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Study of Radon Concentrations in some of Public Ground Water Wells, Sana'a - Yemen.

Ahmed Khalid Abdul-rahim*, Najwa Ahmed Al-Maqtari

Physics Department, Sana'a University, Sana'a, Republic of Yemen

*Corresponding author: aabdulra@gmail.com

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Abstract: This study aims to assess radon-222 concentration levels in public ground water wells in Sana'a city, Yemen. Forty-three well water samples were collected from the study area. The alpha spectroscopy method was used to measure the samples using a RAD7 detector. The radon concentrations were in the range from 0.82 ± 0.38 Bq/L to 38.73 ± 2.98 Bq/L. A moderate correlation of radon levels with aquifer type was observed. Also, weak correlations of radon levels with electric conductivity, pH, and temperature, and no correlation with measured heavy metals were observed. Annual effective dose rates (E) were calculated and were found to be in the range from 0.006 to 0.283 mSv/y. Also, ion analysis of heavy metals was carried out. The analysis shows the maximum concentrations of copper, zinc, arsenic, and lead in groundwater samples were 0.5 mg/L, 0.58 mg/L, 0.7 mg/L, and 0.5 mg/L respectively.

Keywords: Alpha spectroscopy, Concentration, Effective dose, Ground water, Heavy metals, Radon222

1. Introduction:

Radon, a water soluble isotope, is the number one source of natural radioactive substance that is present in groundwater. Radon is a radioactive hazardous noble gas produced by the decay of radium-226. It is a member of the ^{238}U series and an alpha emitting isotope with a half-life of 3.83 days [1]. The natural source of ionizing radiation contributes to more than 70% of the total annual radiation dose rate to people in which about 54% of it is due to radon and its decay products [2]. The global potential assessment of radon release from groundwater is about 500 million curies per year [3].

High concentrations of radon is a health hazard for humans. Knowing radon concentration levels in groundwater helps decrease the risk of developing serious health problems. If radon is inhaled into the lungs, the emitted alpha particles can damage cellular DNA and can cause lung cancer. When high concentrations is ingested, it raises risk of stomach and gastrointestinal cancer [4,19]. The World Health Organization (WHO) has recommended that the concentration of radon-222 in drinking water should not exceed 100 Bq/L [5], while the United States Environmental Protection Agency (EPA) has suggested a limit of 11.1 Bq/L [6, 7, 18, 19]. In this study, radon concentrations in public groundwater wells in Sana'a - Yemen were measured and the consequence health hazards were estimated by calculating the

annual effective dose rates H_E . This study is concerned with groundwater resources because the concentrations of radioactive isotopes in groundwater are higher than in surface water. The higher concentrations in groundwater are due to its contact with igneous rocks and sedimentary beds [8]. The existence of radon in groundwater depends on the existence of uranium (^{238}U) series members in the soil and rocks of the well. Therefore, the occurring of natural radioactivity including radon in groundwater depends on the local geological nature and characteristics of the soil and rocks that the well was composed of [9]. Also in this study, certain heavy metals (lead, arsenic, copper, and zinc) in groundwater samples were measured.

2. Experimental methods:

2.1 Study area:

The study area covered the public groundwater wells that supply Sana'a, Yemen. Sana'a is between latitudes $15^{\circ}16'$ and $15^{\circ}30'N$ and between longitudes $44^{\circ}09'$ and $44^{\circ}16'E$, as shown in figure 1, the study area covers about 259 km² and consists of four main well fields: the Western, Eastern, Musayek and Asser well field. Samples were collected from 43 wells.

Sana'a basin is an inter-mountain plain situated in the central Yemen high lands. Sana'a basin contains the following major aquifers: quaternary alluvium, quaternary volcanic, tertiary volcanic, cretaceous (Tawilah) sandstone,

and Jurassic (Amran) limestone. The cretaceous (Tawilah) sandstone aquifer is nowadays the most important source of water in the area [10, 11].

