Journal of Natural Sciences and Mathematics
Qassim University, Vol. 8, No. 2, pp 121-132 (July 2015/Ramadan 1436H.)

Investigation of Radon Concentrations in Buraydah Municipal Water Network, Saudi Arabia

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Abstract. Radon, a naturally occurring radioactive noble gas, is the main source of the natural radiation that is received by the population. It derives from the traces of radium in rocks and can diffuse directly or as solution in water to the earth's surface. Measurements of ^{222}Rn concentration in a total of 21 water samples collected from Buraydah municipal water network investigated. The concentrations of ^{222}Rn were measured by γ-ray spectrometry using *HPGe* detector and radon detector *RAD7* H_2O techniques. The radon activity concentration is determined by taking the weighted average of the concentrations derived from γ-ray lines associated with ^{214}Pb and ^{214}Bi decay. It was found that the concentration of ^{222}Rn ranged from 1.79 ± 0.5 to 9.28 ± 1.17 *Bq/l*. These values are below 11.1 *Bq/l* the maximum contamination level recommended from the United States Environmental Protection Agency.

Keywords. Radon; Municipal water; Annual effective dose; Buraydah.

1. Introduction

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. There are 17 isotopes of radioactive radon from ^{210}Rn to ^{226}Rn, their half-life range from seconds up to days. The longer half-life is ^{222}Rn of 3.8235 days, the ^{211}Rn and ^{210}Rn with half-life of 14.6 h and 2.4 h, respectively [1]. Radon concentrations are derived from the γ-rays emitted following the decay of the radon progeny ^{214}Pb and ^{214}Bi, after reaching secular equilibrium with ^{222}Rn. The radioactive decay of radon produces floating subatomic particles that can damage living cells. When radon is inhaled, 30% of the radon progeny comes in to contact with air passage in the lung and adhere to the surfaces. The ingestion of radon from water can give rise to an additional exposure dose to the stomach and whole body. Radon is a natural tracer for studying hydrological transport processes. It is transported without substantial adsorption in the matrix. Radon in water measurements has been used to study aquifer-flow rates, estimate recharge rates and residence times of groundwater [2], and study the interaction between groundwater and surface water through discharge and mixing processes [3]. The application to aquifers requires representative sampling of groundwater. In a study of aquifer properties the emphasis is on precision and reproducibility rather than accuracy. In view of the uncertainties in geological and hydrological description of radon genesis properties of aquifers, an accuracy of 25% (systematic uncertainties) for the method is more than sufficient. The aim of this work is to optimize the measurement of radon in water via γ-ray spectrometry and model the time evolution of radon concentrations during sampling. It involves the investigation of statistical and systematic uncertainties due to sampling, measurement and analysis. In recent years, a great interest arose towards the natural radioactivity in water [4] Activity concentration of the ^{222}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. Wen *et al.*,[5] measured ^{222}Rn in groundwater and surface seawater during a full tidal period, estimated ^{222}Rn activity along the coast of Xiangshan, Zhejiang, China. ^{222}Rn activity in Xiangshan coast was in range of 2.4 ×10^4 - 1.7 × 10^5 Bq/m^3 with an average of 9.6 × 10^4 Bq/m^3 for groundwater; 0.2 × 10^2 - 2.8 × 10^2 Bq/m^3 with an average of 1.1 × 10^2 Bq/m^3 for surface seawater. Ravikumar and Somashekar[6] 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 ± 1.48 and 11.43 ± 1.11 *Bq/l*, respectively. The average radon activities in groundwater ranged from 11.6 ± 1.7 to

