THE ACTIVITY CONCENTRATIONS OF ^{222}Rn IN SOME GROUNDWATER WELLS, NAJRAN CITY, SAUDI ARABIA

by

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Scientific paper
http://doi.org/10.2298/NTRP1702166A

In this work, the radon exhalation rate, effective radium content and radiation doses from some groundwater wells in Najran City, Saudi Arabia, were addressed and discussed in detail. This survey of radon concentrations in the groundwater was carried out using the passive measurement technique, where the radon gas passively diffuses into the detector. The obtained results revealed that the radon exhalation rate in terms of area and mass exhibits linear correlations with effective radium in groundwater (correlation coefficient $R^2 = 1$). Also, the majority of radon concentrations are within the UNSCEAR 1993 permitted level and the average annual effective doses obtained for radium and radon are 180 μSv and 860 μSv, respectively.

Key words: radon concentration, CR-39 detector, annual effective dose, groundwater

INTRODUCTION

Groundwater contains different amounts of dissolved radioactive elements from the radionuclides found in the earth. The amount of radionuclides in ground water can vary by several orders of magnitude and is influenced by several physical, chemical and geological properties of the aquifer [1]. The occurrence and distribution of radioactivity in water depend on the local geological characteristics of the source, soil, rock and other factors that control the occurrence and distribution of radionuclides in ground water and the hydrogeological condition and the geochemistry of radionuclides [2].

Radium-226 found in rocks and soil is the main source of radon in groundwater [3, 4]. Radon emanation depends mainly on the content of ^{226}Ra, mineral grain size and transport in the land that is governed by geophysical and geochemical parameters [5]. After radon gas is inhaled, densely ionising alpha particles emitted by deposited short-lived decay products of radon can interact with biological tissues leading to DNA damage [6, 7]. The Kingdom of Saudi Arabia is located in an arid region, which still depends and will continue to depend on groundwater as a source of drinking water and various human activities. The geology of the Najran region is surrounded by many outcrops of different geological formations which are mainly characterized by acidic igneous rocks such as feldspars, biotite, micas and quartz, [8].

There are many ground water wells scattered throughout the region, and these wells are the main source of drinking water in the Najran region, after purification [9]. Measurement of the activity of radium-226 and its daughter ^{222}Rn in water is very important for environmental pollution and public health studies. The data-base concerning radon concentration and radium content in Najran is limited. The main objectives of this study were assessment of the radon exhalation rate, the effective radium content and annual effective doses from some groundwater wells in Najran City, Saudi Arabia. The radon concentration in groundwater has been measured using the can technique during the summer of 2015.

Geology of the study area

Najran is located in the south western part of the Kingdom between 200-170 degrees north latitude and between 440-520 degrees east longitude. Its area is about 365000 km^2, as shown in fig. 1 [10]. There are two types of rocks characterized by their region as acidic igneous rocks such as feldspars, biotite, micas and quartz, and metamorphosed igneous rocks [8].

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Figure 1. Map of Saudi Arabia with Najran city

MATERIALS AND METHODS

In this environmental study, a CR-39 track detector was used to evaluate the radon concentration and effective radium content in some ground-water wells. Thirty-nine water samples were collected from thirteen ground-water wells in the Najran region. Nine ground-water wells are illustrated in fig. 2. The groundwater samples were collected without any kind of treatment. The samples were collected and analysed by using the closed can technique, as shown in fig. 3. Two litres of groundwater from each well were poured into clean plastic bottles. The bottles were closed tightly at the site, in order to prevent radon leakage. Water samples were collected after 20 minutes from running wells, and 500 ml of water from each bottle were placed into a plastic jar with dimensions of (48cm in width, and 24 cm in height). Thirty-nine pieces of CR-39 of thickness (250 m) and an area of about (0.5 cm 0.5 cm) were fixed at the bottom of a cup, which was covered by a piece of sponge with thickness 0.5 cm to make sure that thoron (^{220}Rn, $t_{1/2}$ = 55.60 s) cannot reach the detector. Plastic cups of 8 cm in length and 7 cm in diameter were used in this work. From the interior section, the cups were fixed on the covers of the plastic jar, as shown in fig. 3. These cans were stored at room temperature for a period of 90 days (exposure date: 7/5/2015 to 7/8/2015). After the exposure time, the cups were separated from the plastic jars. The detectors were removed and chemically etched using a 6.25 N solution of NaOH at 70 ºC for 6 h, then CR-39 detectors were washed with tap water for 10 min and then were put in distilled water for 10 min and then the detectors were left to dry. CR-39 detectors were counted visually using an optical microscope with a power of (400X).The efficiency of the detection has been determined in our previous work [11]. It was noticed that the registration efficiency of the CR-39 detector was approximately 100 %.