2.2 Sampling:

Eleven samples from the Western well field, 8 samples from the Eastern, 14 samples from Musayek, and 10 samples from Asser were collected. Figure 1 shows the site coordinates of the samples in an administrative division map.

For sampling, special RAD7 glass vials and other accessories/tools such as tubes, a sampling case, a camera, GPS and some forms were used. Sampling vials were washed with diluted hydrochloric acid and rinsed with distilled water. Before collecting samples, water was allowed to run for several minutes to ensure that the samples came fresh from the source.

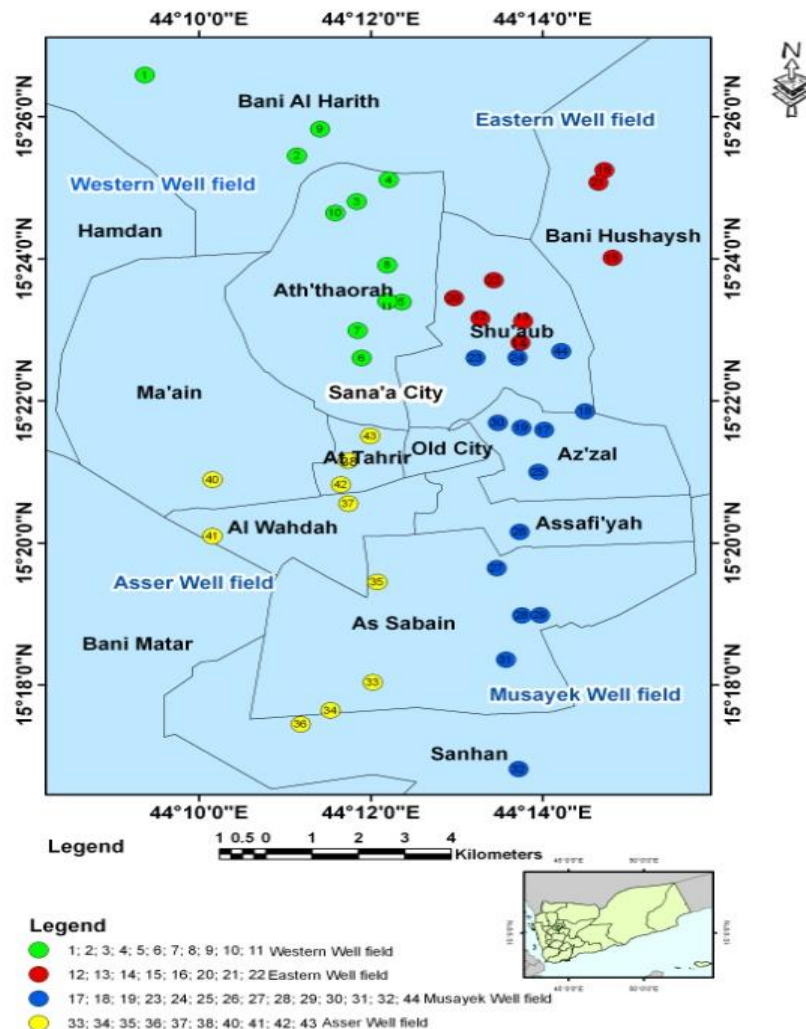


Figure 1. Coordinates of the sample sites in an administrative division

Samples were collected from the groundwater wells directly in order to be a representative of the groundwater source and were collected slowly to ensure that no bubbling happens so that no radon escapes from the samples [12]. After filling the vial, water was allowed to overflow for a short period of time to ensure it is filled with water that has not lost any radon to its surroundings.

Vials were then closed tightly while they were still under water. Finally, the samples were labeled and placed in a plastic case for later measuring.

2.3 Methods:

To measure radon-222 concentrations, the alpha spectroscopy method using the RAD7 detector with the RAD-H₂O accessory were used. A RAD7 is a simple computer-driven electronic detector of semiconductor material, usually silicon, where radon is measured in Bq/m³ or Bq/L. In alpha spectroscopy, the spectra of alpha particles emitted from the sample are analyzed. Radon (222Rn) decays to 218Po by emitting alpha particles and in turn, the 218Po decays to 214Pb by also emitting alpha particles.

