



Assessment of environmental radiological hazards associated with elevated levels of radon gas in drinking water

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ABSTRACT

The purpose of this study was to assess the environmental radiological hazards associated with elevated levels of radon gas in drinking water. Using passive (CR-39 plastic detector) and active (RAD 7) detectors, the radon levels in the air and drinking water (tap water) of 124 homes in 31 distinct locations in Iraqi Kurdistan were determined. Almost anywhere with a high indoor radon level also has a high radon exhalation rate from drinking water. The cities of Similan and Kelak had the highest and lowest radon concentrations using both techniques. A good correlation was found between the values of the average indoor radon concentration for active and passive detection methods. The values are smaller than the internationally recommended limit of 11 Bq/l as proposed by the Environmental Potential Agency (US EPA).

1. Introduction

Alpha particles are primarily generated from the radioactive decay of nuclei such as uranium, thorium, actinium, radium, radon, and thoron, as well as transuranic elements, which are chemical elements with an atomic number greater than or equal to 92. In other words, elements having a mass number of approximately 150 can spontaneously emit alpha particles (NRCNA (National Research Council of the National Academies), 2006). Moreover, radon gas ($^{222}_{88}\text{Rn}$) can be produced through the alpha decay of radium ($^{226}_{88}\text{Ra}$), whereby a sufficient number of atomic nuclei is required for alpha particle decay to take place. As alpha particles are emitted, the nucleus may become excited, leading to the production of gamma rays from the nucleus in order to release the excess energy (Ismail, 2015). Since both the alpha particle and the remainder of the nucleus are positively charged, they repel each other through the Coulomb repulsion process (Maher et al., 2006).

Alpha particles possess a high linear energy transfer (LET) and can travel a distance of up to 7.5 cm in the air, with the majority of their energy being dissipated over a short range. Therefore, ingestion and inhalation of particles that emit alpha radiation can present a radiation

hazard (NRCNA (National Research Council of the National Academies), 2006; Ismail & Jaafar, 2011a). Radon (^{222}Rn) and thoron (^{220}Th) are two natural radioactive gases that are both colorless, odorless, and tasteless (Ismail & Jaafar, 2010d). Upon decay, they emit alpha particles that can deposit onto the lungs, trachea, and stomach, thereby posing a significant health risk (Ismail, 2011; Aswood et al., 2022).

Since radon is an inert gas with limited solubility in bodily fluids, it is diffused evenly throughout the body. There are health concerns, including cancer, associated with exposure to this gas and its solid decay products, polonium-218 and -214 (Ismail & Jaafar, 2011b). By the time the decay products reach the lung, they have undergone further radioactive decay, releasing tiny bursts of energy in the form of alpha particles that may break down DNA or generate free radicals. In addition, radon is also associated with potentially fatal effects on developing fetuses (NRCNA (National Research Council of the National Academies), 2006; Ismail & Jaafar, 2011b). Radon and its short-lived decay products, when breathed in, produce alpha particles that become the primary source of radiation in lung tissues. These decay products, particularly those that are attached to small aerosols or are not attached at all, can harm sensitive lung cells, leading to an increased risk of cancer

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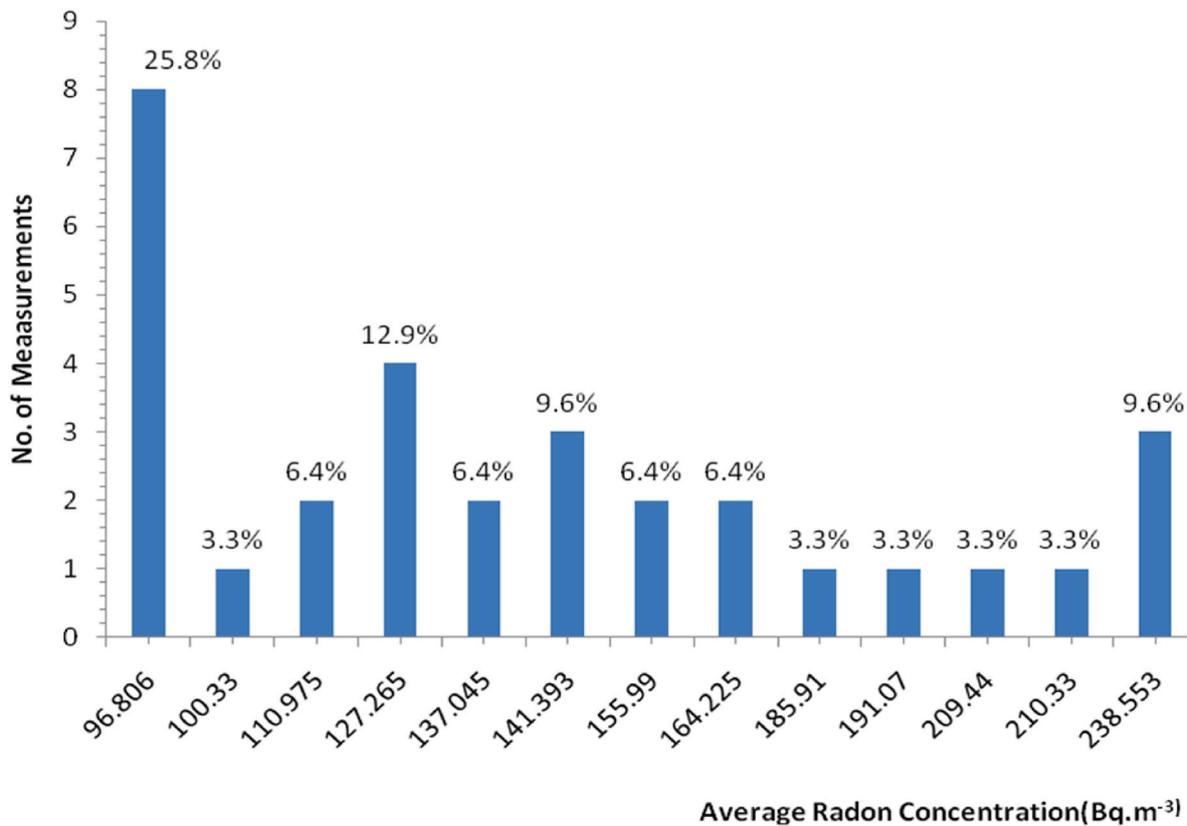


