



Advance application of liquid scintillation counter to estimate radon concentration in groundwater

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Abstract

Radon (^{222}Rn) is radium's immediate radioactive decay product (^{226}Ra) in the uranium decay series. Radon is classified as a group 1 carcinogen due to its hazard to human health, and the evaluation of radon concentrations has become essential. Therefore, an advanced method was applied to a liquid scintillation counter (Hidex 300SL) to measure the radon concentration in groundwater. The estimated radon concentration was between 0.1 and 3.20 Bq/L, with an average of 0.96 Bq/L and with a standard deviation of 0.82 Bq/L. The estimated radon concentrations are in the safe range recommended by the United States Environmental Protection Agency (USEPA) and European Atomic Energy Community (EAEC). However, the values are far below the safe range set by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), European Commission (EC), and World Health Organization (WHO) standards. Nevertheless, the estimated radon concentration by this advanced application ensured no risk of radon exposure from the groundwater of the tested area.

Keywords Liquid scintillation counter · Groundwater · International safety standard · Hidex 300SL, Radon concentration

Introduction

Radon gas and its progeny are the most significant natural radionuclides present in the air, water, ground, underground, groundwater, and surface water (Aleissa et al. 2012; Aleissa et al. 2013; Gebresilasie et al. 2021; Küçükönder and Gümür 2022). The human body can be exposed to the radon gas directly by inhalation and by ingestion of water (Aleissa et al. 2013; Althoyaib and El-Taher 2015). Radon can expose most of the natural radiation to humans and poses the most significant risk for humans by causing several types of cancer, such as liver, colon, and lung cancer (Harrison 2021; Kendall and Smith 2002; UNSCEAR 1988; USEPA 1991). The average annual radon dose is approximately half of the

average natural dose and twice that of all other artificial radiation sources (Mehra and Bala 2014; USEPA 1991). Hence, the measurement of radon activity has become a major essential issue for researchers worldwide (El-Araby 2018; Kuo 2023; Maringer et al. 2013; Massoud et al. 2020). International organizations set different radon concentration levels as safety standards to be aware of radon exposure. For example, the United States Environmental Protection Agency (USEPA) and European Atomic Energy Community (EAEC) have set a value of 11.1 Bq/L as a safe level for drinking water (EC 2019). The World Health Organization (WHO) recommends 100 Bq/L (WHO 2007, 2011) as a safe level of radon in water. For any water source, if the radon concentration exceeds the safety level, the WHO recommends treating the water until the concentration is reduced to below the safety level.

Water is essential for living beings, so the measurement of radon concentration, its annual consumption, and the corresponding health risk assessment are crucial. Many countries worldwide have aimed to estimate the radon concentration in water using various techniques (Hopke et al. 2000; Malik et al. 2015), such as ionization chambers, liquid scintillation counting, Lucas cells, gamma spectroscopy, solid-state alpha detection, and aeration from water (De Simone et al. 2015). The liquid scintillation counter is a modern and

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convenient device for measuring radon concentrations. A Hidex 300 SL scintillation counter is suitable for its high efficiency in measuring radon concentrations. The liquid scintillation counter has a low background count rate and minimum detectable activity. The device is equipped with an autosampler to handle many samples simultaneously. The analytical conditions, background count rate, and counting efficiency are controllable for the liquid scintillation counter. The advantage of a liquid scintillation counter is that it can count even low radon activity. Various types of samples, for example, solids, liquids, suspensions, and gels, can be accommodated with an LSC counter. Moreover, different isotopes can be counted simultaneously. The data analysis, efficiency correction, graph plotting, various calculations, and radioimmunoassay are programmable and can be controlled using a built-in computer.

For the importance of the above, the present work represents the estimation of the radon concentration of groundwater located in the eastern area of Saudi Arabia, and those wells are being used for drinking and domestic purposes. The estimated radon concentrations are compared with the international safety standards set by various international organizations. The results estimated using a liquid scintillation counter are verified with other standard techniques. To the best of our knowledge, this report is the first attempt to estimate the radon concentration in groundwater using a liquid scintillation counter in this area.