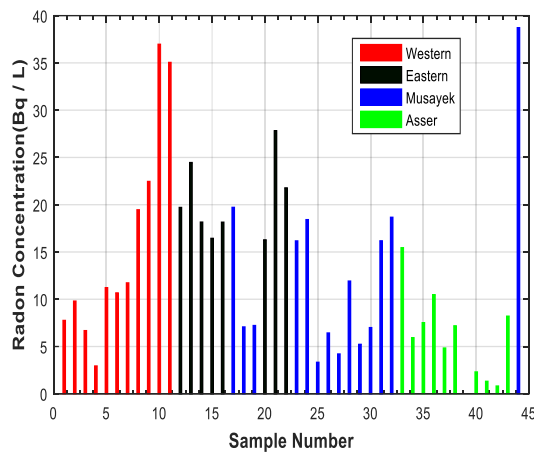


Figure 2. Radon concentration in samples from the four groundwater well fields

Emitted alpha particles from radon or its daughters enter the detector and produce electrical signals proportional in strength to the energy of the emitted alpha particles. The detector then amplifies, filters, and sorts the signals according to their strengths. By selecting the SNIFF¹ mode, the detector uses only 218Po signals to determine radon (²²²Rn) concentrations.

RAD-H₂O is a RAD7 accessory that is used to measure radon in water. It has a set of vials, an aerator, a tube, and connection hoses.

The RAD-H₂O has a closed loop aeration system in which RAD7 pumps air to aerate the sample for five minutes. The air circulates through the water and radon is extracted continuously until 218Po reaches a state of equilibrium in five minutes. The detector repeats counting for 4 five-minute cycles and at the end of the 30 minutes, the RAD7 prints out a summary showing the average radon concentration.

Health hazards caused by radon in groundwater used as drinking water was estimated by calculating the annual effective dose rate (H_E). The annual effective dose rate (H_E) due to the intake of radon from drinking water was estimated using the following equation (13):

$$H_E = C_R I_F E_D \quad (1)$$

Where:

H_E (Sv/y) is the annual effective dose rate to an individual due to ingestion of radon,

C_R (Bq/L) is the activity concentrations of ²²²Rn,

I_F is the annual intake of drinking water in L/year (L/y) where

$I_F = 730 \text{ L/year}$ or 2 L/day [14],

E_D is the ingested dose conversion factor and equals $E_D = 1 \times 10^{-8} \text{ Sv Bq}^{-1}$ [14].

In addition to measure radon(²²²Rn) activity concentration, ion analyses of heavy metals were carried out. The correlation

between radon(²²²Rn) activity concentration levels and the heavy metal concentrations in the samples were examined by calculating the Spearman rank correlation coefficients. Furthermore, the correlation of radon(²²²Rn) activity concentrations with the depth of the well, aquifer type, the power of hydrogen (pH), temperature, and electrical conductivity (EC) were also studied using Spearman correlation rank analyses. Also, analysis of the heavy metals measured in the samples were compared with the guideline values recommended by the World Health Organization, WHO.

3. Results:

3.1 Measurements of Radon concentration:

Table 1 and figure 2 show radon concentration of samples from the four groundwater well fields. For the Western well field, radon-222 activity concentrations ranged from 2.94 ± 1.13 to 36.97 ± 2.51 Bq/L with a mean value of 15.89 ± 11.37 Bq/L. For the Eastern well field, radon-222 activity concentrations varied from 16.28 ± 1.63 to 27.82 ± 1.97 Bq/L with a mean value of 20.35 ± 4.1 Bq/L. For Musayek well field, radon-222 activity concentrations ranged from 3.33 ± 0.85 to 38.73 ± 2.98 Bq/L with a mean value of 12.88 ± 9.5 Bq/L. For Asser well field, radon activity concentrations ranged from 0.82 ± 0.38 to 15.45 ± 1.24 Bq/L with a mean value of 6.41 ± 4.51 Bq/L.