Fig. 1. Average indoor radon concentration in Iraqi Kurdistan; histogram of frequency distribution, lognormal and percentages.

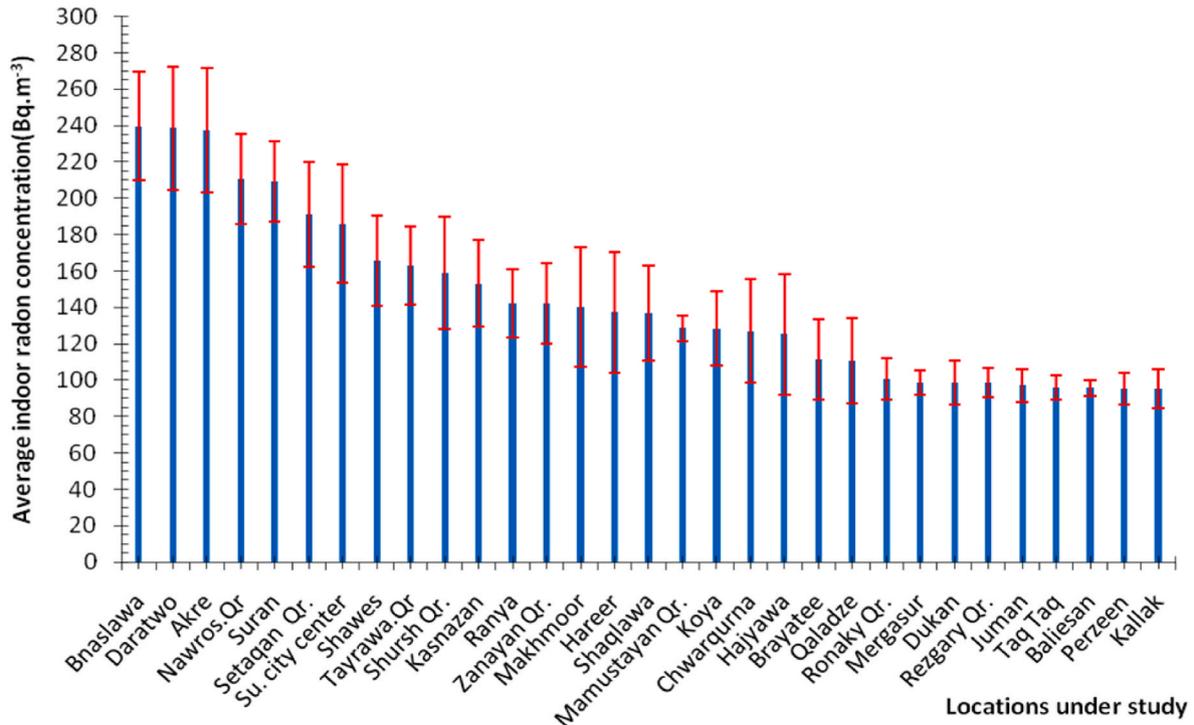


Fig. 2. The average radon concentration among the investigated locations.

development (Field, 2011; Ismail et al., 2011; Mole et al., 1990). In 1979, the World Health Organization (WHO) brought to light the health impacts of being exposed to residential radon via a European working group on indoor air quality. Additionally, radon has also been classified

as a human carcinogen by the International Agency for Research on Cancer (IARC) (World Health Organization (WHO), 2009; IARC (International Agency for Research on Cancer), 1988).

The purpose of this study was to assess the potential environmental

Table 1

The levels of indoor radon concentration and the associated risk factors in the dwellings under investigation in Iraqi Kurdistan.