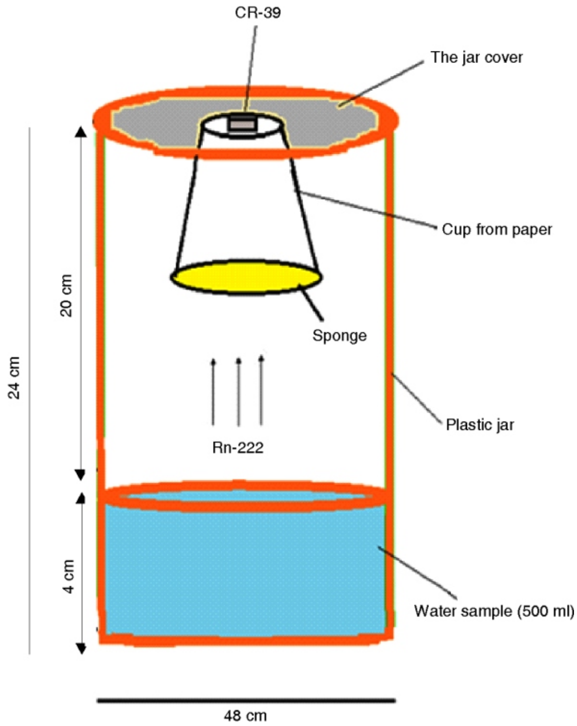

Figure 3. Configuration of the sealed-can technique

The radon activity concentration [Bqm^{-3}] has been calculated by using the following eq. [12].

$$C \quad \frac{\rho}{KT} \tag{1}$$

Figure 2. Nine groundwater wells (white circles) in the Najran region

where K is the calibration constant (Bqm^{-3}d^{-1}) /(tracks per cm^2), ρ – the track density (number of tracks per cm^2), and T – the exposure time (in days). The calibration factor value (0.20 0.01) obtained from our previous work [12] was used to assess the radon activity concentration from the track density.

The effective radium content $C_{\rm Ra}$ (BqL^{-1}) has been determined from the relation (equation?) [13, 14]

$$C_{\rm Ra} \quad \frac{rhA}{kT_{\rm e}M} \tag{2}$$

where ρ is the counted track density, h [m] – the distance between the detector and the top of the sample, K – the calibration factor of the CR-39 detector, M [kg] – the mass of the sample and ($T_{\rm e}$) is the effective exposure time which can be determined using the following equation

$$T_{\rm e} \quad T \quad \frac{(1 \quad {\rm e}^{\lambda_{\rm Rn}T})}{\lambda_{\rm Rn}}$$

where T is the exposure time, and $\lambda_{\rm Rn}$ [h^{-1}] the decay constant for radon . The radon exhalation rate in terms of area ($E_{\rm A}$) and mass ($E_{\rm M}$) was obtained from the relations, reported by [13, 14]

$$E_{\rm A} \quad \frac{C_{\rm Rn}\lambda V}{A \;\; T \quad \frac{1}{\lambda_{\rm Rn}} \;({\rm e}^{\lambda_{\rm Rn}T} \quad 1)} \tag{3}$$

$$E_{\rm M} \quad \frac{C_{\rm Rn}\lambda V}{M \;\; T \quad \frac{1}{\lambda_{\rm Rn}} \;({\rm e}^{\lambda_{\rm Rn}T} \quad 1)} \tag{4}$$

where $C_{\rm Rn}$ [Bqm^{-3}h] is the radon exposure , $\lambda_{\rm Rn}$ [h^{-1}] – the decay constant for radon, A [m^2] – the surface area of water samples, V [m^3] – the volume of the can, T [h] – the time of exposure, M [kg] – the mass of the sample. In order to appreciate the risk associated with the intake of water in the study area, the annual effective dose has been calculated according to the following relation [15, 16]

$$D_{\rm eff} \quad CR\, D_{\rm cf} D_{\rm wc} \tag{5}$$

where $D_{\rm eff}$ [mSv] is the annual effective dose, CR [BqL^{-1}] – the concentration of ^{222}Rn or ^{226}Ra in, $D_{\rm cf}$ – the ingestion dose conversion factor of ^{222}Rn or ^{226}Ra (10^{-8} SvBq^{-1} for ^{222}Rn, and 2 10^{-7} SvBq^{-1} for ^{226}Ra) [17], and $D_{\rm wc}$ – the water consumption (730 L per year) [18].

