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# Radon Concentrations Measurement for groundwater Using Active Detecting Method

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#### Abstract

On global scale, groundwater has been gaining increasing attention as essential and vital water resource. Its demand has been rising rapidly in the last several decades with the overpopulation and enhanced standards of living. In recent years, a great interest arose towards the natural radioactivity in water. Radon concentrations were measured in thirty groundwater samples from Qassim area, Saudi Arabia by using RAD7 an electronic radon detector connected to a RAD- H<sub>2</sub>O accessory (Durridge Co., USA). The measured radon concentration ranges from 0.76 Bq/l to 9.15 Bq/l with an average value of 3.56 Bq/l. The measured values of radon concentration are well in the range within the EPA's maximum contaminant level (MCL) of 11.1 Bq L<sup>-1</sup>. The total annual effective dose resulting from radon in groundwater from Buraydah area were significantly lower than the UNSCEAR and WHO recommended limit for members of the public of 1 mSv year<sup>-1</sup>. The measured values for underground water from the study area suggest that the area is safe for farmers and there is no significant threat to the population as per as radon concentration is concerned.

Keywords: Radon; Groundwater; Annual effective dose; Buraydah.

### 1. Introduction

Radioactive isotopes in nature occur both in the atmosphere and in the lithosphere. The most important radioactive series in the lithosphere are the uranium and thorium series.

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The first members of these series and their decay products are leached out of the rocks and dissolved by groundwater to varying degrees. The gaseous radioactive member of the uranium series, radon is easily dissolvable in water and is enriched in relation to other members of the series. Hence, the radioactivity of groundwater is mainly contributed by radon. Radon-222 and Radon-220, the gaseous daughter products of U-238 and Thorium respectively accounts for more than 50% of the human exposure due to natural radiation. Water is the most important source for life and makes up 70 - 75% of total body weight. While 70% of the world's surface is covered by water, only 0.3 % of the total water resources on earth are suitable for drinking and daily use. Human being provides their water needs from surface water and ground water resources. Ground water has more radioactive contents than surface water since it passes through rocks and soil formations, dissolves many compounds, minerals and radioactive materials. Radioactive isotopes in nature occur both in the atmosphere and in the lithosphere. The most important radioactive series in the lithosphere are the uranium and thorium series. The first members of these series and their decay products are leached out of the rocks and dissolved by groundwater to varying degrees.

In recent years, a great interest arose towards the natural radioactivity in water [1-3]. Activity concentration of the <sup>222</sup>Rn radionuclide was determined in drinking water samples from the Sothern Greater Poland region by liquid scintillation technique. The measured values ranged from 0.42 to 10.52 Bq/l with the geometric mean value of 1.92 Bq/l. The calculated average annual effective doses from ingestion with water and inhalation of this radionuclide escaping from water were 1.15 and 11.8 µSv/y, respectively. Reference [4] measured <sup>222</sup>Rn in groundwater and surface seawater during a full tidal period, estimated <sup>222</sup>Rn activity along the coast of Xiangshan, Zhejiang, China. <sup>222</sup>Rn activity in Xiangshan coast was in range of  $2.4 \times 104 - 1.7 \times 105$  Bq/m<sup>3</sup> with an average of 9.6  $\times$  104 Bq/m<sup>3</sup> for groundwater; 0.2  $\times$  102 - 2.8  $\times$  102 Bq/m<sup>3</sup> with an average of 1.1  $\times$  102 Bq/m<sup>3</sup> for surface seawater. The authors in Reference [5] studied the distribution of radon in ground and surface water samples in Sankey Tank and Mallathahalli Lake areas, the mean radon activity in surface water was 7.24  $\pm$  1.48 and 11.43  $\pm$  1.11 Bq/l, respectively. The average radon activities in groundwater ranged from 11.6  $\pm$  1.7 to  $381.2 \pm 2.0$  Bq/l and  $1.50 \pm 0.83$  to  $18.9 \pm 1.59$  Bq/l, respectively. Correa et al. in Reference [6] analyzed concentration activity of <sup>222</sup>Rn activity concentration in well water. About 70% of water samples from monitored wells presented <sup>222</sup>Rn concentration values above the limit of 11.1 Bq/l recommended by the United States Environmental Protection Agency USEPA. Voltaggio and Spadoni Passive in Reference [7] studied the efficiency of <sup>222</sup>Rn gas accumulators made of polydimethylsiloxane (PDMS) mixed with activated Carbon (AC) for sampling Rn in water. The high Rn volumetric enrichment factor in PDMS-AC disks respect to water resulted in about 206:1, so lowering detection limits for <sup>222</sup>Rn in water to 20 Bg/m<sup>3</sup> when the total activity of Rn progeny in disks is measured by high resolution gamma-ray spectrometry. The authors in Reference [8] estimated radon concentration in groundwater samples at different areas of the districts of SriGanganagar, Hanumangarh, Sikar and Churu in northern Rajasthan. Radon concentration in the groundwater ranged from 0.5  $\pm$  0.3 Bq/l (Chimanpura) to 85.7 $\pm$ 4.9 Bq/l (Khandela) with an average value of 9.03 $\pm$ 1.03 Bq/l. Radon concentration is well below the allowed maximum contamination level (MCL) of radon concentration in water of 11 Bq/l, proposed by Reference [9].