Locations	C _{Rn} (Bq/ m ³)	Potential Alpha Energy Concentration (PAEC) mWL	H _E (mSv/ y)	Lifetime Risks %	Lung Cancer Risks (CPPP)
Akre	237.61 ± 66.11	25.68 ± 5.22	5.99 ± 2.01	2.376	107.86 ± 30.11
Baliesan	95.59 ± 33.23	10.33 ± 2.65	2.41 ± 1.02	0.956	43.39 ± 15.33
Bnaslaw	239.65 ± 77.32	25.91 ± 6.12	6.04 ± 2.31	2.396	108.79 ± 32.18
Brayatee	111.41 ± 38.69	12.04 ± 3.07	2.81 ± 0.98	1.114	50.57 ± 17.45
Chwarqurna	126.96 ± 43.33	13.72 ± 3.22	3.20 ± 1.11	1.269	57.63 ± 18.02
Daratwo	238.4 ± 76.54	25.77 ± 5.77	6.01 ± 2.27	2.384	108.22 ± 31.73
Dukan	98.55 ± 32.94	10.65 ± 2.11	2.48 ± 1.01	0.985	44.73 ± 116.04
Hajyawa	125.31 ± 37.62	13.54 ± 3.08	3.16 ± 1.13	1.253	56.88 ± 17.31
Hareer	137.22 ± 43.72	14.83 ± 3.55	3.46 ± 1.16	1.372	62.29 ± 20.11
Juman	97.12 ± 33.18	10.50 ± 2.41	2.45 ± 1.1	0.971	44.09 ± 15.62
Kallak	95.11 ± 34.75	10.28 ± 2.83	2.4 ± 0.9	0.951	43.17 ± 15.22
Kasnazan	153.11 ± 37.21	16.55 ± 3.42	3.86 ± 1.12	1.531	69.50 ± 21.92
Koya	128.34 ± 35.87	13.87 ± 3.69	3.23 ± 1.11	1.283	58.26 ± 19.01
Makhmoor	140.13 ± 38.11	15.15 ± 3.88	3.53 ± 1.17	1.401	63.61 ± 20.64
Mamustayan Qr.	128.45 ± 41.33	13.88 ± 3.42	3.24 ± 1.04	1.284	58.31 ± 19.1
Mergasuor	98.76 ± 32.78	10.67 ± 2.55	2.49 ± 0.93	0.987	44.83 ± 16.44
Nawros.Qr	210.33 ± 62.36	22.74 ± 4.33	5.30 ± 2.11	2.103	95.48 ± 28.51
Perzeen	95.22 ± 34.22	10.29 ± 2.43	2.40 ± 0.96	0.952	43.22 ± 15.62
Qaladze	110.54 ± 33.44	11.95 ± 2.11	2.78 ± 1.03	1.105	50.18 ± 17.67
Ranya	142.16 ± 40.42	15.36 ± 4.62	3.58 ± 1.14	1.421	64.53 ± 20.77
Rezgary Qr.	98.44 ± 32.89	10.64 ± 2.22	2.48 ± 0.93	0.984	44.68 ± 16.01
Ronaky Qr.	100.33 ± 33.77	10.84 ± 3.01	2.53 ± 0.97	1.003	45.54 ± 16.33
Setaqan Qr.	191.07 ± 39.77	20.65 ± 5.03	4.82 ± 2.30	1.910	86.74 ± 26.77
Shaqlaw	136.87 ± 37.13	14.79 ± 4.01	3.45 ± 1.18	1.368	62.13 ± 21.31
Shawes	165.63 ± 38.26	17.90 ± 4.88	4.17 ± 2.03	1.656	75.19 ± 23.22
Shursh Qr.	158.87 ± 36.32	17.17 ± 4.92	4.0 ± 2.0	1.588	72.12 ± 20.66
Su. city center	185.91 ± 38.21	20.1 ± 5.87	4.69 ± 2.12	1.859	84.39 ± 26.88
Suran	209.44 ± 43.58	22.64 ± 5.11	5.28 ± 2.23	2.094	95.07 ± 27.96
Taq Taq	95.66 ± 39.63	10.34 ± 2.36	2.41 ± 0.95	0.956	43.42 ± 14.87
Tayrawa.Qr	162.82 ± 37.87	17.60 ± 4.00	4.10 ± 1.65	1.628	73.91 ± 20.77
Zanayan Qr.	141.89 ± 38.74	15.34 ± 3.33	3.57 ± 1.19	1.419	64.41 ± 15.08
Average ±SDV.	143.77 ±46.10	15.54 ±4.98	3.62 ±1.16	1.437 ±0.46	65.266 ±20.93

risks associated with dissolved radon gas in domestic water due to an upsurge in airborne radon gas. The study analyzed the concentration of radon in both tap water and air from 124 homes located in 31 different areas of Erbil governorate using passive (CR-39 plastic detector) and active (RAD 7) detectors.

2. Material and methods

2.1. Materials

2.1.1. CR-39NTDs

It is a polymer made of C₁₂H₁₈O₇ with a density of 1.31 g/cm³ and a thickness of 700 m, and each piece is 1 cm × 1.5 cm and has a laser-engraved code. The calibration of nuclear track detector types CR-39 (CR-39NTDs) is necessary to identify their efficiency in registering alpha particles. Registration and detection of alpha particles by the CR-39NTDs depend on the chemical etching condition (type of chemical solution, its normality, its temperature, and the time of etching). So, the calibration considers a preliminary test must be done for this type of detector. The calibration has been done in our previous work (Ismail, 2009). The chemical etching condition for the applied CR-39NTDs was identified by 6N NaOH at 70 °C for 9 h of etching.