RESULTS AND DISCUSSION

Table 1 depicted the ^{222}Rn concentration, effective radium, areal exhalation rate, mass exhalation rate, and annual effective dose for the groundwater samples. This survey leads to the following:

– The activity concentrations of ^{222}Rn range from 12.20 0.83 to 32.50 0.99 Bqm^{-3} with a mean of 24.61 1.63 Bqm^{-3}, and the range activity concentration of Rn-222 is within the permission level (4-40 Bqm^{-3}) [17] for all groundwater samples as shown in tab. 2.
– The concentration of radon in groundwater wells in this study has large variation which may be attributed to high concentrations of ^{226}Ra deposited in aquifer rocks, also, the rocks in the Najran area belong to igneous rocks, as well as some stratified rocks of the sandstone [19], and also maybe due to variable concentrations of sources of the aquifer materials, porosity of the aquifer, permeability, and emanation rates from mineral sources [20].
– The activity concentrations of ^{226}Ra range from 2.07 0.83 to 5.53 0.95 BqL^{-1} with a mean of 4.18 0.28 BqL^{-1} as shown in the tab. 2. About 95 % from the groundwater samples showed radium activity concentrations above the WHO recommended value of 1 BqL^{-1} [21], and 5% from the groundwater samples are lower than this recommended value. The difference in the activity concentrations for both ^{226}Ra, and ^{222}Rn may be due to radon loss during storage in a polyethylene bottle [22-24].
– The areal radon exhalation rate for groundwater samples varied from 10.11 0.83 to 26.93 0.95 mBqm^{-2}h^{-1}, with a mean of 20.39 1.35 mBqm^{-2}h^{-1}. Also, the mass exhalation rates varied from 8.54 0.83 to 22.74 0.95 mBqkg^{-1}h^{-1} with a mean of 17.22 1.14 mBqkg^{-1}h^{-1} as shown in the tab. 2. Groundwater wells have a high mean areal , and the mass radon exhalation rate compared to the global average of the areal , and mass radon exhalation rates (17.50 and 18.00 mBqm^{-2}h^{-1}) respectively [25].
– The log-normal distributions for histogram activity concentrations for ^{222}Rn, and ^{226}Ra along the groundwater samples are presented in fig. 4(a-b). This is indicated by the values of kurtosis which are -0.97, and – 0.97 respectively for ^{222}Rn , and ^{226}Ra which are lower than the expected value of 3, and the values of skewness are 0.28, and 0.27 respectively for ^{222}Rn , and ^{226}Ra which also are larger than the expected value of 0.00 where the normal distribution is a symmetric distribution with well-behaved tails, this is indicated by the skewness which is 0.00, and the kurtosis is 3 [26]. Also the log-normal distributions for the histogram to both areal exhalation rate (kurtosis = –0.97, and skewness = = 0.28), and mass exhalation rate (kurtosis = –0.97, and skewness = 0.28) as shown in tab. 2, and are presented in fig. 4(c and d).
– Figures 5(a-c) show a strong linear relationship between effective radium with both areal, mass exhalation rate and radon concentrations with a correlation coefficient of 1.00.

Table 1. The pH, ^{222}Rn concentration, effective radium, areal exhalation rate, mass exhalation rate, and annual effective dose for groundwater samples

