle right away to electrical sign. Also this detector capable to separate the different between electrical pulses generated from $^{210}$Po and $^{214}$Po with energies 6 MeV and 7.69 MeV, respectively.

The experiment is done in a dry condition, with humidity less than 8% (by purging RAD7 sometimes before starting the test). Then RAD7 is setup onto four cycle mode, each mode about one hour. Finally, the data were transferred from RAD7 to computer. Then, the obtained spectrum was analyses by Capture software.

For quality assurance and tracking the background radiation, radon concentration is measured for a blank container following the same manner as for the soil samples in term of starts, stops and time of exposure.

2.4. Estimation of $^{222}$Rn exhalation rate

After the determination of radon concentration, surface and mass exhalation rate of $^{222}$Rn can be calculated using equations 1 and 2 (Amin et al., 2018).

$$E_x = \frac{\lambda V C_{Rn}}{A \left[ T + \left( e^{-\lambda T} - 1 \right) / \lambda \right]}$$ (1)

$$E_M = \frac{\lambda V C_{Rn}}{M \left[ T + \left( e^{-\lambda T} - 1 \right) / \lambda \right]}$$ (2)

Where: $\lambda$ is the radon decay constant (0.00756 h$^{-1}$), $T$ is the exposure time in hours, $V$ is the volume (0.00104 m$^3$) (the difference between the volume of the container and the volume of the sample), and $A$ is the surface area of the sample (0.014 m$^2$), $M$ is the mass of the soil.
2.5. Radium concentration measurement

In the same time, the activity per unit mass of $^{226}\text{Ra}$ is performed from the examined samples by a gamma-ray spectrometry system consisting of a well-type $3'' \times 3''$ NaI(Tl) scintillation detector. For limiting and reducing background radiation, the detector is surrounded by 4m geometry lead shield with thickness 6 cm and additional shield of 2 mm of electrolytic copper with an upper opening to change the source. In the center of lower segment of lead shield has a small hole about 5 cm in diameter to catch the detector inside the shield. While, photomultiplier tube was wrapped by a thin plastic sheet to decrease the noise of electrical signal created by the shield and avoid direct shield-detector contact. Detector arrangement parameters are shown in Figure 5.

The spectrum is analyzed by using MAESTRO Multi Channel Analyzer (MCA) application software. Energy, resolution, and efficiency calibration of the detector had been calibrated using gamma standard radioactive sources. Channel number convert to energy scale by using photo-peak of the standard International Atomic Energy Agency (IAEA) radioactive sources $^{153}\text{Eu}$, $^{228}\text{Na}$, and $^{60}\text{Co}$ that emit gamma-ray energy of 105.3, 511, 1173, and 1332.49 keV. Figure 6 shows the relation between channel number and energy spectrum of standard sources. After identification of Photo-peak energies with the corresponding channel numbers of standard sources, the same multi sources are used to determine the efficiency ($\varepsilon$) of the detector (Figure 7), by using the following equation (Ramadhan & Abdullah, 2018):

$$\varepsilon = \frac{N}{A \times f_y \times t} \quad (3)$$

Where, $N$ is the net peak area obtained for each Photo-peak, $A$ is the activity value of each radioisotope source at the time of the measurement, $t$ is the time during which the spectrum was acquired, and $f_y$ is the probability of gamma disintegration of the radionuclide; the energy, activity, and probabilities emission of gamma of each standard source shown in Table 1.

After NaI(Tl) was calibrated intern of energy resolution and efficiency, soil sources spectrum were analyzed. Based on the $\gamma$-ray spectrometer counts ($N$) at

![Figure 5](image-url)  
**Figure 5.** (a) the $3'' \times 3''$ well-type NaI(Tl) detector inside the open shield, (b) sample tube, (c) changing the sample, and (d) the combined shield that houses the detector and the PMT that passes through the bottom of the shield.

![Figure 6](image-url)  
**Figure 6.** Energy spectrum verses channel number (source placed inside the well of the NaI(Tl) detector).

![Figure 7](image-url)  
**Figure 7.** Efficiency of the NaI(Tl) detector.
full width half maximum photo-peaks channels of 351.9 keV of $^{214}$Pb and 609 keV of $^{214}$Bi decay products, the activity per unit mass of $^{226}$Ra in samples is calculated as the average of both $^{214}$Bi and $^{214}$Pb radioisotopes (Abdulah & Ramadhan, 2011).