2.1.2. Water bath

The CR-39 NTDs were etched using a "GOTECH TESTING MACHINES INC." water bath, model GT-7039-M, 220 V, 50 Hz. It has an automatic control system and a blending system.

2.1.3. Scanning system of tracks of alpha particles

The setup comprises a high-precision microscope equipped with an auto-focus function, a positioning stage, and a CCD camera. The camera and positioning stage are linked to a computer that runs specialized pattern recognition software.

2.1.4. RAD7

The RAD7 radon detector version 7 manufactured by DURRIDGE CO. was employed to ensure precise long-term measurements and calibration.

2.2. Experimental procedures

2.2.1. Radon in drinking water using CR-39NTDs technique

Using this detection approach, 62 samples (excluding one damaged sample) from 21 major regions were tested, and all samples were obtained directly from tap water. In each area, three samples were collected to calculate the mean value and standard deviation. Samples of drinking water are collected in plastic bottles (tightly closed). Both active and passive techniques have been developed to monitor airborne radon gas in drinking water of the collected samples (Ismail, 2008; Ismail & Jaafar, 2010b). A closed and open diffusion chamber of radon gas that was equipped with pieces of CR-39 NTDs was used as a passive detection method. The detector chamber, with a radius of 3 cm and a height of 7 cm, is a cylindrical cup (Ismail, 2011). To prevent short-lived particles from passing through the filter and entering the chamber, the cup is fitted with a barrier that delays the entry of gases.

A holder holds the CR-39 in place at the bottom of the detector chamber to reduce any potential movement-related errors. Radon enters the holder with a half-time entry of roughly 1 min, which is relatively short compared to the radon half-life of 3.82 days. Consequently, the radon concentration inside the detector chamber quickly equilibrates with the outside concentration, despite any outside concentration variations. Clean plastic bottles were used to collect tap water samples, which were tightly sealed to prevent radon leakage. The dosimeters, containing 282.7 ml (279.94 g) of water, were then placed in the bottles and left exposed for 60 days. Afterward, the dosimeters were retrieved and chemically etched at 6N NaOH at 70 °C for 9 h (Ismail & Jaafar,

Table 2
The concentration of radon in drinking water and its equilibrium factors with the water source.

location name	Source of drinking water	Average radon	
		Concentration (C_{wRn}^{222}) Bq/(L.d) using CR-39 NTDS	Concentration (C_{wRn}^{222}) Bq/(L.h) using RAD7
Su. Goiza	Artesian wells + River basins	1.087 ± 0.23	0.684 ± 0.10
Su. Mamostayan	Artesian wells + River basins	2.828 ± 0.29	0.686 ± 0.11
Su.Sabonkeran	Artesian wells	1.188 ± 0.34	0.51 ± 0.39
Su.Qiyasan	Artesian wells	2.561 ± 0.37	0.464 ± 0.22
Hajyawa	Spring waters	3.964 ± 1.08	0.81 ± 0.34
Chwarqurna	Artesian wells	3.207 ± 0.78	0.708 ± 0.70
Ranya	spring waters + River basins	3.182 ± 0.66	0.704 ± 0.33
Er. Mamostayan	Artesian wells	1.334 ± 0.81	0.604 ± 0.14
Er.Setaqan	surface wells	1.437 ± 0.43	0.624 ± 0.36
Er. Nowroz	Spring + surface wells	2.783 ± 0.41	0.678 ± 0.533
Er. Tyrawa	surface wells	3.086 ± 0.83	0.691 ± 0.19
Shawes	surface wells + Artesian wells	3.101 ± 0.77	0.672 ± 0.28
Koya	Spring waters	3.16 ± 0.68	0.694 ± 0.75
Daratwo	Spring waters	2.414 ± 0.66	0.69 ± 0.21
Similan	Spring waters	4.43 ± 0.93	0.964 ± 0.33
Suran	Spring waters + surface wells	4.143 ± 0.86	0.878 ± 0.26
Bnslawa	spring waters + surface wells	3.207 ± 1.02	0.717 ± 0.26
Akre	Spring waters	3.832 ± 0.83	0.722 ± 0.28
Kelak	surface wells	1.048 ± 0.11	0.416 ± 0.13