3.2 Measurements of Heavy elements:

Table 2 presents the results of the ion analyses of the following heavy elements: lead, zinc, arsenic, and copper in the ground water samples. According to the World Health Organization, WHO, the concentration guideline values for drinking water for the following heavy metals should not exceed: 0.01 mg/L, 3 mg/L, 0.01 mg/L, and 2 mg/L for lead, zinc, arsenic and copper respectively [5].

Results show that the concentration of lead in 19 samples were higher than the WHO guideline value and ranged from 0.1 to 0.5 mg/L. Results also show the arsenic concentrations were higher than the WHO guideline value in 14 samples ranging from 0.1 to 0.7 mg/L. Copper and zinc concentrations in all samples were lower than the WHO guideline values.

3.3 Correlation Analyses:

Spearman's rank correlation coefficients (r_s^2) and significant levels (p-values) of radon concentrations with well depth, EC, pH, temperature, and aquifer associated with the samples are presented in table 3 and figure 3. Also table 3 and figure 4 present correlation coefficients of radon concentrations with heavy metal concentrations (copper, zinc, arsenic, and lead).

Weak correlations between radon concentrations and well depths, pH, EC, and temperature were observed. However, a moderate correlation between radon concentration levels and aquifer type were observed. On the other hand as shown in table 3 and figure 4, no correlations between radon concentrations and measured heavy metal concentrations were observed.

Table 1. Radon concentration in groundwater samples and other parameters

Sample ID#	Well Fiel	Radon Con. (Bq/L)	H _E (mSv/y)	Aqu- ifie	Well depth (m)	EC (µs/cm)	T (°C)	pH
1	W	7.76±0.77	0.057	S	292	812	22	7.4
2	W	9.80±1.13	0.072	S	323	836	28	7.5
3	W	6.68±0.87	0.049	S	400	678	27	7.4
4	W	2.94±1.13	0.021	S	345	1102	26	7.2
5	W	11.22±1.71	0.082	S	417	542	23	7.3
6	W	10.67±1.61	0.078	S	430	537	29	7.2
7	W	11.73±1.36	0.086	S	422	523	27	7.2
8	W	19.46±0.79	0.142	S	423	683	30	7.3
9	W	22.46±1.98	0.164	S	357	819	28	7.2
10	W	36.97±2.51	0.270	S	355	628	28	7.3
11	W	35.05±2.44	0.256	S	402	623	28	7.4
12	E	19.71±2.11	0.144	S	340	631	27	7
13	E	24.46±2.18	0.179	S	389	963	25	7.1
14	E	18.16±1.06	0.133	S	389	822	27	7.1
15	E	16.44±1.62	0.120	S	251	644	27	7.2
16	E	18.15±1.08	0.133	S	415	550	27	7.1
17	M	19.72±1.46	0.144	VS	660	710	27	7.1
18	M	7.07±1.05	0.052	VS	620	708	27	7.2
19	M	7.22±1.05	0.053	VS	646	728	30	7
20	E	16.28±1.63	0.119	VS	399	719	27	7.3
21	E	27.82±1.97	0.203	S	401	631	27	7
22	E	21.78±1.61	0.159	S	380	1473	22	7
23	M	16.17±2.29	0.118	S	415	868	27	7.1
24	M	18.42±0.83	0.134	V	350	814	23	7.1
25	M	3.33±0.85	0.024	VS	482	622	28	7.6
26	M	6.43±0.93	0.047	VS	480	468	28	7.4
27	M	4.21±0.51	0.031	VS	766	487	31	7.4
28	M	11.92±1.32	0.087	VS	823	501	28	7.5
29	M	5.23±1.09	0.038	VS	870	431	29	7.5
30	M	7.00±0.9	0.051	VS	630	710	28	7.2
31	M	16.18±1.67	0.118	VS	860	382	31	8.1
32	M	18.67±1.69	0.136	VS	1000	398	31	8.1
33	A	15.45±1.24	0.113	VS	884	542	30	7.4
34	A	5.93±1.47	0.043	VS	915	542	39	7.1
35	A	7.52±0.87	0.055	VS	900	425	39	8.3
36	A	10.48±1.39	0.077	VS	1010	482	29	7.3
37	A	4.84±0.53	0.035	VS	680	439	32	7.6
38	A	7.19±1.38	0.052	VS	400	457	32	7.4
40	A	2.31±0.47	0.017	V	467	536	37	7.6
41	A	1.32±0.18	0.010	VS	567	528	36	9.6
42	A	0.82±0.38	0.006	V	580	542	35	7.7
43	A	8.21±1.87	0.060	VS	620	427	34	7.6
44	M	38.73±2.98	0.283	S	418	746	27	7.3
Mean		13.53 ± 3.9	0.1					
W.F: Water Field, W:Western, E: Eastern, M:Musayek, A:Asser, S:Sandstone, V: Volcanic, VS: Volcanic Sandstone								

