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Relationship between radon concentration and physicochemical parameters in groundwater of Erbil city, Iraq

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ABSTRACT
This study portrays the relationship between Radon activity concentration and physicochemical parameters in groundwater in Erbil city, Iraq. Primary data of groundwater samples were collected from five different zones in Erbil, during February 2019. Water samples were collected for both Radon measurement and physicochemical analysis. Water samples were taken from 24 different wells as a reference, then directly measured their radioactivity at the laboratory of Erbil Environment Office. Dissolved Radon 222Rn in a sampled groundwater has been measured using an electronic Radon detector RAID 7. Determination of various parameters such as pH, total dissolved solids, total hardness, Sulfate, and Magnesium in groundwater has also been measured. Moreover, regression analysis illustrated that there is no significant relationship observed between pH, TDS, and hardness. Determination of various parameters such as pH, total dissolved solids, total hardness, Sulfate, and Magnesium with Radon activity concentration because these are no significant at the 95% confidence level. Otherwise, the correlation of determinate (R²) values show the relationship between Sulfate and Magnesium with Radon concentration because these are significant at the 95% confidence level. All measurements were done in almost identical environmental conditions to avoid the minor differences in meteorological parameters.

1. Introduction
Water is a crucial natural resource for most life on the planet (Srilatha et al., 2014). However, while water covers approximately 70% of our planet’s surface, only 0.3% of the total water resources on our planet are portable and suitable for daily life. Freshwater is an essential and vital element for all living organisms, and even for survival (Akar et al., 2012). The human race accesses both surface water and groundwater. Dissolved chemical species exist naturally in surface and groundwater as a result of leaching from rocks that are in contact with the water. However, the sum of the concentrations of dissolved chemical species, i.e. Calcium (Ca²⁺), Magnesium (Mg²⁺), Sodium (Na⁺), Potassium (K⁺), Hydrogen carbonate (HCO₃⁻), Carbonate (CO₃²⁻), Sulfate (SO₄²⁻), Chloride (Cl⁻), Fluorine (F⁻), Nitrate (NO₃⁻), and Silicon tetra hydroxide dissolved in the water can negatively affect the water quality of daily life. The number of water cations (Magnesium, Sodium, etc.), anions (Sulfates, Chloride, etc.), and radionuclides (Potassium, Radium, Thorium, etc.) are variable in the groundwater (Reddy et al., 2017). The radioactive nuclides in groundwater are much higher than the surface water because it passes through soil formations and rock, dissolving many radiochemical compounds and minerals (Akar et al., 2012; Srinivasa et al., 2018). For instance, when groundwater moves naturally through radium isotopes bearing soil and rocks, the radiochemical element radium is dissolved and transported with the water.

Radon is a radioactive, colorless, odorless gas that is soluble in water (Inácio et al., 2017; Keramati et al., 2018; Ramola et al., 2008). Radon’s half-life is 3.8 days (Kumar et al., 2016; Malakootian & Nejhad, 2017). Therefore, areas that are underlain by granites and similar rocks containing comparatively more uranium typically have higher average count rates of Radon (Shilpa et al., 2017). In general, the voids and fractures from rocks and soil cause Radon to move from its source to enter the environment. Consequently, the Radon released by the decay of radioactive elements can quickly dissolve in groundwater and be carried to water-supply wells. Previously, several studies have used Radon as a natural tracer to investigate whether the surface water infiltrates aquifers in the same area. However, it has rarely been applied to estimate the residence time of groundwater because of its short half-life (Choubey et al., 2003). The quality of groundwater is equally important as its quantity owing to the suitability of water for daily life. The changing quality of
groundwater in an area is a function of chemical and physical parameters that are profoundly affected by anthropogenic activities and geological formations (Asabere & Bempah, 2013; Kaliprasad & Narayana, 2018).

Recently, many researchers have been working diligently to evaluate Radon activity concentration in different environments all over the world to control and reduce their multiple effects on human health (Duggal et al., 2013; Mansour et al., 2005). Motivated by such R & D efforts, the prime objective of our study is to evaluate the concentration level of Radon in groundwater source used for drinking and to examine the relationship between Radon activity concentration with physicochemical parameters in groundwater of Erbil city, Iraq.