Well no.	Sample code	pH	^{222}Rn [Bqm^{-3}]	Effective radium [BgL^{-1}]	Areal exhalation rate [mBqm^{-2}h^{-1}]	Mass exhalation rate [mBqkg^{-1}h^{-1}]	Annual effective dose due to ingestion [mSv]	
							^{222}Rn	^{226}Ra
1	1A	6.80	39.50 0.63	6.72 0.40	32.72 1.97	27.6 1.67	0.29 0.02	1.40 0.08
	1B	7.00	11.00 0.54	1.87 0.37	9.11 1.81	7.70	0.08 0.01	0.40 0.07
	1C	7.00	36.50 0.51	6.21 0.32	30.24 1.58	25.54 1.33	0.27 0.02	1.30 0.07
2	2A	7.00	18.00 0.25	3.06 0.18	14.91 0.88	12.59 0.74	0.13 0.01	0.60 0.04
	2B	6.80	7.60 0.69	1.29 0.47	6.30 2.26	5.32 1.91	0.06 0.02	0.30 0.08
	2C	6.80	11.00 0.54	1.87 0.37	9.11 1.81	7.70 1.52	0.08 0.01	0.40 0.07
3	3A	7.00	20.00 0.17	3.40 0.13	16.57 0.61	13.99 0.52	0.15 0.02	0.70 0.02
	3B	6.80	18.60 0.23	3.16 0.17	15.41 0.80	13.01 0.67	0.14 0.02	0.60 0.04
	3C	7.00	38.80 0.61	6.60 0.38	32.14 1.88	27.14 1.59	0.28 0.02	1.30 0.07
4	4A	6.80	24.70 0.02	4.20 0.00	20.46 0.01	17.28 0.01	0.18 0.02	0.90 0.01
	4B	7.00	33.00 0.37	5.61 0.23	27.34 1.11	23.09 0.94	0.24 0.01	1.10 0.03
	4C	6.80	36.50 0. 51	6.21 0.32	30.24 1.58	25.54 1.33	0.27 0.01	1.30 0.07
5	5A	7.00	41.70 0.73	7.09 0.46	34.55 2.27	29.17 1.91	0.3 0.02	1.40 0.08
	5B	7.00	18.60 0.23	3.16 0.17	15.41 0.80	13.01 0.67	0.14 0.02	0.60 0.04
	5C	6.80	32.90 0.36	5.59 0.22	27.26 1.10	23.02 0.93	0.24 0.01	1.10 0.03
6	6A	7.00	41.70 0.73	7.09 0.46	34.55 2.27	29.17 1.91	0.30 0.02	1.40 0.08
	6B	6.80	13.50 0.44	2.30 0.31	11.18 1.48	9.44 1.25	0.10 0.01	0.50 0.05
	6C	7.00	24.7 0.02	4.20 0.02	20.46 0.01	17.28 0.01	0.18 0.01	0.90 0.01
7	7A	7.00	5.00 0.79	0.85 0.54	4.140 2.60	3.50 2.20	0.04 0.01	0.20 0.12
	7B	6.80	11.17 0.54	1.90 0.37	9.25 1.79	7.81 1.51	0.08 0.01	0.40 0.07
	7C	6.80	34.00 0.41	5.78 0.25	28.17 1.24	23.79 1.05	0.25 0.01	1.20 0.05
8	8A	6.80	17.80 0.26	3.03 0.19	14.75 0.90	12.45 0.76	0.13 0.01	0.60 0.04
	8B	7.00	18.80 0.22	3.20 0.16	15.57 0.77	13.15 0.65	0.14 0.01	0.70 0.02
	8C	7.00	18.00 0.25	3.06 0.18	14.91 0.88	12.59 0.74	0.13 0.01	0.60 0.04
9	9A	6.80	45.00 0.86	7.65 0.55	37.28 2.70	31.48 2.28	0.33 0.02	1.60 0.12
	9B	7.00	14.70 0.39	2.50 0.27	12.18 1.32	10.28 1.11	0.11 0.01	0.50 0.06
	9C	7.00	19.70 0.18	3.35 0.14	16.32 0.65	13.78 0.55	0.14 0.01	0.70 0.03
10	10A	7.00	33.20 0.37	5.64 0.23	27.50 1.14	23.23 0.96	0.24 0.01	1.20 0.05
	10B	7.00	28.00 0.16	4.76 0.09	23.2 0.45	19.59 0.38	0.20 0.01	1.00 0.02
	10C	7.00	22.90 0.05	3.89 0.05	18.97 0.23	16.02 0.19	0.17 0.01	0.80 0.01
11	11A	6.80	44.00 0.82	7.48 0.53	36.45 2.57	30.78 2.17	0.32 0.02	1.50 0.10
	11B	7.00	18.00 0.25	3.06 0.18	14.91 0.88	12.59 0.74	0.13 0.01	0.60 0.04
	11C	6.80	4.20 0.83	0.71 ± 0.56	3.48 2.71	2.94 2.29	0.03 0.02	0.10 0.12
12	12A	6.80	33.00 0.37	5.61 0.23	27.34 1.11	23.09 0.94	0.24 0.01	1.10 0.04
	12B	7.00	16.50 0.32	2.81 0.22	13.67 1.08	11.54 0.91	0.12 0.01	0.60 0.04
	11C	7.00	48.00 0.99	8.16 0.63	39.77 3.10	33.58 2.62	0.35 0.03	1.70 0.13
13	13A	7.00	18.80 0.22	3.20 0.16	15.57 0.77	13.15 0.65	0.14 0.01	0.70 0.03
	13B	6.80	26.00 0.08	4.42 0.04	21.54 0.18	18.19 0.16	0.19 0.01	0.90 0.01
	13C	6.80	14.70 0.39	2.50 0.27	12.18 1.32	10.28 1.11	0.11 0.01	0.50 0.06