In Saudi Arabia, studies on natural radioactivity contents in the environments are dispersed in last few years. The authors in Reference [10] measured Twenty-nine groundwater samples, collected from Wadi Nu'man wells, Mecca Province, Saudi Arabia. The <sup>222</sup>Rn concentration ranged from 10-100 Bg/l with an average value of about 40 Bg/l. In Reference [11] Aleissa et al., measured <sup>222</sup>Rn radioactivity concentration levels in 171 well waters located in and around the city of Riyadh in Saudi Arabia. The analyses were performed by an ultra-low level liquid scintillation spectrometer equipped with an  $\alpha/\beta$  discrimination device. The measured <sup>222</sup>Rn activities of deep wells ranged from 0.34±0.05 to 3.52±0.30 Bq/l (average: 1.01±0.10 Bq/l), whereas those of shallow wells ranged from 0.72±0.08 to 7.21±0.58 Bq/l (average: 2.74±0.24 Bq/l). In Reference [12] Alabdulaaly measured radon levels in eight water supply municipalities of the Central Region of Saudi Arabia. The well water radon level was in the range of 0.89-35.44 Bq/l with an overall weighted geometric mean value of 8.80 Bq/l. Kadi In reference [13] found <sup>222</sup>Rn in some groundwater samples, the concentration of <sup>238</sup>U and <sup>222</sup>Rn has been assessed in underground water samples collected from the Makkah Al-Mukarramah area west of Saudi Arabia. Observed radon activities lie in the range 0.6-3.9 Bq/l. In Reference [14] Alabdulaaly assayed radon levels in a water distribution network of the capital city of Saudi Arabia, Riyadh. All samples have shown low radon levels with an average concentration of 0.2 Bq/l and a range values of 0.1-1.0 Bq/l. The authors in Reference [15] studied <sup>222</sup>Rn levels in the groundwater supplies of the capital city of Saudi Arabia (Riyadh). All samples have low radon levels with an average concentration of  $2.99 \pm 0.29$  and  $3.44 \pm 0.35$  Bq/l ( $61.8 \pm 7.8$  and  $92.9 \pm 9.5$  pCi/l) for the deep and shallow well waters, respectively.

### 2. Materials and Methods

### 2.1. Sampling: on-site activities

A total of 30 samples from Qassim area were Selected for investigation. The wells were purged through pumping for 10 min to ensure sample quality. All the water samples were collected in special glass bottles 250 mL capacity designed for radon in-water activity measurement ensuring minimum radon loss by degassing and without any air contact as shown in Reference [16].

#### 2.2. Laboratory measurements

<sup>222</sup>Rn measurement of ground water samples was carried out using a radon-in-air monitor RAD-7 (Durridge Co. Ltd) using RAD H2O technique (Figure 1) with closed loop aeration concept as it is cleared in Reference [17]. RAD H2O technique employs closed loop concept, consisting of three components, (a) the RAD7 or radon monitor, on the left, (b) the water vial with aerator, in the case near the front, and (c) the tube of desiccant, supported by the retort stand above as marked in Figure1a. Schematic representations of the radon-in-air monitor RAD-7 with RAD H2O given in Figure 2. The radon activity was measured using a radon-in-air monitor (RAD7) coupled with a specially fabricated closed loop of aeration system that strip/free radon from the water. The sample bottles of 250 mL were connected to the RAD-7 and the internal air pump of the radon-monitor was used for re-circulating a closed air-loop through the water sample, purging radon from the water into the air-loop. The air was re-circulated through the water continuously to extract the radon until RAD-H2O system reaches a state of equilibrium. After reaching equilibrium between water, air, and radon progeny attached to (PIPS) detector, the radon activity concentration measured in the air loop was used for calculating the initial radon-in-water concentration of the respective sample. The RAD-7 allows determination of radon-in-

air activity concentrations by detecting the alpha-decaying radon progeny <sup>218</sup>Po and <sup>214</sup>Po using a passivated implanted planar silicon detector (PIPS). The radon monitor (RAD7) uses a high electric field above a silicon semi-conductor detected at ground potential to attract the positively charged polonium daughters, <sup>218+</sup>Po ( $t_{1/2} = 3.1$  min; alpha energy = 6.00 MeV) and <sup>214+</sup>Po ( $t_{1/2} = 164 \,\mu$ s; alpha energy = 7.67 MeV), which are counted as a measure of <sup>222</sup>Rn concentration in air. At the end of the run (30 min after the start), the RAD7 prints out a summary, showing the average radon reading. The time elapsed for the sample collection and analysis corrected using the equation