Materials and method

Study areas

The present area of investigation is located in the northeastern part of Saudi Arabia. Three cities are covered in this study: Hafr Al Batin, Thybiyah, and Qaisumah, as shown in Fig. 1. Hafr Al Batin (Al Batin valley) is named for its location in the dry valley of Wadi Al Batin. Wadi Al Batin is a part of Wadi al Rummah, the largest and longest dry river in the Arabian Peninsula, approximately 2000 km long (Aly et al. 2015; El-Taher and Al-Turki 2016; El-Taher et al. 2020). The central part of Wadi Al Rummah comes through the Al Qassim oasis, one of the leading agricultural regions (Aly et al. 2015). During summer days, the climate is arid and hot (40–50 °C), and it falls during winter nights to (–2–8 °C). It rarely rains during winter, and no rainfall is recorded during summer.

Geology of the study areas

Hafr Al Batin, located in northeastern Saudi Arabia, exhibits a geological landscape dominated by sedimentary rock formations. These sedimentary rocks, including sandstone,

limestone, shale, and siltstone, bear testament to the area's ancient geological history, with the potential for valuable insights through fossil discoveries. The region is renowned for the Saq Aquifer, an essential groundwater source stored within porous sedimentary layers. Geological structures, such as fault lines and fractures, can influence groundwater dynamics and the distribution of potential mineral resources such as limestone. While situated within the generally stable Arabian Plate, occasional seismic activity remains possible due to its tectonic setting. In light of its geological properties, Hafr Al Batin presents opportunities for further exploration, resource assessment, and geological hazard evaluation, ensuring responsible land use and environmental preservation in this unique geological landscape. The areas have good water quality due to their aquifers and geological structure (Vincent 2008). Based on the report of Vincent and Alsharhan et al. the main aquifers are based on primary and secondary origins. The primary origins are Quaternary sands, including quartz sandstones and conglomerates with primary porosity. The secondary porosity with oolitic limestone (Alsharhan et al. 2001; Vincent 2008). Hafr Al Batin is located on a Quaternary sand aquifer, where deep wells supply relatively good water compared to shallow wells. Radon can be generated between the rocks as an intermediate radioactive decay of uranium/thorium radioactive substances. Therefore, radon can be present in aquifer rocks dissolved in groundwater. It was found that groundwater associated with granite and metamorphic rocks has relatively higher radon concentrations (Durrani 1999). Since radon gas is dissolved in groundwater, it might escape from water during contact with the atmosphere. Serious attention was taken during the sampling process to obtain the actual radon concentration. Groundwater samples from 18 source wells were collected into plastic bottles and capped immediately. For better performance, in most cases, the measurements were carried out within 4 h of sample collection.

Radon measurement by LSC

The Hidex 300SL is one of the most useful liquid scintillation devices based on the triple-to-double coincidence ratio (TDCR). A photograph of such a measurement device in our laboratory is shown in Fig. 2a. The device has three photomultiplier tubes on the same plane at 120 degrees (Fig. 2b, c). The device is based on the triple-to-double coincidence ratio (TDCR) (Arun et al. 2021; Broda 2003). A scintillation principle is shown in Figure S1 in the supporting information. The detector has three photomultipliers that enable counting of the triple-to-double coincidence ratio, a method to obtain the counting efficiency of an unknown sample without any standard source. TDCR is a method that can be applied for both chemical and color quenching, aqua- and organic samples, cocktails, and isotopes. First,