381.2 ± 2.0 *Bq/l* and 1.50 ± 0.83 to 18.9 ± 1.59 *Bq/l*, respectively. Correa et al., [7] analyzed concentration activity of ^{222}Rn activity concentration in well water can contribute to indoor radon concentration levels, which represent a radiation risk for the public in Curitiba, Paraná State, Brazil were performed. About 70% of water samples from monitored wells presented ^{222}Rn concentration values above the limit of 11.1 *Bq/l* recommended by the United States Environmental Protection Agency *USEPA*. The obtained activity values varied between 1.6 *Bq/l* and 215 *Bq/l* for radon concentration, and radium concentrations deviated within an interval of 0.50 Bq/l and 6.8 Bq/l. [8] Voltaggio and Spadoni Passive studied the efficiency of ^{222}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 ^{222}Rn in water to 20 Bq/m^3 when the total activity of *Rn* progeny in disks is measured by high resolution gamma-ray spectrometry. Rani *et al*.,[9] 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 ± 0.3 *Bq/l* (Chimanpura) to 85.7±4.9 *Bq/l* (Khandela) with an average value of 9.03±1.03 *Bq/l*. Radon concentration is well below the allowed maximum contamination level (*MCL*) of radon concentration in water of 11.1 *Bq/l*, proposed by (*USEPA*). In Saudi Arabia, studies on natural radioactivity contents in the environments are dispersed in last few years. Shabana *et al.*,[10] measured radon (^{222}Rn) twenty-nine groundwater samples, collected from Wadi Nu'man wells, Mecca Province, Saudi Arabia. The water contains high concentrations of ^{222}Rn and considerable levels of natural uranium. The ^{222}Rn concentration ranged from 10-100 *Bq/l* with an average value of about 40 *Bq/l*. Aleissa *et al.*, [11] measured ^{222}Rn radioactivity concentration levels in well waters located in and around the city of Riyadh in Saudi Arabia. Water samples were collected from 171 wells in six different

locations. The analyses were performed by an ultra-low level liquid scintillation spectrometer equipped with an α/β discrimination device. The measured ^{222}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*). The ^{222}Rn concentrations levels were found to be in compliance with the proposed national limits of 11.1 *Bq/l* and depend on the water source. Alabdulaaly[12-14] measured radon levels in eight water supply municipalities of the Central Region of Saudi Arabia. Samples were collected from 77 wells and 6 treatment plants supplying drinking water to over 500000 inhabitants. 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*. Most of the raw water radon was removed by the plants treatment processes. Aeration and filtration resulted in 60.5% radon removal compared to a removal in the range of 78.7-96.5% in treatment plants containing reverse osmosis or electrodialysis processes along with aeration. The plants product water contains radon levels in the range of 0.15-5.71

Bq/l whereas two water systems with no treatment contain levels of 2.07 and 1.19 *Bq/l*. Alabdulaaly[13] assayed radon levels in a water distribution network of the capital city of Saudi Arabia, Riyadh, the ^{222}Rn levels in the city distribution network was carried out on 89 samples were collected from different locations representing the city districts. 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*.

El-Taher[15] carried out Measurement of radon concentrations and their annual effective dose exposure in groundwater from Qassim province, Saudi Arabia. The measured radon concentration ranges from 0.76 to 4.69 Bq/l with an average value of 2.8 *Bq/l*. The measured values of radon concentration are well in the range within the USEPA's Maximum Contaminant Level (MCL) of 11.1 *Bq/l*. The total annual effective dose resulting from radon in groundwater of Qassim province were significantly lower than the (United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [16], World Health Organization (WHO) [17] and Ludovic and Céline [18] in a review of ICRP recommendations, all recommended limit for members of the public of 1 *mSv/y*. Kadi[19] found ^{222}Rn in some groundwater samples, the concentration of ^{238}U and ^{222}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*.

2. Materials and Methods

The aquifers supply the main municipal water network with groundwater that, are contaminated with naturally radioactive materials. All these aquifers represent the source of groundwater in Buraydah Region are supplied from Al-Saq huge aquifer. Therefore attention must be paid to investigate the effect of contaminated water on human health. The present study is intended to explore levels of radioactive elements radon ^{222}Rn in municipal water network from Buraydah city. Radiological study to estimate the annual effective dose will be carried out, assessment the contribution of radioactive radon in the accumulated exposure dose. Water samples collected from 19 samples scattered across Buraydah city and analyzed for routine physiochemical properties according to the standard methods for the examination of waste water and waters [20].