 $C = C_0 e^{-\lambda t} \quad \dots \qquad (1)$ 

where C is the measured concentration, C<sub>0</sub> initial concentration (to be calculate) after the decay correction and **t** is the time elapsed since collection (days),  $\lambda = (0.693)/(t_{1/2}) = 0.181$ ,  $t_{1/2} = 3.83$  days.

## 2.3. Calculation the annual effective dose

Radon gas is the largest contributor to the collective exposition to natural radiation of the population in the world [18-19,5]. The inhalation of short-lived decay products of radon (<sup>222</sup>Rn) accounts on average about 50% of the effective equivalent dose on the human being as cleared in Reference [20]. The annual effective dose to an individual consumer due to intake of radon from drinking water is evaluated using the relationship as defined from Reference [21].

where  $D_w$  is the annual effective dose (Sv y<sup>-1</sup>) due to ingestion of radio-nuclides from the consumption of water,  $C_w$  concentration of <sup>222</sup>Rn in the ingested drinking water (Bq L<sup>-1</sup>),  $C_{Rw}$  annual intake of drinking water (L y<sup>-1</sup>),  $D_{cw}$  is the ingested dose conversion factor for <sup>222</sup>Rn (Sv Bq<sup>-1</sup>) [22-23]. For calculation of effective dose, a dose conversion factor of 5 x 10<sup>-9</sup> Sv Bq<sup>-1</sup> suggested by the United Nations Scientific Committee on the Effects of Atomic Radiation has been used [24-25]. Annual effective dose due to intake of <sup>222</sup>Rn from drinking water has been calculated considering that an adult (Age >1 8 year), on average, takes 730 L water annually. Following ingestion of <sup>222</sup>Rn dissolved in drinking water, annual effective doses ( $\mu$ Sv y<sup>-1</sup>) and effective doses per liter (nSv L<sup>-1</sup>) were calculated.

## 3. Results and Discussion

The measurements for radon concentration have been carried out for groundwater samples from Buraydah city, Saudi Arabia, RAD7. The radon concentrations ranged from 0.76 Bq/l to 9.15 Bq/l with an average value of 3.56 Bq/l. The obtained results are far less compared to radon results obtained by [26-29]. Hence, an attempt has been carried out in the current study to estimate the total annual effective dose resulting from radon in the sampled groundwater and it was noticed that annual effective dose-rate (AED) and effective dose-rate per liter (EDL) were varying with increase in radon concentration. The calculated effective dose per liter (EDL) and annual effective dose (AED) were ranging from 3.8 to 45.75 nSv/l and 2.77 to 33.39  $\mu$ Sv/y, respectively (Table 1). It was evident that the total annual effective doses resulting from Buraydah were

significantly lower than the recommended limit 1 mSv/y for the public [30-31].

Several national and international health organizations have determined acceptable action levels for radon concentrations. The USEPA defined a value of 11.1 Bq/l for radon concentration in water in its report in 1999 in Reference [9]. United Nations Scientific Committee on the Effects of Atomic Radiations Reference [30] defined a value of 40 Bq/l and the WHO defined in Reference [31] a value of 100 Bq/l as an action limit. Table 1 represents the overall radon concentration levels and their annual effective dose exposure. It can be seen that radon activity varies from 0.76 Bq/l to 9.15 Bq/l with an average value of 3.56 Bq/l. Although, all the samples are within the maximum contaminant level (MCL) of 11.1 Bq/l as shown in reference [9].



Figure 1: Measurement apparatus RAD 7 (RAD H2O User Manual)

The spatial variations in radon concentration could be a function of the geological structure of the area, depth of the water source and also differences in the climate and geo-hydrological processes that occurs in the area. When the measured radon concentration values are compared with the allowed maximum contamination level for radon concentration in water (which is 11.1 Bq/l), proposed by the USEPA in Reference [9], it can be seen that the present value are below this recommended value .Also, when the measured values for radon concentration are compared with the European Commission Recommendations on the protection of the public against exposure to radon in drinking water supplies which recommends action levels of 100 Bq/l for public water supplies, it can be seen that the levels we measured were below these limits.