2. Study of the area

Erbil city is located in the north of Iraq at 36.19°N-latitude and 44.009°E-longitude, with an altitude of 414 m above sea level. Erbil is in the middle of a large high hill called the plain of Erbil. The western boundary of the plain of Erbil is close to the Tigris River, and the northern and north-eastern parts of the city are high and comprise of Hasarost mountains (Hameed, 2013). Geologically, Erbil city is a flat area that is wholly covered with alluvial deposits which lithologically consists of post-granulated clay, sand, and silt (Ramola et al., 2008). Erbil’s climate may be classified as subtropical with a semi-arid continental climate, with large temperature variability between seasons. The summer months (June, July, and August) are quite hot and dry. The average temperature is around 43 °C during summer days and the temperature reaches as high as 48 °C on the hottest day (Qadir & Saeed, 2011). The winter months (December, January, and February) can be cold and wet, with an average temperature around 2 °C and the lowest temperature may fall to around −5 °C (Qadir & Saeed, 2011). The annual average rainfall is around 300–400 millimeters (Choubey et al., 2003). The average relative humidity is 80% and 20% in winter and summer, respectively, with slow wind speed at about 3–5 annually (Asabere & Bempah, 2013). These natural background radiations can vary significantly depending on the geological and environmental factors. In view of this, in the present investigation, an attempt was made to measure Radon concentration and Radon inhalation rate in the water wells in Erbil city, in order to assess the radiation dose delivered to the public, and to examine the relationship between $^{222}$Rn Radon concentrations with physicochemical parameters in groundwater of Erbil city, Iraq.

3. Materials and method

3.1. Sample collection

Water samples were taken from the environments of a water well used as a source for drinking water. A total of 24 samples from different locations in Erbil city were collected for investigation during February 2019. And the rest of the locations/points use main pipe water (Ifraz Project), and others have the same geological materials according to the geological map of Erbil City (Gardi, 2017). Glass water bottles 250 ml and 500 ml were used for collecting groundwater samples for both Radon measurement and physicochemical analysis, respectively. The bottles were thoroughly washed prior to sample collection and were later properly labeled. Typically, the wells were purged for 10 min through pumping to ensure sample quality. All the water samples were collected in the special glass bottle of 500 ml for physicochemical analysis and 250 ml capacity bottles designed primarily for measuring Radon activity in water to avoid minimum loss of Radon by defacing and any air contact. After sampling, the water samples were brought to the laboratory (with minimal loss of time and keep their temperature below 20 °C) and analyzed by analytical techniques available in the laboratory, immediately. Data obtained from the experiment were analyzed utilizing statistical tools to analyze significant relationship and correlations between variables. In this research, ($R^2$) was used to determine the significant relationship between Radon concentration with physicochemical parameters.

3.2. Measurement of radon concentration

The water samples were analyzed using an electronic Radon detector RAD 7. A RAD/H2O accessory was connected to the RAD 7 to measure the concentration of Radon in the collected samples. The sample bottles of 250 ml have been connected to the RAD 7, and the internal air pump of the Radon monitor has been used for re-circulating a closed air-loop through the water sample, purging Radon from the water into the air-loop. The air has been re-circulated through the water continuously to extract the Radon until the RAD/H2O system reaches the equilibrium state. Finally, the initial Radon concentration of the respective water sample has been calculated by using the Radon activity concentration measured in the air-loop. The RAD 7 detector recalibrated recently in January 2019 to achieve the standard calibration accuracy, as Environmental Protection Agency (EPA) recommends for all continuous Radon instruments. The setup diagram of RAD/H2O is shown in (Figure 1 (Durridge Company I, 2011)).
3.3. Physicochemical analysis

The investigated parameters were mainly pH, total dissolved solid (TDS), total hardness, Sulfate (SO$_4^{2-}$), and Magnesium (Mg$^{2+}$) ions using standard analytical techniques available at the laboratory of Erbil Environment Office.

3.4. pH

pH measurement shows the acidity or alkalinity of a water sample. pH is the negative logarithm of the hydrogen ion concentration or algebraically $pH = -\log_{10} [H^+]$. $orpH = \log_{10}[H^+]$. It is the main method frequently employed in drinking water quality monitoring, for example, via, laboratory quality pH meter and the electrode, pH multi-parameter probes, and pH paper strips (Müller et al., 2018).

3.5. Total dissolved solids (Tds)

TDS was determined by using a digital conductivity meter. TDS is the total weight of all solids that are dissolved in a provided water volume, expressed in the unit of parts per million (PPM) and unit of a milligram per unit volume of water (mg/l) (Jeyaraj et al., 2019).