In this study radon in water was measured in a counting geometry of a screw-top Marinelli beaker (*AEC-Amersham*, code *NQB2205*, volume 1.3 L) filled to the top. The γ-ray spectra were measured with a *Canberra* p-type *HPGe* detector of 50% relative efficiency, and 1.95 *keV FWHM* resolution at the ^{60}Co transition line of 1332.5 *keV*. Canberra standard well-type lead shield is provided and the detector equipped with standard nuclear electronics to process the detector signals. Preamplifier is internally connected under vacuum within the *HPGe* detector aluminum cap. Digital Spectrum Analyzer (*DSA-1000*) is connected to the *HPGe*, it provides the detector with voltage needed for it to work. In addition, output signal from the preamplifier is connected to it for the amplification process and eventually the signal produced by the

DSA-1000 is transferred to a computer equipped with a 8192 channels *MCA*. The system has to be first energy calibrated using standard sources e.g. ^{137}Cs and ^{60}Co. Radon concentrations were derived from the intensity of six γ-lines emitted by radon progeny: E_γ = 295 and 352 *keV* of ^{214}Pb, and $E\gamma$ = 609, 934, 1120 and 1764 *keV* from ^{214}Bi. Measurements commenced after radioactive equilibrium between radon and its short-lived progeny had been achieved (5 *h*). Each sample was counted for 3 h. In addition, tight sealed samples stored for 3 weeks, to measure the radon in-growth from ^{226}Ra, and counted for a minimum of 10 h.

For this set-up the minimum detectable activity (*MDA*), based on the E_γ= 352 *keV* γ-line and a counting time of 2 *h*, was found to be 0.2 *Bq/l* (3σ). This value is one to two orders of magnitude lower than the measured values. The reported radon concentration is the weighted average for the concentration of the individual γ-lines. The detector-efficiency calibration was carried out according to the method reported by [21, 22]. The relative, full-energy peak (*FEP*) efficiencies of E_γ= 295, 352, 609, 934, 1120 and 1764 *keV* were determined from groundwater samples of unknown ^{222}Rn concentration. This procedure involves the listed branching ratios [1] and the assumption that coincident summing is negligible.

^{222}Rn measurement of water samples was carried out using a radon detector *RAD7* (*Durridge Co. Ltd*) using *RAD H2O* technique. To validate the results radon concentrations measured by γ-spectrometry using the continuous radon monitor, *RAD7 Durridge*, estimate the correlation factor between the *HPGe* and the *RAD7* results. The radon activity was measured using a radon detector (*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-H_2O system reaches a state of equilibrium. After reaching equilibrium between water, air, and radon progeny attached to the 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 ^{218}Po and ^{214}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

3. Calculation the Annual Effective Dose

Radioactive radon gas is the major contributor to the collective exposure from natural radioactivity delivered to the population in the world [6, 23, 24]. The inhalation of short-lived decays products of radon (^{222}Rn) accounts for about 50% of the effective equivalent dose to the human being [6]. The annual effective dose to an individual consumer due to intake of radon from groundwater is evaluated using the relationship [23].

$$D_w = C_w \cdot C_{rw} \cdot D_{cw} \quad \textbf{(1)}$$

where D_w is the annual effective dose (*Sv/y*) due to ingestion of radio-nuclides from the consumption of water, C_w concentration of ^{222}Rn in the ingested drinking water (*Bq/l*), C_{rw} annual intake of drinking water (l/y), D_{cw} is the ingested dose conversion factor for ^{222}Rn (*Sv/Bq*) [25]. For calculation of effective dose, a dose conversion factor of 5 x 10^{-9} *Sv/Bq* suggested by the International Commission on Radiological Protection (*ICRP*) has been used [26]. Annual effective dose due to intake of ^{222}Rn from drinking water has been calculated considering that an adult (Age > 18 year), on average, takes 730 *l* water annually [27]. Following ingestion of ^{222}Rn dissolved in drinking water, annual effective doses (*μSv/y*) and effective doses per liter (*nSv/l*) were calculated.