Figure 2: A: Schematic representation of the RAD 7 instrument for measuring radon in water; B: aerator assembly

 Table 1: Radon concentration and their annual effective dose exposure in ground water from Qassim area,

 Saudi Arabia.

Sample	Rn-222	Annual Effective dose	Annual effective doses	Total Annual
N0.		rate	per liter	Effective dose rate
	$\operatorname{Bq} L^{-1}$			
		EDE ( $\mu$ Sv y <sup>-1</sup> )	EDL (nSvL <sup>-1</sup> )	$(mSv y^{-1})$
1	1.34	4.89	6.70	0.0049
2	1.28	4.67	6.40	0.0047
3	1.02	3.72	5.10	0.0037
4	0.76	2.77	3.80	0.0028
5	1.00	3.65	5.00	0.0037
6	1.08	3.94	5.40	0.0040
7	2.70	9.65	13.50	0.0097
8	2.44	8.90	12.20	0.0089
9	3.29	12.01	16.45	0.0120
10	3.17	11.57	15.85	0.0116

١٢	3.01	10.95	9.23	0.0110
	5.01	17.12	22.45	0.0171
11	4.69	17.12	23.45	0.0171
1٤	4.42	16.14	22.10	0.0161
10	4.52	16.51	22.60	0.0165
1٦	4.24	15.49	21.20	0.0155
1۷	4.44	16.22	22.20	0.0162
1^	3.20	11.68	16.00	0.0117
1۹	4.49	16.38	22.45	0.0164
۲.	9.15	33.39	45.75	0.0334
۲۱	8.26	30.14	41.30	0.0301
۲۲	5.12	18.69	25.60	0.0187
2۳	5.52	20.14	27.60	0.0201
2٤	7.88	28.75	39.40	0.0208
2°	3.40	12.40	17.00	0.0124
26	3.12	11.38	15.60	0.0114
27	2.60	09.48	13.00	0.0095
28	2.05	07.48	10.25	0.0075
29	2.15	07.84	10.75	0.0079
30	3.30	12.04	16.50	0.0120
Min	0.76	2.77	3.80	0.0028
Max	9.15	33.39	45.75	0.0334
Average	3.56	13	17.65	0.0126

The radon concentrations found in this work are presented together with comparable measurements from the rest of the world in Tables 2. Radon concentration in groundwater may vary with time because of factors such as dilution by recharge and changes in recharge area due to pumping etc. The seasonal changes may be high or low depending on the factors responsible for enrichment of radon in groundwater. A study on radon concentration in tube wells by Sonkawade et al. in Reference [32] found that de-ionization of water reduces the radon concentration. Also, the concentration of radon was found to be inversely correlated with the pH value of water samples. Various studies conducted in different terrains on the concentration of radon in groundwater indicates a direct relation between the presence of uranium and thorium in the parent rock and radon enrichment in groundwater. In tectonically disturbed areas high radon concentrations are generally related to changes in geology, soil type, and structural controls. High radon concentrations in groundwater and soil are observed above structural planes like fault, fracture, fold, and lineaments. It is used as a natural tracer in many hydro geological investigations and for quantifying submarine discharge along sea coats.

Water type	Country	Range (Bq/l)	Reference
Drinking	India	0.87-32.10	[33]
Groundwater	Brazil	0.95-36.00	[34]
Well	Turkey	0.70–31.70	[35]
Well	Mexico	1.78–39.75	[36]
Ground	Italy	1.80-52.70	[37]
Drinking	Poland	0.42-10.52	[3]
Groundwater	China	110-36.00	[4]
Groundwater	India	11.7–381.2	[5]
Groundwater	Brazil	1.6–215	[6]
Groundwater	India	0.50-85.7	[8]
Groundwater	Saudi Arabia	10-100	[10]
Groundwater	Saudi Arabia	0.34-3.52	[11]
Groundwater	Saudi Arabia	0.76-4.69	[38]
Drinking	Saudi Arabia	0.89-35.44	[12]
Groundwater	Saudi Arabia	0.76- 9.15	Present work

Table 2: Range of radon concentrations in various types of water worldwide

## 4. Conclusion

A total of 30 groundwater samples collected from Buraydah, Qassim area were examined for <sup>222</sup>Rn. The results obtained show that the radon concentration in water are below 11 Bq/l the maximum contamination level recommended from the U.S. Environmental Protection Agency. Even the effective dose per liter and annual effective dose values were varying with respect to the increase in radon concentration and were significantly lower than the UNSCEAR and WHO recommended limit for members of the public of 1 mSv y<sup>-1.</sup>

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