Table 2. Summarized results for pH, Rn-222, effective radium, areal exhalation rate, mass exhalation rate, and annual effective dose due to ingestion of radon, and radium

Statistics	pH	Rn-222 [Bqm^{-3}]	Effective radium [BqL^{-1}]	Areal exhalation rate [mBqm^{-2}h^{-1}]	Mass exhalation rate [mBqkg^{-1}h^{-1}]	Annual effective dose due to ingestion [mSv]	
						^{222}Rn	^{226}Ra
Mean	6.92	24.61	4.18	20.39	17.22	0.18	0.86
Range	6.80-7.00	12.20-32.50	2.07-5.53	10.11-26.93	8.54-22.74	0.09-0.24	0.43-1.13
Standard deviation	0.10	11.93	2.03	9.88	8.34	0.086	0.41
Median	7.00	20.00	3.40	16.57	13.99	0.15	0.70
Kurtosis	–2.04	–0.97	–0.97	–0.97	–0.97	–0.96	–0.89
Skewness	0.27	0.28	0.27	0.28	0.28	0.28	0.28
Variance	0.01	142.27	4.11	97.66	69.64	0.01	0.17

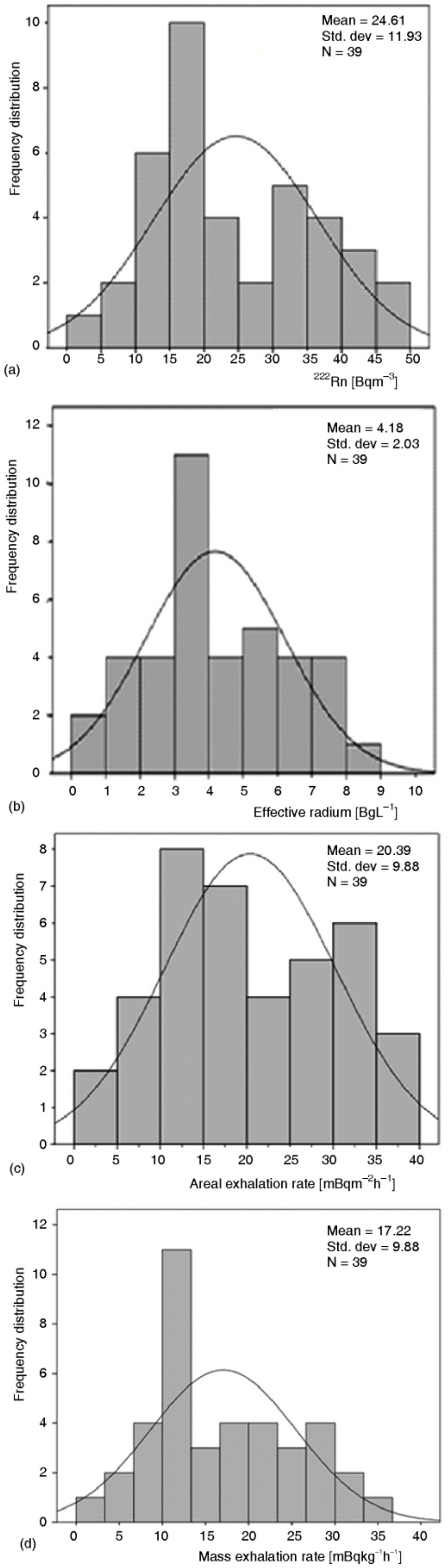

Figure 4. A histogram illustrating the change in (c) Areal exhalation rate [mBqkg^{-1}h^{-1}], (d) Mass exhalation rate [mBqkg^{-1}h^{-1}] in some groundwater samples in Najran city