4. Results and Discussion

The average ratio of radon concentrations derived from the 609 *keV* γ-line indicates that the effect of coincident summing for this γ-ray line worthwhile to be considered. In regard to the uncertainties in the ratios that derived from the other γ-ray lines, will change the overall effect on the average a few percent on maximum higher. By benefit from using the efficiency transfer we could neglect the effect of coincidence summing [21]. The *HPGe* radon concentration measurements were checked further in two ways. The first way involves extracting the ^{222}Rn decay constant λ. For this, four water samples were measured several times over 4 days using the *HPGe* detector. Depending on the ^{222}Rn concentration, the measurement time for these samples ranged between 7 and 10 h. The decay of the ^{222}Rn concentrations was fitted with an exponential decay curve. The ^{222}Rn decay constants for the four samples were found to be $(2.51 \pm 0.05)\times10^{-6}$, $(2.11 \pm 0.02)\times10^{-6}$, $(2.08 \pm 0.11)\times10^{-6}$ and $(2.27 \pm 0.10)\times10^{-6}$ s^{-1}, with a weighted average of $(2.16 \pm 0.08) \times10^{-6}$ s^{-1}, which is consistent with the known λ-value of 2.10×10^{-6} s^{-1}. This implies that there is no significant ^{222}Rn loss during the measurement process. The second way is to validate these results for radon concentrations by comparing it with those measured by α-spectrometry using the continuous radon monitor, *RAD7* Durridge as another mean. Water samples were collected in large volume containers (~3 *l*), split into sub-samples, and decanted into Marinelli beakers and glass vials for the *HPGe* and the *RAD7* measurements, respectively.

The radon concentrations ranged from 1 to 5.3 *Bq/l* and the correlation factor between the *HPGe* and the *RAD7* results was found to approach 99%. This result also supports the insignificance of coincidence summing effects for this study. The obtained results are far less compared to radon results obtained by [28-31]. 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 9.0 to 46.4 *nSv/l* and 5.29 to 33.39 *μSv/y*, respectively (Table 1). It was evident that the total annual effective doses resulting from radon in groundwater from Buraydah were significantly lower than the recommended limit 1 mSv/y for the public [16, 17]**.**

The radon concentrations found in this work are presented together with comparable measurements from the rest of the world in Table 2. 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 [32]. United Nations Scientific Committee on the Effects of Atomic Radiations [16] defined a value of 40 Bq/l [16] and the *WHO* defined a value of 100 *Bq/l* as an action limit [17]. Table 1 represents the overall radon concentration levels and their annual effective dose exposure. It can be seen that radon activity varies from 1.79 *Bq/l* to 9.28 *Bq/l* with an average value of 4.73 *Bq/l*. Although, all the samples are within the maximum contaminant level (*MCL*) of 11.1 *Bq/l* [32]. 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 [32], 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. In Table 2, the values obtained here are compared with those of reported in the literature from other countries.

Table (1). Radon concentration and their annual effective dose exposure in municipal water network from Buraydah City, Saudi Arabia

Sample No.	Rn-222 (*Bq/l*)	Rn-222 (*pCi/l*)	Annual effective dose-rate per liter EDL (*nSv/l*)	Annual Effective dose-rate AED (*µSv/y*)
1	1.91±0.16	51.5	9.5	6.95
2	3.30±0.23	89.1	16.5	12.04
3	2.33±0.12	63.0	11.7	8.51
4	2.38±0.10	64.2	11.9	8.67
5	3.69±0.26	99.7	18.4	13.47
6	8.70±0.27	235.2	43.5	31.76
7	3.79±0.27	102.4	18.9	13.83
8	3.35±0.14	90.4	16.7	12.21
9	1.79±0.09	48.4	9.0	6.54
10	4.21±0.35	113.8	21.1	15.37
11	2.50±0.16	67.6	12.5	9.14
12	5.31±0.15	143.5	26.6	19.38
13	9.28±0.21	250.7	46.4	33.86
14	8.43±0.26	227.8	42.1	30.77
15	5.31±0.18	143.5	26.5	19.38
16	5.73±0.22	154.9	28.7	20.92
17	7.36±0.28	199.0	36.8	26.87
18	4.77±0.15	128.9	23.8	17.41
19	4.14±0.29	111.9	20.7	15.11
20	7.35±0.30	198.8	36.8	26.85
21	3.73±0.26	100.7	18.6	13.61
Min	1.79±0.50	48.4	09.0	6.54
Max	9.28±1.17	251	46.4	33.86
Average	4.73	127.86	23.65	17.27