3.6. Total hardness

Measuring total Hardness in the water sample was determined by titration method which required 25 ml distilled water and 25 ml of water sample into a 250 ml Erlenmeyer flask, followed by 0.02 gm of Irochrom and 4 drops of buffer solution. The contents were later titrated with Ethylenediaminetetraacetic acid (EDTA) solution. The titration was continued till the color change from purple to blue and calculate the total Hardness; Result = 1.25mltitration $\times$ 40 (Ferreira et al., 2019).

3.7. Sulfate SO$_4^{2-}$

Sulfate amount in the water samples was determined by the turbid metric method (Khan et al., 2020).

3.8. MagnesiumMg$^{2+}$

The Magnesium amount in the water samples was determined by the portable photometer (Banerji & Mitra, 2019; Nnaji et al., 2019).

4. Results and discussion

A total of 24 groundwater samples have been collected from different neighborhoods (Newroz, Karezan, Badawa, Shexahmed, Brayaty, Raparen, Mufti, Runaki, Eskan, Zanko Galawezh) of Erbil city. A screenshot of the ArcGIS program including the different sample locations on the Erbil Master Plan Map is shown in (Figure 2). The Radon activity concentration values are groundwater at various locations in the study area are given in (Table 1). All measurements have been made in similar climatic and environmental conditions during February 2019, so that the variation in these
meteorological parameters was small. The temperature is between 16.4 and 22.5 °C. The Radon concentration in water varied from 4 to 12.18 Bq/l, this is due to the geological material of these specific zones. The mineralized area with high concentrations of radium which include uranium minerals and granitic rocks would expect to show elevated radon activity concentration in groundwater (Choubey & Ramola, 1997), which is within the proposed safe limit of 4–40 Bq/l by the United Nations Scientific Committee on the Effects of Atomic Radiation (Nations, 2011). Also, the annual mean effective doses of groundwater samples due to ingestion and inhalation have been calculated by using the parameters established in the reference (Mittal et al., 2016). Ingestion could be calculated by Eq. 1.

\[ E_{w,i} (\mu Sv y^{-1}) = C_{Rn,w} \times C_W \times (EDC) \] (1)

where \( E_{w,i} \) is the effective ingestion dose, \( C_{Rn,w} \) is the activity concentration of Radon in water (Bq/l), \( C_W \) is the weighted estimated of water consumption (730 l/y), and \( (EDC) \) is the ingestion Effective Dose Coefficient 3.5 nSv/Bq. And inhalation could be calculated by Eq. 2, as below.

\[ E_{w,h} (\mu Sv y^{-1}) = C_{Rn,w} \times R_{a,w} \times F \times O \times (DCF) \] (2)

where \( E_{w,h} \) is the effective inhalation dose, \( C_{Rn,w} \) is the activity concentration of Radon in water (Bq/l), \( R_{a,w} \) is the ratio of Radon in the air to Radon in tap water \( (10^{-4}) \), \( F \) is the factor of equilibrium between Radon and its decay products \( (0.4) \), \( O \) is the average

Figure 2. Screenshot of ArcGIS platform showing the sample locations on Erbil city map – northern Iraq.
Table 1. Radon activity concentration in groundwater samples of some Erbil neighborhoods.