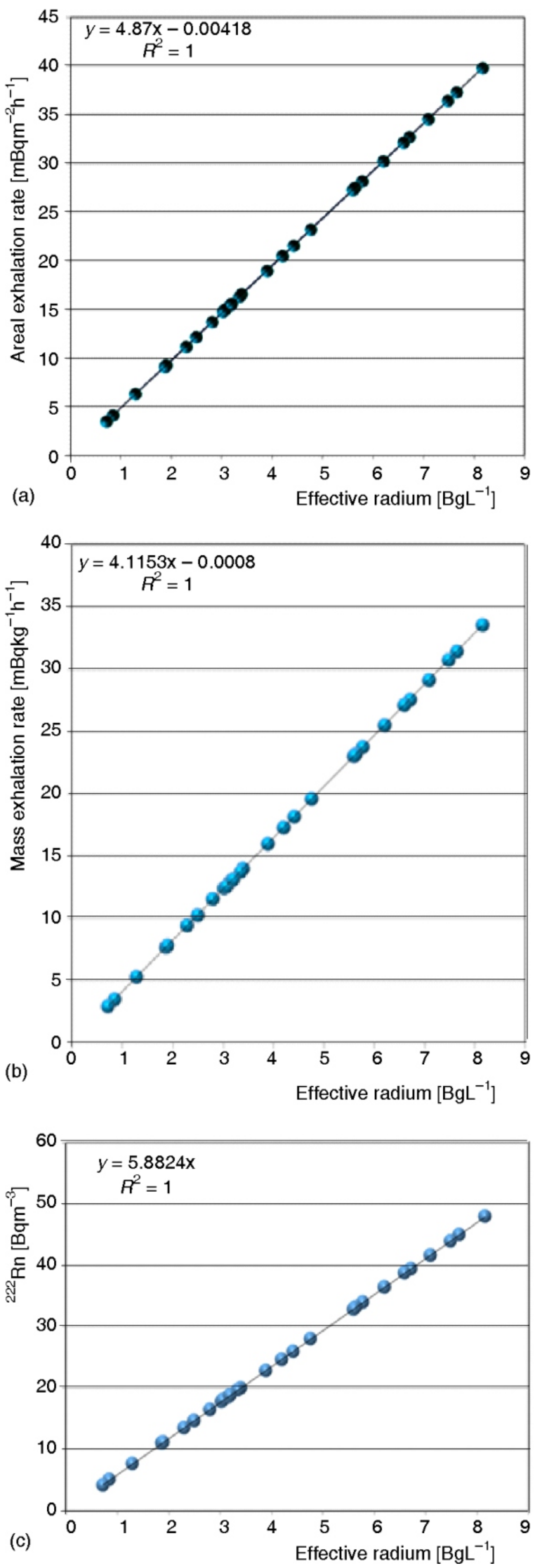

Figure 5. Linear regression of the areal exhalation rate *vs.* effective radium in groundwater wells (a), linear regression of the mass exhalation rate *vs.* effective radium in groundwater wells (b), and linear regression of effective radium *vs.* radon concentration in groundwater wells (c)

Table 3. Activity concentration of ^{222}Rn in different sources of water in Saudi Arabia, and different parts of the world