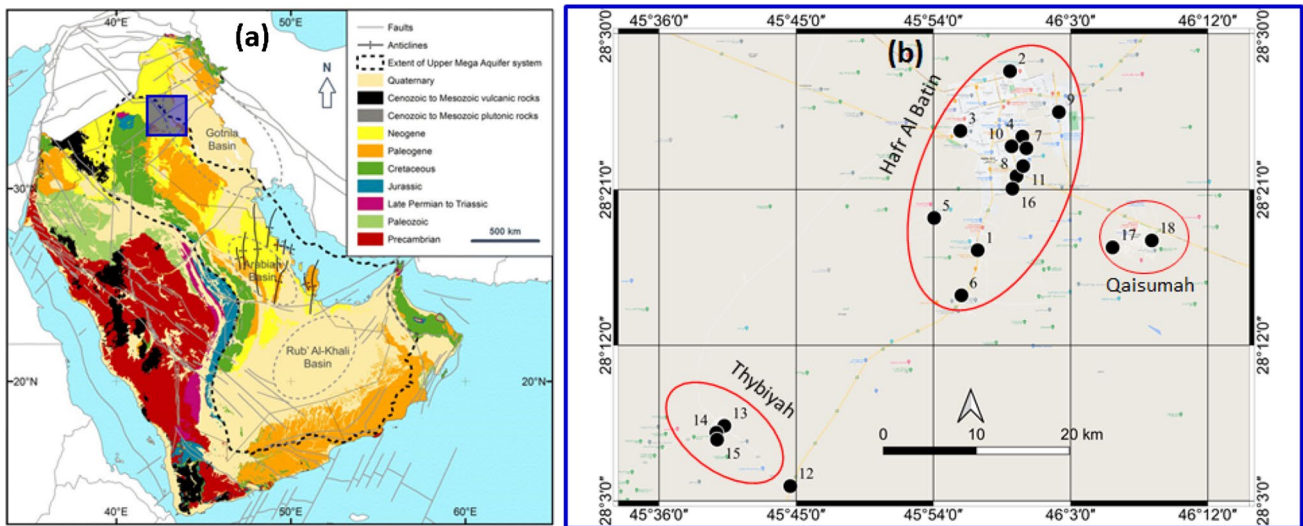


Fig. 1 a Geological map of Saudi Arabia, and b study areas of Hafr Al Batin, Thybiyah and Qaisumah, Eastern Province, Saudi Arabia

an appropriate scintillation cocktail (Aqualight Plus, Hidex, UK) was added to the scintillation vial. Then, the water sample was inserted below the scintillation cocktail by ensuring no aeration of the water and secured with a tight cap. The vial was kept for an hour so that radon’s short-term decay product entered equilibrium. Then, the scintillation counter technique was used to measure the activity of radon from the photon emission rate from the fluids. Before counting with the scintillation counter, the procedure below was followed.

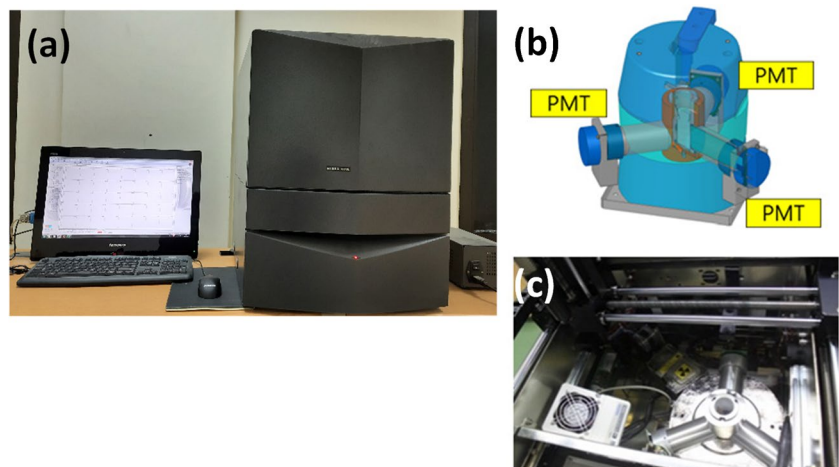
The samples were always kept in cool and dark places since luminescence can have a significant effect. In addition, Hidex 300SL discharges static electricity from the sample to avoid the static electricity effect. Samples were prepared by adding 12 ml of a water-soluble cocktail to 8 ml of an aqueous sample. The scintillation device then measured the radon activity by emitting photons. It should be mentioned that special attention should be taken during the sample

preparation; otherwise, the estimated result will be influenced by escaping the radon gas from the water sample.

Results and discussion

Before analysis of an unknown sample using an LSC, the device should be monitored by testing background radiation, calibration, and efficiency checked biweekly or monthly using a standard source. Such a controlled chart for calibration using a standard source of certified activity-free solution, carbon 14 (¹⁴C) source, and tritium source (³H) is shown in Fig. 3. The activity-free solution shows an almost constant reading with a minimal experimental error in the last eight years (Fig. 3a). This implies that radiation is always present in the activity-free solution and environment, even without a radioactive source. For the standard source

Fig. 2 a Front view of the Hidex 300SL, b schematic diagram, and c photo of the detector



of carbon 14, no significant changes in count per minute (CPM) were detected due to its longer half-life (approximately 5715±40 years) (Choppin et al. 2002).