| Sample location | Water well no. | Depth/m | Temp.°C | Radon activity concentration/Bq/L | Mean annual effective dose/μSv y⁻¹ |
|-----------------|----------------|---------|---------|----------------------------------|----------------------------------|
| Newroz          | 17             | 157     | 18.5    | 6.44                             | 16.45, 16.23, 32.68               |
| Newroz          | 13             | 300     | 18.5    | 11.32                            | 28.92, 28.53, 57.45              |
| Newroz          | 7              | 150     | 17      | 5.21                             | 13.31, 13.13, 26.44              |
| Newroz          | 5              | 120     | 16.4    | 4                                | 10.22, 10.08, 20.30              |
| Newroz          | 12             | 143     | 17.3    | 4.17                             | 10.65, 10.51, 21.16              |
| Karezan         | 15             | 338     | 17.9    | 9.68                             | 24.73, 24.39, 49.12              |
| Badawa          | 20             | 300     | 18.8    | 12.18                            | 31.12, 30.69, 61.81              |
| Badawa          | 18             | 250     | 19.1    | 8.71                             | 22.25, 21.95, 44.20              |
| Badawa          | 14             | 200     | 19.4    | 9.31                             | 23.79, 23.46, 47.25              |
| Badawa          | 9              | 175     | 19.4    | 9.17                             | 23.43, 23.11, 46.54              |
| Shexahmed       | 2              | 300     | 18.5    | 11.2                             | 28.62, 28.22, 56.84              |
| Brajati         | 6              | 150     | 18.8    | 5.78                             | 14.77, 14.57, 29.34              |
| Raparen         | 14             | 150     | 19.1    | 7.5                              | 19.16, 18.90, 38.06              |
| Raparen         | 10             | 150     | 19.1    | 7.66                             | 19.57, 19.30, 38.87              |
| Mufri           | 8              | 500     | 18.8    | 10.5                             | 26.83, 26.46, 53.29              |
| Mufri           | 3              | 300     | 19.7    | 11.5                             | 29.38, 28.98, 58.36              |
| Mufri           | 2              | 160     | 20      | 6.49                             | 16.58, 16.35, 32.93              |
| Runaki          | 6              | 300     | 19.7    | 6.49                             | 16.58, 16.35, 32.93              |
| Eskan           | 1              | 120     | 19.7    | 8.41                             | 21.49, 21.19, 42.68              |
| Zanko           | 12             | 300     | 22.5    | 4.82                             | 12.32, 12.15, 24.47              |
| Zanko           | 3              | 137     | 22.5    | 4.31                             | 11.01, 10.86, 21.87              |
| Galawezh        | 8              | 300     | 22.5    | 10.65                            | 27.21, 26.94, 54.05              |
| Galawezh        | 10             | 300     | 20.3    | 5.21                             | 13.31, 13.13, 26.44              |
| Galawezh        | 9              | 300     | 20      | 9.42                             | 24.07, 23.74, 47.81              |

| Range           | 120–300        | 16.4–22.5| 4–12.18 | 10.22–31.12 | 10.08–30.69 | 20.30–61.81 |
| Average         | 233.3          | 16.4–22.5| 7.92     | 20.24        | 19.96        | 40.20        |

door occupancy time per person (7000 h/y), and (DCF) is the Radon exposure Dose Conversion Factor for 9 nSv/h.(Bq/m³). In (Table 1), it is clear that the annual mean effective dose due to ingestion changes from 10.22 to 31.12 μSv y⁻¹ with a mean value of 20.24 μSv y⁻¹. The inhalation annual means of the effective dose of groundwater changes from 10.08 to 30.69 μSv y⁻¹ with a mean value of 19.96 μSv y⁻¹. Moreover, the minimum to a maximum value of total dose estimated due to Radon concentration in groundwater is 20.30 μSv y⁻¹ and 61.81 μSv y⁻¹ respectively, with an average value of 40.20 μSv y⁻¹ as shown in (Table 1). The total annual effective dose of all the studied groundwater samples is found to be fine within the proposed safe limit of 0.1 mSv y⁻¹ by World Health Organization (WHO) and European Council (EU) (Mittal et al., 2016). In (Table 2) a comparison of Radon concentration in studied water samples from various cities of (Mosul (Najam, 2014), Akashat (Tawfik, 2013), Najaf (Abjojassim et al., 2015), Baghdad (Kadhim, 2015), Babel (Kadhim et al., 2016), and Sulaymaniyah (Youssuf & Abdullah, 2011)) with Erbil (Present work) in Iraq is given. It can be seen that the concentration of Radon in water samples of Akashat and Sulaymaniyah might be in close agreement with the present work. From which it can be seen that the Radon concentration in water samples of Najaf, Baghdad, and Babel lower than the present work. However, the concentration of Radon in Mosul, is higher than the present investigation.