Region	Sources of water	^{222}Rn activity concentration [BqL^{-1}]	Reference
Thirteen regions in Saudi Arabia	Wells supplying drinking water	0.01- 67.40 (4.62) 0.24 - 24.90	[27]
Jeddah City, Saudi Arabia	Tap water Flush water Gallon water Mineral water Other drinking water	0.92-2.12 (1.72) 0.86-1.96 (1.54) 0.81-3.56 (2.21) 5.56-14.87 (9.92) 4.39-l 1.02 (8.06)	[28]
Central region of Saudi Arabia	Groundwater	0.89- 35.44 (8.80)	[29]
Samtah-Jazan, Saudi Arabia	Mineral water Distilled water Gallon water Tap water	15.02 9.18 6.40 3.06	[30]
Riyadh City, Saudi Arabia	Water distribution network	0.10-1.00 (0.20)	[31]
Hail province, Saudi Arabia	Groundwater	0.17-5.41 (2.60)	[32]
Al-Jawa, Saudi Arabia	Well waters	1.45-9.15 (4.49)	[33]
Libya	Drinking water samples from different water wells	1.02-6.42 (3.46)	[34]
Lebanon (many locations)	Well and spring water	0.46-49.60	[35]
Malaysia	Well water Stream water Tap water	12.40-17.00 (14.7) 6.70-9.40 (7.90) 2.70-7.00 (5.37)	[36]
Cyprus	Groundwater	0.10-5.00 (1.40)	[37]
Spain	Wells and springs	0.80-26.00 (11.40)	[38]
China	Groundwater	0.71-3735.00 (229.40)	[39]
Pakistan	Drinking water (Hand pump) Drinking water (motor pump)	0.042-0.125 (0.081) 0.075-0.158 (0.120)	[40]
Najran, Saudi Arabia	Well groundwater	12.20-32.50 (24.61)	Present study

– The mean annual effective dose for ingestion of ^{222}Rn is 0.18 0.01 mSv with a ranged of 0.09 0.02 to 0.24 0.03 mSv, as shown in tab. 2.
– The annual effective dose for ingestion of radium is 0.86 0.06 with a ranged of 0.43 0.07 to 1.13 0.04 mSv, as shown in tab. 2.
– By comparing our data for radon concentrations in this work with the results of other studies for different parts in in the world, it's obvious that ,the average value of the radon concentration in groundwater in Najran city was lower when compared to the values reported in China, and was higher than the values reported in Pakistan as shown in tab. 3.
– These results indicated that the calculated annual effective dose due to ingestion of radon, and radium in water are within the average value 0.10 mSv reported by the WHO [18].
– In this study neutral-pH values were most commonly found for all groundwater samples, and the range of pH of the groundwater samples in this study lies within the WHO guidelines (6.50-9.50) [41].

CONCLUSION

The present study showed that the average annual effective dose due to ingestion of radon for all the groundwater samples was below the safe limit of the WHO (WHO, 2004), and for ^{226}Ra it was slightly higher than this recommended value. Also, this study showed that there is a linear relationship between effective radium contents and radon concentrations with strong correlation (R^2 = 1.00) in all groundwater samples in the Najran region. Finally, our results clearly show that groundwater from some wells in the Najran region is safe in so far as the health hazards of radon. There was a wide range of radon concentrations in groundwater wells from 12.20 0.83 to 32.5 0.99 Bqm^{-3} with an average of 24.61 1.63 Bqm^{-3}, and this range of radon concentrations are is within the safe limit recommended by UNSCEAR.

ACKNOWLDGMENTS

This project was funded by the Deanship of Scientific Research (DSR), Najran University, Najran, under Grant No. (Nu/ESCI/14/017).The authors, therefore, greatly acknowledge with thanks DSR, Najran University for technical and financial support.

AUTHORS' CONTRIBUTIONS

Authors A. M. Abdalla and T. I. Al-Naggar collected the samples, carried out experiments, analysed and discussed the results, wrote the manuscript, drew the figures, and prepared the tables.

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Received on August 31, 2016
Accepted on May 3, 2017

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КОНЦЕНТРАЦИЈЕ АКТИВНОСТИ ^{222}Rn У БУНАРИМА ПОДЗЕМНИХ ВОДА У НАЈРАН СИТИЈУ, САУДИЈСКА АРАБИЈА

У раду се детаљно разматра јачина ексхалације радона, ефективни садржај радијума и доза зрачења из бунара подземних вода у Најран Ситију у Саудијској Арабији. Испитивање концентрације радона у подземним водама спроведено је применом пасивне технике мерења, при којој радон пасивно дифундује у детектор. Добијени резултати показују да је брзина ексхалације радона исказана кроз површину и масу линеарно корелирана са ефективном количином радијума у подземним водама (коефицијент корелације $R^2 = 1$). Такође, већина измерених концентрација радона је у дозвољеним границама прописаним извештајем UNSCEAR 1993, а средње годишње ефективне дозе које потичу од радијума и радона су 180 μSv и 860 μSv, респективно.

Кључне речи: концентрација радона, CR-39 детектор, годишња ефективна доза, подземна вода