On the other hand, for ³H, a significant decrease in CPM was found eight years after initializing the device (0.6162 times the initial count) (Fig. 3b). A rough estimation shows a half-life of approximately 11.75 years for ³H, which is close to the published data of 12.3 years reported in the literature (Lucas and Unterweger 2000). It should be mentioned that if the measured value is approximately 1000 times higher than the background count, the background effect can be ignored.

The measured concentrations of radon for 18 groundwater samples collected from the Hafr Al Batin, Thybiyah,

and Qaisumah areas, Saudi Arabia, are tabulated in Table 1. It was found that the concentration varied in the range of 0.1 to 3.20 Bq/L, with an average of 0.96 Bq/L and a standard deviation of 0.82 Bq/L. Many national and international organizations have established their radon levels worldwide. For instance, the United States Environmental Protection Agency (USEPA) set a value of 11.1 Bq/L for the radon concentration in groundwater (USEPA 2000). The UNSCEAR set 40 Bq/L as a safe limit in their published report, (UNSCEAR 2000) while the European Commission (EC) and World Health Organization (WHO) have described a value of 100 Bq/L as an action limit (EC 2019; WHO 2011). As observed,

Fig. 3 Control chart for the calibration source; for the certified solution of **a** activity-free water for background, and **b** certified source of ³H (solid circle) and certified source of ¹⁴C (open circle)

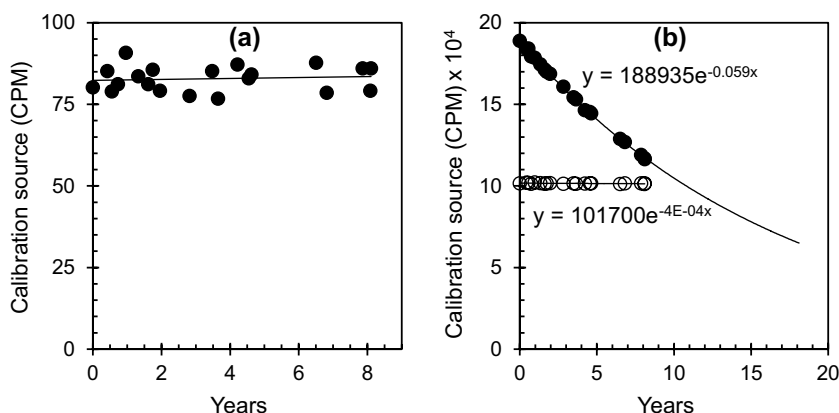


Table 1 Estimated radon concentration using LSC

Serial no.	Sample ID	Area	Depth of Wells	Purpose of use	Hidex 300SL (Bq/L)
1	02d01	Hafr	Shallow	Domestic use only	0.70
2	03DSt01	Hafr	Deep	Drinking water treatment plant	1.86
3	05DSt03	Hafr	Deep	Drinking water treatment plant	1.19
4	07DSt05	Hafr	Deep	Drinking water treatment plant	0.60
5	09d03	Hafr	Shallow	Domestic and irrigation	1.65
6	10d04	Hafr	Shallow	Domestic use only	3.50
7	11St06	Hafr	Deep	Drinking water treatment plant	1.71
8	12St07	Hafr	Deep	Drinking water treatment plant	0.14
9	15d07	Hafr	Shallow	Domestic use only	0.50
10	19St08	Hafr	Deep	Drinking water treatment plant	3.10
11	20d11	Hafr	Shallow	Domestic use only	1.87
12	24d15	Thybiyah	Shallow	Domestic use only	2.50
13	25d16	Thybiyah	Shallow	Domestic use only	2.48
14	28St11	Thybiyah	Deep	Drinking water treatment plant	2.20
15	32St12	Thybiyah	Deep	Drinking water treatment plant	1.45
16	38d24	Hafr	Shallow	Domestic use only	1.10
17	40d26	Qaisumah	Shallow	Domestic use only	0.20
18	44d30	Qaisumah	Shallow	Domestic use only	0.10

Depth of the shallow well: ~400 m
 Depth of the deep well: 800 m and above

all samples measured radon levels below those limits. Approximately 90% of the samples were far less than the radon levels reported earlier (Al-Jaseem et al. 2016; Aljaloud and ElBatouti 2021). Furthermore, the estimated radon concentrations in the present study are comparable to those found in the rest of Saudi Arabia (Abuelhia 2017; Abuelhia 2019; Aleissa et al. 2012; Aleissa et al. 2013). For example, a range of 1.45–9.15 Bq/L was found in the Al-Jawa area, as reported by Althoyaib et al. (2015); 0.76–4.69 Bq/L was found for groundwater from the Al-Qassim area, reported by El-Taher et al. (2016); (0.92–2.12) Bq/L was found in the Jeddah area reported by Tayyeb et al. (1998); 0.11–9.20 Bq/L was found in the Dammam area, reported by Abuelhia (2017); and 1.74–4.32 Bq/L was found in the Jazan area, as reported by El-Araby et al. (2019).