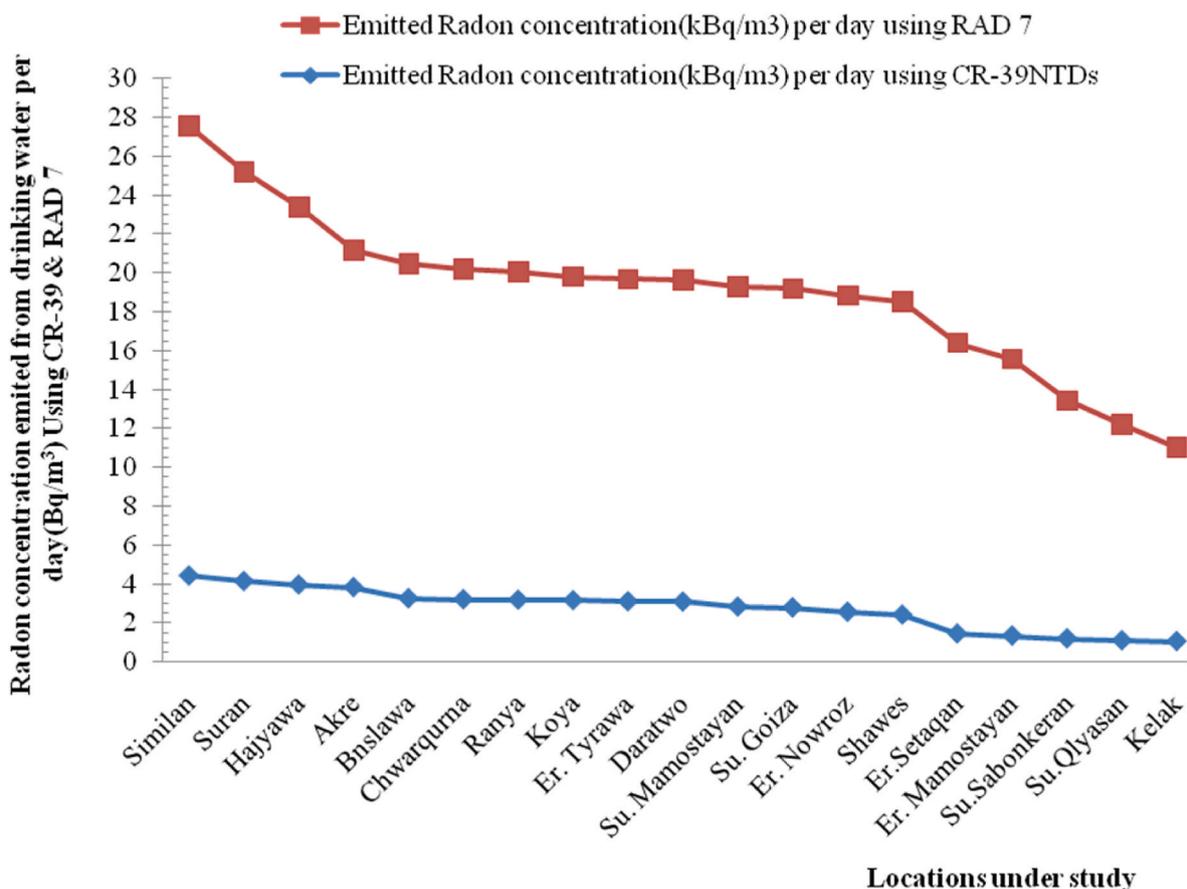


Fig. 3. Non-uniform distribution of radon concentration emanation from drinking water for selected locations, using passive (CR-39NTDs) and active (RAD 7) detecting methods.

2010c). To determine the number of tracks per cm^2 on each detector, 90 different folds were scanned using an optical microscope with a magnification of $400\times$.

2.2.2. Radon in drinking water using RAD7 technique

RAD7 was used as an active method to measure the airborne radon concentration that was emitted from the water samples. To dissolved radon gas from the depths continuously, moving and circulation of the

water were done with the measurements using the rubber air pump. The measuring process is based on detecting alpha particles produced from the disintegration of radon and its products using a polycarbonate solid alpha detector and converting alpha radiation directly to an electrical signal. Thus, during a short time (24 h), it will give an average concentration of airborne radon gas, so the measurements are short (Ismail, 2011). 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