Table 2. Heavy metal concentrations in groundwater samples

Sample ID#	Cu (mg/L)	Zn (mg/L)	As (mg/L)	Pb (mg/L)
1	0.3	N	N	0.1
2	0.2	N	N	0.1
3	0.1	N	N	0.1
4	0.2	N	N	0.1
5	N	0.06	N	0.1
6	N	N	N	0.1
7	0.1	0.58	N	N
8	0.3	0.12	N	N
9	0.1	N	0.2	N
10	0.3	N	N	N
11	N	N	N	N
12	0.2	N	N	N
13	0.3	N	N	N
14	0.5	N	N	N
15	0.1	N	N	N
16	0.2	N	N	N
17	0.2	N	N	N
18	0.1	N	N	N
19	0.2	N	N	N
20	N	0.12	N	N
21	0.1	N	0.2	N
22	0.2	0.35	N	N
23	0.2	N	N	N
24	0.1	N	N	N
25	0.2	N	0.2	N
26	0.2	N	0.2	N
27	0.3	N	N	N
28	0.2	N	N	N
29	0.2	N	0.1	N
30	0.1	N	N	0.1
31	0.1	N	0.2	0.1
32	0.2	N	0.4	0.3
33	0.3	N	0.4	0.1
34	0.2	N	N	N
35	0.3	N	0.2	0.5
36	0.2	N	N	0.2
37	0.2	N	0.2	0.2
38	0.2	N	N	0.3
40	0.2	N	N	0.3
41	0.1	N	0.1	0.4
42	0.2	N	0.3	0.4
43	0.2	0.3	0.5	0.2
44	0.2	0.1	0.7	0.3
N: Not Detectable				

Table 3. Spearman correlation coefficients of radon concentrations with various water parameters and with the measured heavy metal concentrations

	Well Depth	pH	EC	Aquif.	Temp.	Copper	Zinc	Arsenic	Lead
r_s value	- 0.37	- 0.47	0.45	0.54	- 0.47	0.08	0.27	0.44	-0.29
p- value	0.015	0.001	0.003	0.000	0.001	0.64	0.08	0.12	0.22

Undetectable values of zinc, arsenic, lead were excluded from correlation analysis.