### Table 1. The present activities, energies, and probability of gamma of the used radioactive sources.

| Standard sources | A (Bq) | E (keV) | $\epsilon$ (%) |
|------------------|--------|---------|----------------|
| $^{137}$Eu       | 640    | 105.3   | 21.1           |
| $^{222}$Na       | 1176   | 511     | 180            |
| $^{60}$Co        | 24,498 | 1173.26 | 99.85          |
|                  |        | 1332.49 | 99.98          |

### Table 2. Measurements of Soil $^{222}$Rn concentration, $^{226}$Ra content, area exhalation $E_a$ and mass exhalation $E_m$ of soil samples; Duhok district (points 1–4), Somel district (points 5–7), and Amed district (points 8–10).

| Points | Depth (cm) | $^{222}$Rn (Bq. m$^{-3}$) | $^{226}$Ra (Bq. kg$^{-1}$) | $E_a$ (Bq. m$^{-3}$ h$^{-1}$) | $E_m$ (Bq. kg$^{-1}$ h$^{-1}$) |
|--------|------------|---------------------------|---------------------------|-----------------------------|---------------------------------|
| Point 1| 40         | 341                       | 28                        | 5.6                         | 51                              |
|        | 30         | 300                       | 26                        | 5.0                         | 45                              |
|        | 20         | 290                       | 26                        | 4.8                         | 43                              |
|        | 10         | 275                       | 20                        | 4.5                         | 41                              |
|        | 40         | 273                       | 36                        | 4.5                         | 41                              |
|        | 30         | 270                       | 33                        | 4.5                         | 40                              |
|        | 20         | 255                       | 30                        | 4.2                         | 38                              |
|        | 10         | 184                       | 22                        | 3.0                         | 27                              |
|        | 40         | 384                       | 50                        | 6.4                         | 57                              |
|        | 30         | 380                       | 48.6                      | 6.3                         | 56                              |
|        | 20         | 403                       | 48                        | 6.7                         | 60                              |
| Point 3| 10         | 362                       | 44                        | 6.0                         | 54                              |
|        | 40         | 320                       | 31                        | 5.3                         | 48                              |
|        | 30         | 300                       | 30                        | 5.0                         | 45                              |
|        | 20         | 290                       | 30                        | 4.8                         | 43                              |
|        | 10         | 250                       | 27                        | 4.1                         | 37                              |
|        | 40         | 290                       | 33                        | 4.8                         | 43                              |
|        | 30         | 285                       | 31                        | 4.7                         | 42                              |
|        | 20         | 285                       | 25                        | 4.7                         | 42                              |
|        | 10         | 200                       | 25                        | 3.3                         | 30                              |
|        | 40         | 281                       | 24                        | 4.6                         | 42                              |
|        | 30         | 275                       | 20                        | 4.5                         | 41                              |
|        | 20         | 261                       | 19                        | 4.3                         | 39                              |
| Point 6| 10         | 190                       | 18                        | 3.1                         | 28                              |
|        | 40         | 310                       | 28                        | 5.1                         | 46                              |
|        | 30         | 290                       | 26                        | 4.8                         | 43                              |
|        | 20         | 292                       | 26                        | 4.8                         | 43                              |
|        | 10         | 285                       | 23                        | 4.7                         | 42                              |
|        | 40         | 243                       | 22                        | 4.0                         | 36                              |
|        | 30         | 190                       | 18                        | 3.1                         | 28                              |
|        | 20         | 148                       | 13                        | 2.4                         | 22                              |
| Point 8 | 10        | 72                        | 8                         | 1.2                         | 10                              |
|        | 40        | 280                       | 25                        | 4.6                         | 42                              |
|        | 30        | 210                       | 23                        | 3.5                         | 31                              |
| Point 9 | 10        | 198                       | 18                        | 3.3                         | 29                              |
|        | 10        | 120                       | 14                        | 2.0                         | 17                              |
|        | 40        | 300                       | 28                        | 5.0                         | 44                              |
|        | 30        | 225                       | 26                        | 3.7                         | 33                              |
| Point 10| 10        | 200                       | 20                        | 3.3                         | 29                              |

### 3. Results and discussion

The activity concentrations of $^{222}$Rn, $^{226}$Ra, and surface and mass radon exhalation rate $E_a$ and $E_m$ for different depths of soil samples from Duhok governorate were determined and represented in Table 2.

The mean soil depth activity of $^{222}$Rn and $^{226}$Ra in Duhok district ranged from 246 ± 41 to 382 ± 17 Bq. m$^{-3}$ and 25 ± 3.4 to 48 ± 2.6 Bq. kg$^{-1}$, with an average of 305 ± 44 Bq. m$^{-3}$ and 33 ± 5 Bq. kg$^{-1}$ respectively. The mean soil depth activity of $^{222}$Rn in Somel district ranged from 252 ± 63 to 294 ± 11 Bq. m$^{-3}$ with an average of 270 ± 13 Bq. m$^{-3}$ and of $^{226}$Ra 20 ± 5 to 29 ± 4 Bq. kg$^{-1}$ with an average of 25 ± 2.3 Bq. kg$^{-1}$, respectively. Furthermore, the minimum and maximum mean of radon concentration in each sample point in Amed district are 163 ± 72 to 221 ± 58 Bq. m$^{-3}$, respectively with a mean of 196 ± 17 Bq. m$^{-3}$ While radium content ranged from 15 ± 6 to 23 ± 6 Bq. kg$^{-1}$ with a mean of 19.3 ± 2 Bq. kg$^{-1}$.