Physicochemical parameters of drinking water, for example, pH, total dissolved solids, Hardness, Sulfate, and Magnesium have been measured to evaluate the influence on Radon concentration. pH values at all zones were acceptable when compared against the WHO standards for groundwater wells in Erbil – Iraq as shown in (Figure 3). pH values of the collected water samples varied between 7.6 and 8.3. The higher pH is a frequent result of high bicarbonate and carbonate concentrations (Idoko & Oklo, 2012). The R² values in (Figure 3) showing there are no relationship between pH values and Radon concentration because these are no significant at the 95% confidence level. This is mainly due to the fact that Radon is an inert gas (Srilatha et al., 2014). TDS results are acceptable at all zones when compared to the WHO standards for groundwater wells in Erbil, Iraq. TDS of groundwater wells are all within the WHO permissible limit of 2000 μS/cm for drinking ground-water. TDS test provides many dissolved ions in groundwater and is employed as an indicator test to show the general groundwater quality (Asabere & Bempah, 2013). TDS indicates the nature of water quality for salinity (Srilatha et al., 2014). High TDS value influences the taste and corrosive property of the water (Srilatha et al., 2014).

Table 2. Activity concentration of radon in groundwater samples of different cities in Iraq.

| City-Iraq     | Radon activity concentration/Bq/L |
|---------------|----------------------------------|
| Mosul         | 17.4 ± 0.8to35.1 ± 1 ± 2         |
| Akashat       | 8.02 ± 0.1to11.7 ± 0.16           |
| Najaf         | 0.0432 ± 0.003to98.876 ± 0.226    |
| Baghdad       | 0.2890 ± 0.072                   |
| Babel         | 0.54 ± 0.49to7.92 ± 0.52          |
| Sulaymaniyah  | 7.5390 ± 0.011.184               |
| Erbil         | 4to12.18                         |
Figure 3. Radon activity concentration vs pH value.

Figure 4. Radon activity concentration vs TDS.

$R^2$ values in (Figure 4) Showing there are no relationship between TDS and Radon concentration because these are not significant at the 95% confidence level. This is because Radon has no smell, taste, or color (Shuttleworth, 2018). In addition, the Hardness values are acceptable at all zones when compared against the WHO standards for groundwater wells in Erbil, Iraq. The hardness values of the collected water samples varied between 164 and 352 mg/l. Hardness is a potent substance in drinking water, and it contains the sum of the polyvalent cations and is expressed as an equivalent amount of CaCO$_3$ (Musa & Ahanonu, 2013). $R^2$ values in (Figure 5) illustrates no relationship between Hardness and Radon concentration because these are no significant at the 95% confidence level. This is mainly due to the fact that Radon is an inert gas (Idriss et al., 2011; Srilatha et al., 2014). Also, Kupwade & Langade point out that the Sulfide minerals add the soluble Sulfate into the underground water through the oxidation procedure. Sulfate concentrations in groundwater well samples can be seen in (Figure 6). Concentrations of Sulfate are within the WHO guide range and are acceptable at all Erbil well zones. On the other hand, $R^2$ values in (Figure 6) demonstrate the relationship between Sulfate and Radon concentration because these are significant at the 95% confidence level. Furthermore, Magnesium values are acceptable at all zones when compared against the WHO standards for groundwater wells in Erbil Iraq as shown in (Figure 7). Magnesium values of the collected water samples varied between 4.86 and 34.06 mg/l. $R^2$ values in (Figure 7) points out the relationship between Magnesium and Radon concentration because these are significant at the 95% confidence level.

Figure 5. Radon activity concentration vs hardness.
Conclusion

This study demonstrated that the activity concentrations of Radon in all groundwater samples in some locations of Erbil city are comparable with the similar study carried out in other parts of Iraq. Radon concentrations in all groundwater samples in Erbil are within the proposed safe limits by the United Nations Scientific Committee on the Effects of Atomic Radiation. Although in Erbil there are no standards for drinking water with regard to Radon concentration. However, it seems, that the total annual effective dose from the 24 groundwater locations of the studied area is determined to be fine and within the safe limit of 0.1 mSv·yr⁻¹ proposed by WHO. We presented that all parameters were acceptable for drinking water at all zones when compared to the WHO standards for groundwater wells in Erbil, Iraq. Regression (R²) values (0.0711 and 0.0809, respectively) show the relationship between Sulfate and Magnesium with Radon activity concentration because these are significant at the 95% confidence level. In contrast, R² values (0.0194, 0.0036, and 0.0129, respectively) show that there is no relationship between pH, TDS, and Hardness with Radon activity concentration because these are not significant at the 95% confidence level.

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Disclosure statement

The authors declare that there is no conflict of interest.

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Figure 6. Radon activity concentration vs $SO_4^{2-}$.  

Figure 7. Radon activity concentration vs $Mg^{2+}$.  

5. Conclusion

The authors declare that there is no conflict of interest.
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