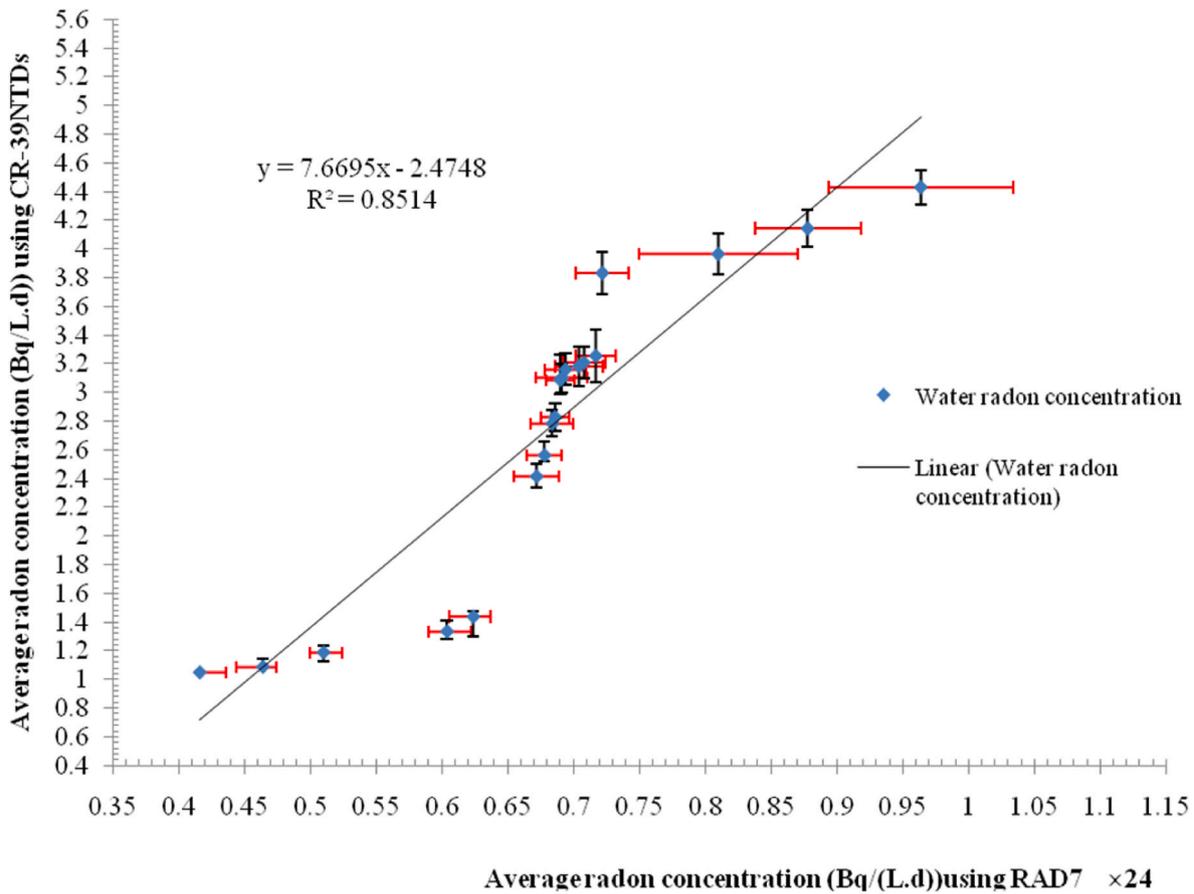


Fig. 4. Correlation between passive (CR-39NTDs) and active (RAD7) detecting technology for the values of average radon concentration.

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.

2.3. Radon measurements and inhalation dose estimation

2.3.1. Radon concentration in water, C_{wRn} (Bq/m³)

The radon concentration emitted from drinking water, denoted as C_{wRn} (Bq/m³), can be determined using the following equation (Ismail, 2004; Suresh et al., 2020).

$$C_{wRn}^{222} = \frac{D_0}{K} \tag{1}$$

where, K is the detector sensitivity ($K = 0.2315$ track/cm².d per Bq/m³) and D_0 is the track density (track/cm². day) of the closed-can technique.

2.3.2. Inhalation dose estimation (bronchial epithelium)

Inhalation exposure is defined in terms of the air concentration of radon progeny in working level (WL) units, and working level month (WLM) is an exposure equivalent to 1WL for 170 h. A working level is defined as a concentration of radon progeny through ²¹⁸Po totaling 1.3×10^5 MeV of potential alpha energy per liter of air, and it can be calculated as follows (Ismail, 2007).

$$WL = F \times \frac{C_{aRn}^{222}}{3700}; F = a \exp\left(\frac{bD_0}{D}\right) \tag{2}$$

Where, C_{aRn}^{222} is the contribution of radon concentration in drinking

water to indoor radon concentration (Bq/m³), F is the equilibrium factor, D and D_0 represent the track densities (track/cm².day) of the open (D: without a filter) and the closed (D_0 : with a filter) –can techniques respectively. The values of the two constants a & b are 14.958 and –7.436 (Ismail, 2007), respectively.

As conclusive evidence is lacking concerning the location of the relevant target cells, it appears there are different assumptions about a conversion factor for the bronchial epithelium; more details are mentioned by Cross et al. (Cross et al., 1984). In addition, a mean value of 0.007 Gy/WLM has been adopted in this work.

The transfer coefficient from water to air is 10^4 , so 37000 Bq/l in water converts to 3.7 Bq/l in air using 0.007 Gy/WLM as a reasonable lifetime conversion coefficient for environmental exposures. The process involves the utilization of an alpha quality factor of 20, an equilibrium factor (F), and continuous exposure to a radon concentration of 3.7 Bq/l, which is converted to an approximate bronchial epithelium dose (BED) equivalent of 4 mSv/y. In the current study, the equilibrium factor was a variable, and therefore, we adjusted its value as follows:

$$\text{Bronchial epithelium dose} = C_{aRn}^{222} \text{ (Bq/l)} \times F \times 2.16216 \text{ mSv/y} \tag{3}$$

2.3.3. Contribution of radon concentration in water to indoor radon, C_{aRn} (Bq/m³)

The correlation between radon levels in drinking water and indoor air poses a significant concern in environmental radioactivity. As Zalewski et al. (Zalewski et al., 2001) stated, radon present in water may be released into the indoor air during domestic water use, thereby potentially impacting indoor radon concentrations. The contribution of radon concentration in drinking water to indoor radon concentration (C_{aRn}^{222} , Bq/m³) can be calculated using the following equation:

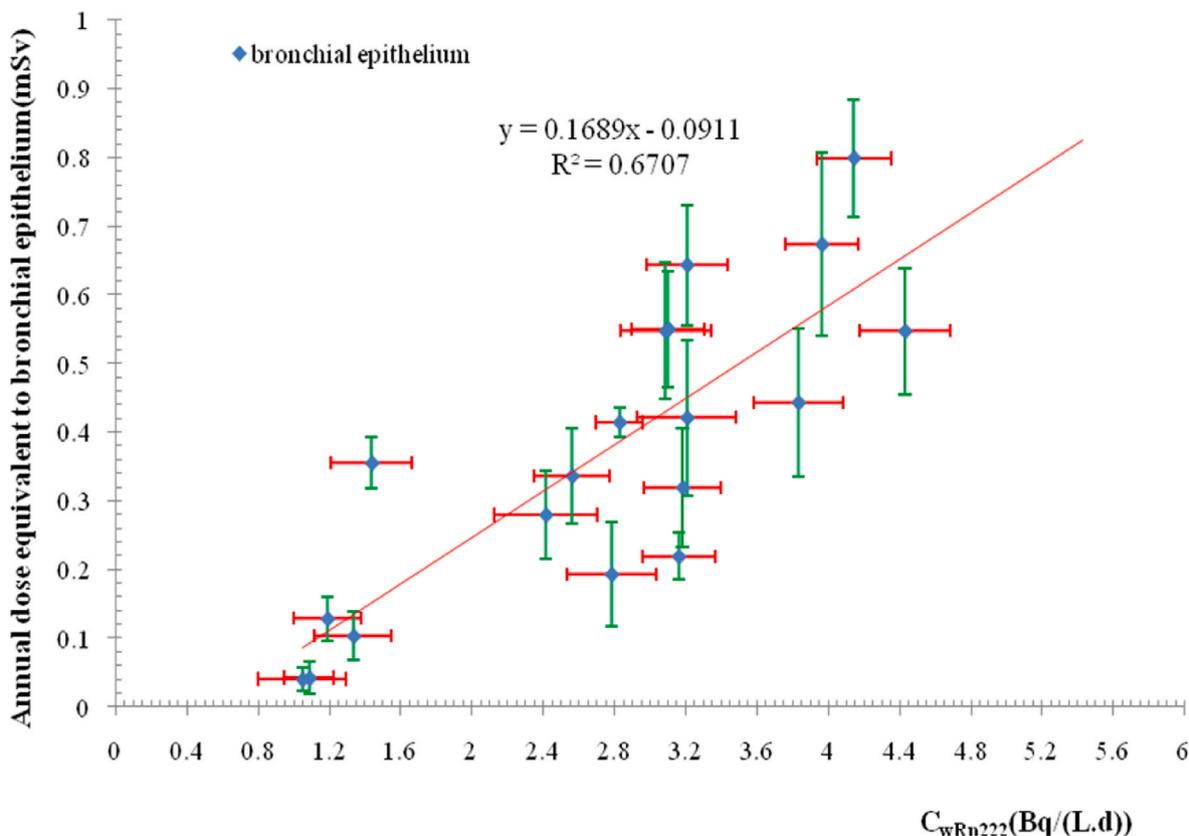


Fig. 5. Equivalent annual doses to the bronchial epithelium and the concentration of radon in drinking water.

Table 3

Radon concentration and its contribution to indoor radon concentration using CR-39NTDs in water, and its dose equivalent to the bronchial epithelium.

Location	CwRn NTD		CaRn NTD	D _o	D	F	bronchial mSv/d
	Bq/(L.d)	Bq/(m ³ .d)	Bq/(m ³ .d)				
Su. Goiza	1.087	1087	0.388	232.68	1143.84	0.05	0.042
Su. mamostayan	2.828	2828	1.010	275.12	106.79	0.19	0.415
Su. Sabonkeran	1.188	1188	0.424	930.74	1910.29	0.14	0.128
Su. Qlyसान	2.561	2561	0.915	282.9	1158.62	0.17	0.336
Hajyawa	3.964	3964	1.416	1149.28	1378.43	0.22	0.673
Chwarqurna	3.207	3207	1.145	191.66	1038.95	0.26	0.644
Ranya	3.182	3182	1.136	753.43	1266.38	0.13	0.319
Er. Mamostayan	1.334	1334	0.476	308.78	1382.68	0.1	0.103
Er. Setaqan	1.437	1437	0.513	232.68	318.26	0.32	0.355
Er. Nowroz	2.783	2783	0.994	644.31	152554	0.09	0.193
Er. Tyrawa	3.086	3086	1.102	1233.45	963.98	0.23	0.548
Shawes	3.101	3101	1.108	717.86	2598.45	0.23	0.551
Koya	3.160	3160	1.129	191.66	724.23	0.09	0.220
Daratwo	2.414	2414	0.862	459.01	1739.83	0.15	0.280
Similan	4.430	4430	1.582	594.1	1920.05	0.16	0.547
Suran	4.143	4143	1.480	727.76	301.99	0.25	0.800
Bnaslaw	3.207	3207	1.145	973.9	1501.61	0.17	0.421
Akre	3.832	3832	1.369	579.23	1589.95	0.15	0.444
Kelak	1.048	1048	0.374	150.15	29.83	0.05	0.040

$$C_{aRn^{222}} = C_{wRn^{222}} \times W \times \frac{e}{V \times \lambda c} \tag{4}$$