It can be noted from the results that the concentration of $^{222}$Rn and $^{226}$Ra in soil samples varied appreciably from one points to another. Maximum radon concentration and radium content were found at Duhok District (points 3) while minimum recorded at Amed district (point 8) about 80 km to the north-east from point 3. The variation may be due to the geo-chemical process in soil or geological condition of locations (El-Araby et al., 2019; Jayasheela et al., 2013; Shashikumar et al., 2008). Many researchers found a positive correlation between radon concentration and radium concentration (Jayasheela et al., 2013; Shashikumar et al., 2008). In the present study the same trend is observed as shown in Figure 8. A positive correlation has been observed between radon and radium concentration with correlation coefficients of 0.98. This means that, radium content in soil is the main source of radon.

![Figure 8. Correlation between average of $^{222}$Rn and $^{226}$Ra concentration at each different points.](image-url)
Furthermore, the outcomes show that the activity concentrations of radon and radium content in the investigated soil samples were found to be directly correlated with the soil depth (10 cm – 40 cm), except the point 3 (in this point radon concentration in depth 20 cm is more than that in 30 and 40 cm).

Figure 9 shows the variation of $^{222}$Rn concentration as a function of soil depths in three different districts of Duhok provinces. The outcomes suggest that the maximum soil gas radon observed at depth 40 cm and minimum at 10 cm. The obtained results from the present study indicate that the concentration of radon gas in soil increases with the soil depth. This is because of the variation of radium content, soil grain size, and soil porosity. Present results show behavior which agrees with that findings of many other researchers (Amin et al., 2018; Antonopoulos-Domis et al., 2009; El-Zohry et al., 2017; Kaliprasad & Naryana, 2018).

Figure 10 illustrates the correlations between exhalation rate of radon and average concentration of radon and radium in each soil samples while Figure 11 shows the relation between surface and mass radon exhalation rate. Statistical t-test demonstrates that a strong positive correlation has been observed between radon exhalation rate with radon concentration ($R^2 = 1$). In the same manner as $^{222}$Rn and $^{226}$Ra, the highest value of $E_x$ and $E_M$ recorded in point 3 while lowest value measured in point 8. This is because there is a direct relationship between level of radon and its exhalation (Amin et al., 2018; El-Araby et al., 2019; Shashikumar et al., 2008). $^{222}$Rn exhalation rate depend on radioactive disintegration of $^{226}$Ra to produce radon, the moisture condition of the soil in the vicinity of the escaped radon atom, the direction of recoil of radon gas in the grain and its diffusion in the pore space (Elzain, 2015a). Hence, it is clear that there is a positive relation between radon exhalation with the concentrations of both radon and radium in soil (El-Araby et al., 2019; Elzain, 2015b). Finally, Figure 12 shows the average radon exhalation rate intern of area at each sample points.

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**Figure 9.** Average radon concentration in the three districts as a function of soil depth.

**Figure 10.** Relationship between radon exhalation rate and radon concentration at Duhok province (radon concentration at each point is the average of all different depths).

**Figure 11.** Relationship between area and mass radon exhalation rate (exhalation at each point is the average of all different depths).

**Figure 12.** The average of radon area exhalation rates for 10 soil sample points.
The results of this study are broadly in agreement with the research carried out throughout the world. Table 3 shows comparison between present work and other local study (Ismail & Jaafar, 2010). Also the comparison is carried out with nine of the worldwide investigations about same topic.

The total average values of $^{222}$Rn, $^{226}$Ra, $E_X$, and $E_M$ in Duhok provinces were 263 Bq.m$^{-3}$, 26.5 Bq.kg$^{-1}$, 39 Bq.m$^{-2}$.h$^{-1}$, and 4.2 Bq.kg$^{-1}$.h$^{-1}$ respectively and all of them below the average global values of 300 Bq.m$^{-3}$, 35 Bq.kg$^{-1}$, and 57.6 Bq.m$^{-2}$.h$^{-1}$, respectively (Abbasb et al., 2020). As a whole, the study has confirmed that soil gas radon and $^{226}$Ra content in the study area and consequently the associated radon exhalation does not expose the human health to associated risk.

**4. Conclusion**

Radium and Radon exhalation rates for soil have been measured in three districts of Duhok province by alpha and gamma spectroscopy. A strong correlation was observed between radon gas in soil and radon surface and mass exhalation rate. From this study, it is clear that, there is a positive correlation between radon exhalation with the concentrations of radon and radium in soil. Furthermore, the present study indicate that the soil-gas radon increases with soil depth which might be due to the radium content, soil grain size, and soil porosity. The results show that the value of radon concentration, radium content, and radon exhalation rates are less than the mean world value.

**Disclosure statement**

No potential conflict of interest was reported by the author(s).

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