A comprehensive analysis of the data presented in Table 1 unveils a discernible trend: when examining radon concentrations, the average values in shallow wells consistently demonstrate a lower tendency compared to their counterparts in deep wells. This general pattern holds true with remarkable consistency, and it serves as valuable insight into the distribution of radon within the groundwater sources. However, it is important to note the presence of certain exceptions that defy this overarching trend. These exceptions are notable, as they deviate from the established pattern, adding complexity to the assessment. Notably, some shallow wells situated within the lower valley of the arid Al Batin River exhibit elevated radon concentrations that are atypical for their category. This localized variation suggests that geological and hydrogeological factors unique to this particular region may contribute to the observed discrepancies. Additionally, exceptions are not confined solely to shallow wells; they extend to certain deep wells as well. Furthermore, deep wells positioned atop the hillside display deviations from the anticipated pattern. These exceptions are indicative of the complex interplay of geological, hydrological, and environmental variables that influence radon concentration in groundwater (Massoud et al. 2020).

The standard deviation (σ) was calculated mathematically for each groundwater sample. Statistically, the standard deviation of a series of samples indicates the deviation from the mean value and is a measure of the precision of the measurements. Radioactive decay follows the Poisson distribution law, from which it is possible to calculate the standard deviation as the square root of recorded counts by the RAD7 detector. When no decay is recorded, instead of reporting zero uncertainty, RAD7 reports an uncertainty value based on 2σ . In this study, the cycle time was increased to 30 min for maximum certainty. The radon concentration in the water sample obtained in Bq/m^3 was converted to Bq/L.

If the sample was collected earlier but analyzed later, there must be a decay due to the delay in measuring radon

concentration. Therefore, a correction should be made for the sample decay depending on the sampling time. The decay correction factor (*DCF*) is simply an exponential function with a time constant of 132.4 h and is calculated based on the following formula:

$$DCF = e^{(T/132.4)} \quad (1)$$

where T is the duration (in hours) between sampling and counting by the LSC instrument, and 132.4 h is the mean lifetime of the ^{222}Rn atom (radon has a half-life of 3.825 days, multiplied by 24 h per day and divided by the natural logarithm of 2) (Salvato 2003). Such a decay correction factor with time is plotted in Fig. 4 (solid line). From the DCF plot, it can be seen that samples were appropriately sealed, stored, and counted by LSC within 24 h, and no significant correction was needed. However, a correction is needed if the sample is not counted immediately, and the DCF increases exponentially with time. In this study, in most cases, the sample was counted immediately within 4 h of sample collection except for six (open circle in Fig. 4) collected from Thybiyah and Qaisumah cities (those cities are far from the LSC measurement lab). A proper correction using the DCF equation was made and is tabulated in Table 1.

The estimated results using the LSC method are comparable to other countries surrounding the Kingdom. Table 2 compares the estimated radon concentrations of water in several countries. The radon concentration obtained in the current study follows the calculated values obtained from surrounding countries, such as Yemen, UAE, Kuwait, Iraq, and Jordan, as tabulated in Table 2. On the other hand, the

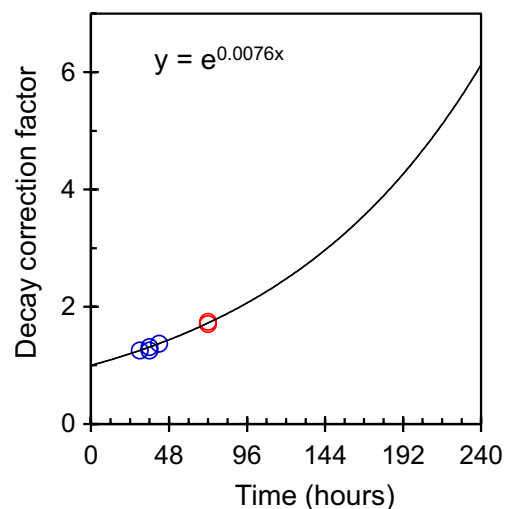


Fig. 4 The sampling time-dependent decay correction factor (DCF) is shown as a solid line, and few samples corrected with DCF collected from the Thybiyah and Qaisumah areas are far from our LSC-measured lab

Table 2 Reported radon concentrations at different locations around Saudi Arabia.