Table (2). Range of radon concentrations in various types of water worldwide

Water type	Country	Range (*Bq/l*)	average annual effective doses (*μSv/y*)	Reference
Drinking	Poland	0.42-10.52	1.53-38.40	[4]
Groundwater	China	110-36.00	36.5-131.4	[5]
Surface water Groundwater	India	7.24–11.43 11.7–381.2	26.43-41.72 42.71-1391.38	[6]
Groundwater	Brazil	1.6–215	5.84-784.75	[7]
Groundwater	India	0.50–85.7	1.83-312.81	[9]
Groundwater	Saudi Arabia (Mecca province)	10-100	36.5-365	[10]
Groundwater	Saudi Arabia (Riyadh province)	0.34-3.52	1.28-12.85	[11]
Groundwater	Saudi Arabia (Qassim province)	0.76- 4.69	2.77-17.12	[15]
Drinking	Saudi Arabia (Riyadh province)	0.89-35.44 0.10-1.00	3.25-129.36 0.37-3.65	[12, 13]
Drinking	Saudi Arabia (Qassim province)	1.45- 9.15	5.29-33.40	Present work

5. Conclusion

A total of 21 water samples from Buraydah municipal water network, that are driven from wells, were examined for ^{222}Rn. The results obtained show that the radon concentration in water are below 11.1 *Bq/l* the maximum contamination level recommended from the U.S. Environmental Protection Agency. The total radon concentrations for all samples exceed 1.85 *Bq/l* as minimum contamination levels up to 9.26 *Bq/l* as a maximum contamination level. The measurement of radon by γ-ray spectrometry using an HPGe detector and Marinelli beakers has been investigated. The results show that the radon concentration in water can be found with good precision (*5%*) in a reasonable time by taking the weighted average of radon concentrations derived from six γ-lines associated with ^{214}Bi and ^{214}Pb decay. The absolute accuracy of the method approach *3%*.

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التحقيق في تركيزات غاز الرادون في شبكة مياه البلدية لمدينة بريدة، المملكة العربية السعودية

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قسم التغذية وعلوم الاطعمة، كلية التصاميم والإقتصاد المنزلي، جامعة القصيم، مبنى ٣٨٩٧، بريدة ٨٣١٤-٥٢٣٨٣، المملكة العربية السعودية

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ملخص البحث. هذه الدراسة تهدف الى تقدير الرادون وهو غاز مشع من الغازات النبيلة التي تحدث بشكل طبيعي، وهو المصدر الرئيسي للإشعاع الطبيعي التي يتلقاها الجمهور. ينشأ هذا الغاز نتيجة تحلل عنصر الراديوم في الصخور والذي يمكن ان ينتقل مباشرة للمياه الجوفية أو بمزج هذه المياه مع المياه على سطح الأرض. التحقيق قياسات تركيز ^{٢٢٢}Rn في عدد ٢١ عينة مياه تم جمعها من شبكة مياه البلدية بمدينة بريدة. تم قياس تركيزات ^{٢٢٢}Rn باستخدام المطياف الجامي بواسطة كاشف (HPGe) وكاشف الرادون H2O RAD7. تم تحديد تركيزات غاز الرادون النشط بأخذ متوسط مرجح لتركيزات المستمدة من خطوط الاضمحلال لأشعة جاما المرتبطة بالنظيرين ^{٢١٤}Pb و^{٢١٤}Bi. وقد وجد أن تركيز ^{٢٢٢}Rn تراوحت بين ١,٧٩ ± ٠،٥ حتي ٩،٢٨ ± ١,١٧ بيكريل / لتر. هذه القيم هي أقل من ١١,١ بيكريل / لتر الحد الأقصى لمستوى التلوث الموصى بها من وكالة حماية البيئة في الولايات المتحدة.

كلمات استدلالية: الرادون. مياه البلديات؛ الجرعة الفعالة السنوية؛ بريدة.