Where, $C_{wRn^{222}}$ is the radon concentration of drinking water (in kBq/m³), W is the consumption of water (in m³/h per person), V is the bulk of an indoor room, e is the coefficient of radon transfers from domestic water to indoor air and λc is the exchange rate of air (unit is h⁻¹). Equation (4) can be rewritten as

$$C_{aRn^{222}} = C_{wRn^{222}} \times f \tag{5}$$

$$\text{Where, } f = W \times \frac{e}{V \times \lambda c} \tag{6}$$

The notation f denotes the conversion coefficient of radon in water, which represents the contribution of water radon to indoor radon. According to references (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 1993; Xinwei, 2006), the coefficient of radon transfers from domestic water to indoor air, e is commonly 0.5, λc is 0.7/h, W is 0.01 m³/h per person, V is 20 m³ per person. The conversion coefficient of radon in water (f) was determined to be 3.57×10^{-4} . This indicates that the presence of 1 kBq/m³ of ²²²Rn

in drinking water can result in an increase of 0.357 Bq/m³ in the indoor radon concentration.

3. Results and discussion

As depicted in Fig. 1, the average concentration of indoor radon in certain locations within Iraqi Kurdistan followed a logarithmic scale during the summer, with 64.4% of readings below 150 Bq/m³, 19.4% between 150 and 200 Bq/m³, and 16.2% above 200 Bq/m³. Average indoor radon concentrations vary from location to location, as shown in Fig. 2. The mentioned variables depended on the type of dwelling and the ventilation rate.

As can be seen in Table 1, excellent ventilation rates as mentioned in previous work (Ismail & Jaafar, 2010) resulted in low radon concentrations whereas low ventilation rates resulted in high concentrations. The reason behind this is that the progeny of radon, particularly ²¹⁸Po and ²¹⁴Po, were treated as airborne particles, which could be suspended in the air along with dust and other loose materials inside the houses. Therefore, the ventilation rate of indoor air is greatly affected. The radon concentration was lower (139.11 ± 17.26 Bq/m³) in areas with good ventilation (13.7%), whereas it was higher (189.78 ± 55.91 Bq/m³) in areas with poor ventilation (56.45%) (Ismail & Jaafar, 2010).

Dissolved radon in drinking water for the houses that have had their indoor radon evaluated using short- and long-term measurements. The values of dissolved radon concentration, its contribution to indoor radon concentration, and the equilibrium factor between radon and its progeny are listed in Table 2. The mentioned values varied from one location to another, depending on the source of drinking water. In addition, the average radon concentration in drinking water was variable from 1.048 ± 0.11 Bq/l to 4.43 ± 0.93 Bq/l per day and from 0.416 ± 0.13 Bq/l to 0.964 ± 0.33 Bq/l per hour for passive and active detecting techniques, respectively. The results were below the recommended level of WHO (20 Bq/l), and corresponded with the radon gas in drinking water in other countries (El-Araby et al., 2019; Nuhu et al., 2020; Suresh et al., 2020).

For both techniques, the high and low values of radon concentration were in the cities of Similan and Kelak, respectively, as shown in Fig. 3. On the other hand, Fig. 4 shows a good correlation between the values of average radon concentration between both active and passive detecting methods, which proves the high efficiency of CR-39 NTDs. Further, the values are smaller than the internationally recommended limit of 11 Bq/l as proposed by the Environmental Potential Agency, USA (EPA (Environmental Protection Agency), 2009). The impact of radon in drinking water on indoor radon concentration is notably greater in homes supplied with drinking water from artisan wells.

Fig. 5 illustrates a strong correlation ($R^2 = 0.67$) between the annual equivalent doses to the bronchial epithelium and the concentration of radon present in drinking water. From this experimental relation, the annual dose equivalent to the bronchial epithelium (mSv) = 0.168 × C_{wRn222} (Bq/l) - 0.091. There is a significant variation in the radon concentration levels of different types of natural water. Typically, spring and surface water contain lower levels of radon, while drilled well water has higher levels. Thus, the impact of water radon levels on indoor radon concentrations is influenced by the source of the drinking water.

The results of the annual dose equivalent to the bronchial epithelium are presented in Table 3. The average annual dose equivalent to the bronchial epithelium ranges from 0.025 to 0.843 mSv/y, with the highest value observed in Hajyawa and the lowest in Kelak. These findings suggest that dissolved radon has an impact on the bronchial epithelium.

4. Conclusions

The environmental radiological hazards associated with elevated levels of radon gas in drinking water were estimated using passive and active detectors for different sources of tap water. Airborne radon present in drinking water was found to contribute to increased indoor radon

levels. Almost anywhere with a high indoor radon level also has a high radon exhalation rate from drinking water. Thus, a good correlation ($R^2 = 0.67$) was found between the annual equivalent doses to the bronchial epithelium and the concentration of radon present in drinking water. Furthermore, there was a significant variation in the radon concentration levels of different types of natural water. Typically, spring and surface water contain lower levels of radon, while drilled well water has higher levels. Notably, all values were lower than the internationally recommended limit of 11 Bq/l set by the US Environmental Protection Agency.

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