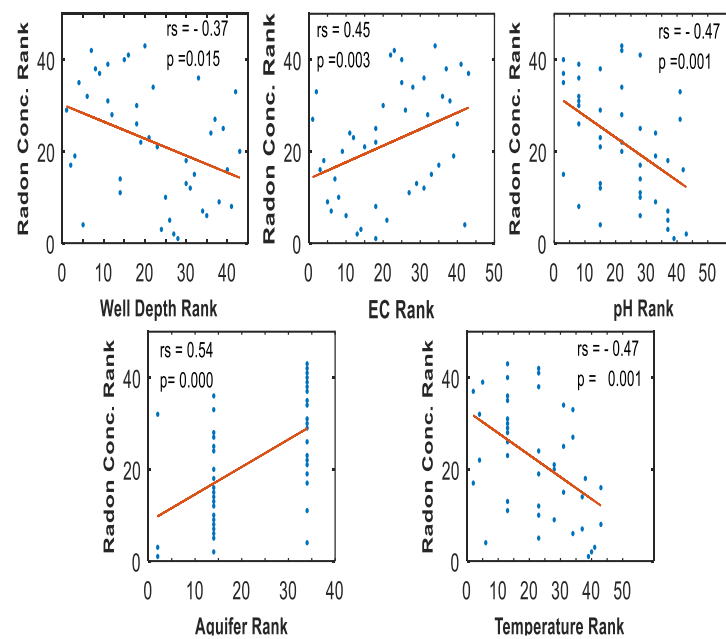


Figure 3. Spearman correlation coefficients of radon concentration with well depth, EC, pH, aquifer type, and temperature.

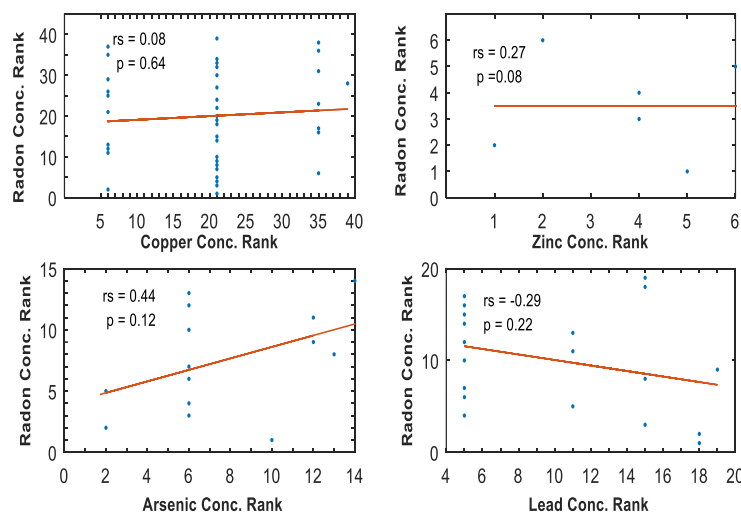


Figure 4. Spearman correlation coefficients of radon concentration with copper, zinc, arsenic, and lead

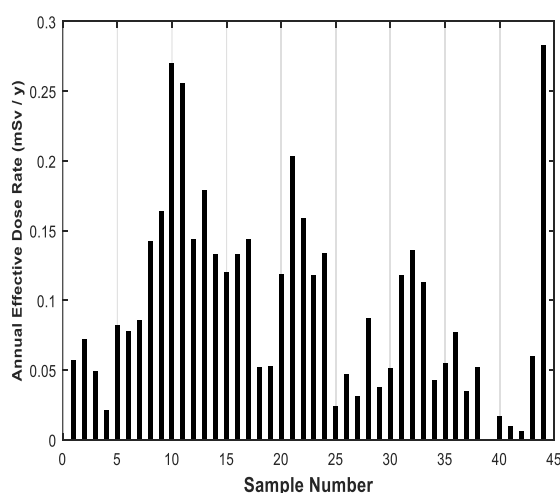


Figure 5. Annual effective dose rates associated with all water samples.

3.4 Health Hazard calculations:

As shown in table 1 and figure 5, the annual effective dose rate (H_E) values ranged from 0.006 to 0.283 mSv/y with a mean of 0.1 mSv/y. The values of H_E due to intake of radon associated with 19 samples were above 0.1mSv/y, the WHO individual dose criteria (IDC) from ingestion of drinking water [15].

All samples from the Eastern well field, six of the fourteen Musayek samples, four of the eleven Western samples and one of the ten Asser samples exceeded the average world value. The annual effective dose rates (H_E) associated with radon intake from the Western, Eastern, Musayek, and Asser well fields ranged from 0.021 to 0.27 mSv/y, 0.119 to 0.203 mSv/y, 0.024 to 0.283 mSv/y, .006 to 0.113 mSv/y respectively with means of 0.116 mSv/y, 0.149 mSv/y, 0.094 mSv/y, and 0.047 mSv respectively.