Type of water	Location	Radon concentration, C_{Rn} (Bq/L)
Drinking bottle water	Kuwait City, Kuwait	1.02–6.05 (Seoud 2018)
Drinking bottle water	Algeria	2.6–14.0 (Amrani 2002)
Drinking water	Kufa, Iraq	0.0039–0.221 (Abojassim 2014)
Drinking water	Bam village, Iran	1.20–9.88 (Malakootian and Nejhad 2017)
Drinking water	Baoji, China	30.0–127.0 (Xinwei 2006)
Drinking water	Dammam, Saudi Arabia	0.11–9.20 (Abuelhia 2019)
Well water	Afyonkarahisar, Turkey	0.70–31.70 (Yalim et al. 2007)
Well water	Jordan	3.10–5.10 (Al-Bataina et al. 1997)
Well water	Baita Stei, Romania	4.78–35.50 (Moldovan et al. 2014)
Groundwater	Khartoum, Sudan	1.58–345.10 (Idriss et al. 2011)
Groundwater	Al-Jawa, Saudi Arabia	1.45–9.15 (Althoyaib and El-Taher 2015)
Groundwater	Al-Qassim, Saudi Arabia	0.76–4.69 (El-Taher 2012)
Groundwater	Jeddah, Saudi Arabia	0.92–2.12 (Tayyeb et al. 1998)
Groundwater	Jazan, Saudi Arabia	1.74–4.32 (El-Araby et al. 2019)
Groundwater	Hafr Al Batin, Saudi Arabia	0.1–3.5 (present study)

results in the present study are far below those in Turkey, Algeria, India, Romania, Sudan, Iran, and China. The geological nature of the sampling area, soil nature, and environment might be the reason for the difference in radon concentrations (Asuni et al. 2021). Moreover, the irradiative emissions from radon, such as neutrons, neutron flux, water samples, contamination of other radioactive sources, and pH, can also influence the experimental results. (Aljaloud and ElBatouti 2021; Martin 2006) We are now estimating the radon concentration using other standard methods to better compare the advantages and disadvantages of the techniques, which will be reported elsewhere.

Conclusion

An advanced application using a liquid scintillation counter was used to ascertain the radon concentration in groundwater samples collected from diverse locations in the northeastern region of Saudi Arabia. The results of this meticulous analysis revealed that the estimated radon concentrations of these groundwater samples ranged from as low as 0.1 Bq/L to a maximum of 3.20 Bq/L, with a calculated average concentration of approximately 0.96 Bq/L and a standard deviation of 0.82 Bq/L. In the most northern part of the area, particularly within the lower valley, shallow wells were identified as having significantly higher radon concentrations. In contrast, wells situated in the hillside areas showed notably lower levels of radon. This variation in radon concentration is likely attributable to the geological composition of the rocks and sediments that the groundwater passes through. Remarkably, these findings indicate that the radon concentrations detected in

the groundwater of this region consistently fall well below the safety thresholds established by both the United States Environmental Protection Agency (USPEA) and the European Atomic Energy Community (EAEC). Furthermore, they notably and comfortably remain significantly below the safe level recommendations established by two globally recognized authoritative bodies: the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) and the World Health Organization (WHO). The results concluded that the potential radon exposure to humans residing within the tested areas of the Eastern Province of Saudi Arabia is unequivocally and substantially lower than the stringent safety limits stipulated by international standards. As a result, this comprehensive assessment offers reassuring evidence that there exists no discernible risk of radon exposure stemming from the groundwater within the sampled cities of this region. Such meticulous scientific investigations are paramount in ensuring the continued safety and well-being of the local population and serve as a testament to the rigorous scientific methods employed in environmental monitoring and risk assessment.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s12517-023-11846-8>.

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Data availability The dataset used in this study is available from the corresponding author upon reasonable request except for data that are subject to third-party restrictions.

Declarations

Conflict of interest The authors declare no competing interests.

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