4. Discussion:

As mentioned in the results section, radon-222 concentrations ranged from 0.82 ± 0.38 to 38.73 ± 2.98 Bq/L with a mean value of 13.53 ± 3.9 Bq/L. All samples had radon activity concentration levels below the WHO's recommended value (100 Bq/L). Twenty two samples out of the forty three samples had activity concentrations higher than 11.1 Bq/ L, EPA³¹ and European Atomic Energy Community (EAEC) s recommended limit value of radon concentration [18]. Asser well field only had one sample higher than EPA's limit value and the rest of the higher concentrations were from the other groundwater well fields.

There are some factors that may influence radon activity levels in groundwater such as the well's depth (1), the aquifer type [16], the heavy metal content in the sample and radium mobility[16]. Extensive water withdraw may increase radium mobility in the aquifer and therefore increase the level of radon. Standard water parameters such as pH, temperature and EC give an indication of the mobility of radionuclides in an aquifer [16]. In our study, analysis shows weak (close to moderate) negative correlation ($r_s = -0.47$, $p = 0.001$) of radon with pH. The cause of this inversely correlation of radon levels with pH can be interpreted as: because pH influences the solubility and mobility of radionuclides in water, as pH decreases, the

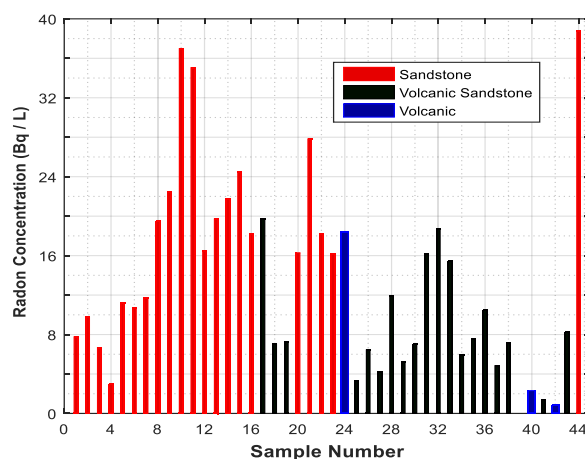


Figure 6. Radon concentration variations with aquifer system of groundwater well.

solubility increases which causes radium (radon parent) to be more soluble and hence increases radon concentration levels in groundwater [17]. Also, a weak (close to moderate) correlation ($r_s = 0.45$, $p = 0.003$) of radon with electric conductivity in samples was observed. Electric conductivity is directly related to the total dissolved solids (TDS) content in a sample where TDS content in water samples may influence radon levels. This explained the correlation between radon levels and EC. On the other hand, a negative weak correlation of radon with temperature was observed. This correlation is unexpected since radon levels is expected to increase with an increase of temperature (positive correlation).

The increase of radon levels with the increase of temperature is due to the increase of radionuclide diffusion rates with the increase of temperature [12]. However in our study, the temperature range is too small to reflect a significant variation in radon levels, and this may be the reason for this unexpected negative correlation. Furthermore, no correlation between radon concentrations and the measured heavy metal concentrations were observed. On the other hand a significant moderate correlation ($r_s = 0.54$, $p = 0.000$) of radon concentration with aquifer types was observed. This indicates that the variation of radon concentrations is related to the type of the aquifer system of the groundwater well.

Some of the groundwater wells studied are within a sandstone aquifer system, a few within a tertiary volcanic aquifer system and some within a volcanic sandstone aquifer (majorly sandstone), see figure 6. The dominated aquifer system of the Western and Eastern well fields is sandstone. On the other hand, Musayek and Asser well fields are dominated by a volcanic sandstone aquifer system.

The radon-222 activity concentrations in the samples from wells with a pure sandstone aquifer system were higher than the concentrations in the samples from wells with a pure volcanic or a volcanic sandstone aquifer system. Samples 9, 10, 11, 13, 21, 22, and 44 which were collected from wells with a sandstone aquifer had highest activity concentrations among all samples. The sandstone aquifer tends to have an elevated activity concentration of natural radionuclides compared to

other aquifer systems [16]. Sandstone can generally be considered as a weathering derivative of quartz-dominated rocks [16]. Rocks with a quartz structure are expected to have high natural radioactivity concentrations.

Health hazard calculations show the annual effective dose rates (HE) ranged from 0.006 to 0.283 mSv/y with a mean of 0.1 mSv/y. Values of HE associated with 19 samples exceeded 0.1 mSv/y, the individual dose criteria (IDC). Thirty-six percent of the Western well field's samples, 100% of the Eastern's, 43% of Musayek's and 9% of Asser's samples were associated with values of HE higher than the individual dose criteria (IDC). Heavy element analyses show copper and zinc in all samples had lower concentrations than the WHO guideline values, while lead had higher concentrations in 19 samples and arsenic in 14 samples.

5. Conclusion:

Our analyses show that there exists an association between radon-222 concentration levels in public groundwater and the well's aquifer type. Samples obtained from the sandstone aquifer system had the highest radon concentrations, followed by those obtained from the volcanic sandstone aquifer system. The samples with the lowest radon concentrations were obtained from a pure volcanic aquifer system. In analyzing the concentrations with respect to geographical location, it was observed that the lowest radon-222 concentration levels in samples were collected from Asser well field. However, this water field showed higher concentration values of heavy metals. The overall average value of the annual effective dose (HE) rates associated with all samples is equal to 0.1mSv/y, the WHO Individual dose criteria (IDC) value. The annual effective dose (HE) associated with the intake of radon from the Eastern well field samples were the highest while for Asser the rates were the lowest. Heavy metal measurements show high concentrations of lead and arsenic in some samples especially in the Asser well field.

Margins:

¹SNIFF mode is the RAD7 device mode allows only Po-218, the daughter of Rn-222, to be counted.

²rs stands for Spearman's rank correlation coefficients statistical analysis

³EPA stands for :US. Environmental Protection Agency

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دراسة تراكيز الرادون - 122 في بعض حقول المياه الجوفية في صنعاء - اليمن

أحمد خالد عبدالرحيم^{1*}, نجوى أحمد المقطري¹

الملخص : تهدف هذه الدراسة ألي تقدير مستويات تركيز الرادون-122 في حقول أبار المياه الجوفية في مدينة صنعاء - اليمن. تم تجميع 43 عينة من منطقة الدراسة واستخدمت طريقة مطيافية ألفا لقياس العينات باستخدام كاشف RAD7. بينت النتائج أن تركيز الرادون في المدى من 0.38 ± 0.82 Bq/L إلي 38.73 ± 2.98 Bq/L. بينت التحليلات وجود ارتباط من الدرجة المتوسطة بين تركيز الرادون وطبيعة التكوين الصخري للحوض المائي وارتباط ضعيف بين تركيز الرادون وكلا من الموصلية الكهربائية والأس الهيدروجيني pH ودرجة الحرارة كما بينت التحليلات وجود ارتباط بين تركيز الرادون وتركيز العناصر الثقيلة المقاسة في عينات الماء. تم تقدير معدل الجرعة الفعالة السنوية وكان معدل الجرعة من 0.006 إلي 0.283 mSv/y كذلك بين التحليل الأيوني للعناصر الثقيلة أن تركيز العناصر الثقيلة : النحاس والخرصين والزرنيخ والرصاص يساوي بالترتيب 0.5 mg/L, 0.7 mg/L, 0.58 mg/L و 0.5 mg/L. الكلمات المفتاحية: رادون 122, تركيز, المياه الجوفية, مطياف ألفا, الجرعة الفعالة, المعادن الثقيلة, ارتباط.