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Hydrological and Radiological Studies of Water Resources by Using Radon in Hashimiya Area- Middle of Iraq.

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Abstract

Radon concentrations are measured for water samples collected from twenty wells which were drilled in Hashimiya area in addition to twelve samples of surface water using Alpha Gaurd. 140 samples, 7 for each well, were collected represent wet season in continuous pumping and 20 samples, one for each well, were collected represent dry season. Concentration of radon in groundwater is many times of its concentration in surface water. The minimum concentration in groundwater is about (7) Bq/L and (5) Bq/L while the maximum concentration is about (31) Bq/L and (19) Bq/L in wet season and dry season respectively. The range of radon concentrations in river water is between (1.06) Bq/L and (1.21) Bq/L. This study has indicated that there is a flow from river water towards shallow groundwater in the closed wells especially in wet period. This is concluded by occurring a dilution of groundwater with that of the river water of low concentration of radon. On the other hand, there is a flow of deep groundwater with elevated concentration of radon to the shallow groundwater which caused to an increase of the concentration of radon in these wells. From the radiological point of view, due to consuming their water, all the wells offer annual effective dose greater than 0.1 mSv which is the recommended value by UNSCEAR. So, these considered as radon contaminated wells and need for remediation before their water is used or consumed.

Keywords: Radon, groundwater, radiological, Iraq.

دراسة هيدروولوجية واشعاعية لمصادر المياه باستخدام الرادون في منطقة الهاشمية-وسط العراق

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الخلاصة

جرى قياس تركيز غاز الرادون في المياه الجوفية التي جمعت من عشرين بئرا تم حفرها في منطقة الهاشمية-وسط العراق بالإضافة إلى اثنتي عشرة عينة من المياه السطحية باستخدام جهاز- الفا جارد. تم جمع 140 عينة من المياه الجوفية 7 عينات من كل بئر تمثل فترة الزيادة المائية في ضخ مستمر، عينة كل نصف ساعة و 20 عينة من المياه الجوفية عينة واحدة من بئر تمثل فترة الجفاف. كانت تراكيز غاز الرادون في المياه الجوفية حوالي عدة اضعاف تركيزه في المياه السطحية. بلغ الحد الأدنى للتركيز في المياه الجوفية حوالي (7) بيكريل / لتر و (5) بيكريل / لتر في حين أن أقصى تركيز حوالي (31) بيكريل / لتر و (19)

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بيكريل / لتر في موسم الزيادة المائية وموسم الجفاف على التوالي. في المقارنة، فإن تركيز غاز الرادون في مياه النهر ما بين (1.06) بيكريل / لتر و (1.21) بيكريل / لتر. وجد في هذه الدراسة ان هناك حركة لمياه النهر باتجاه المياه الجوفية في الآبار الضحلة وخاصة في الفترة الرطبة. واستنتج ذلك نتيجة وجود تخفيف لتركيز الرادون في المياه الجوفية نتيجة اختلاطها بمياه النهر ذات التراكيز المنخفضة من غاز الرادون. من ناحية أخرى، هناك تدفق المياه الجوفية العميقة ذات التراكيز المرتفعة نسبياً من غاز الرادون باتجاه المياه الجوفية الضحلة أدى الى رفع تركيز غاز الرادون في هذه الآبار. من وجهة نظر البيئة الإشعاعية، ونتيجة لاستهلاك المياه من جميع الآبار وجد انها تسبب جرعة اشعاعية فعالة سنوية أكبر من 0.1 ملي سيفرت . لذلك، تعد هذه الآبار ملوثة بالرادون وتحتاج مياهها الى المعالجة قبل استخدامها او استهلاكها من قبل المواطنين في المنطقة.

Introduction

Surface and groundwater resources assessment represent one of the main issues in planning and management water resources in worldwide and it pushes researchers to achieve more and more researches using different tools and techniques [1-4]. Due to the scarcity in fresh water, it is important to investigate all water resources and their interconnection to set adequate plans for the best utilizing of water resources. Hydrogeological traditional techniques, stable isotopes and radioactive isotopes are used as environmental tracers in this field. They have been proved that Radon-222 could be a useful natural tracer; because radon has a large spatial footprint which allows the estimation of aquifer flow system [5] and because of its concentrations in groundwater differ than that in surface water, although the main source of radon in surface water comes from groundwater. Groundwater has relatively high concentration of radon in comparison with surface water. It's half-life, which is comparable to the mean residence time of water and simple instrumentation required for Rn²²² analysis makes it an excellent tracer of groundwater recharge [6-10] and surface water-groundwater interaction [4, 11-13]. For near surface groundwater, which is the focus of this study, the radon concentration is thought to be influenced by infiltrating low radon surface water or by discharging deep groundwater with high radon concentration.

The present study is an attempt to examine the use of radon as indicator in groundwater – surface water interaction and estimate the radiological effect and hazard of using groundwater with relatively high concentration of radon. Obviously, the study is very important, to evaluate the quality of water resources by radiological analyses and estimate the interconnection between surface water and groundwater. Radiological test was implemented on the surface and groundwater samples for radon gas concentrations.

The study area

The study area represents a strategic importance of Hashimiya district, since it is the main source of the food package for the population. It is characterized by a flat surface. And declined gently from north toward the south by 2.8 cm/km. Hilla River is the sole surface water resource in the study area, which enters the area from the western side and leaves it at the south-eastern side (Figure-1). Hilla River flows about 60km from Hindiya barrage on the Euphrates River before it reaches Hashimiya district. Due to the dropping of the water level of the Euphrates River, the water becomes scarcity in study area, subsequently this leads to the increasing of dependence of the population on groundwater by digging wells for the purpose of domestic consumption.

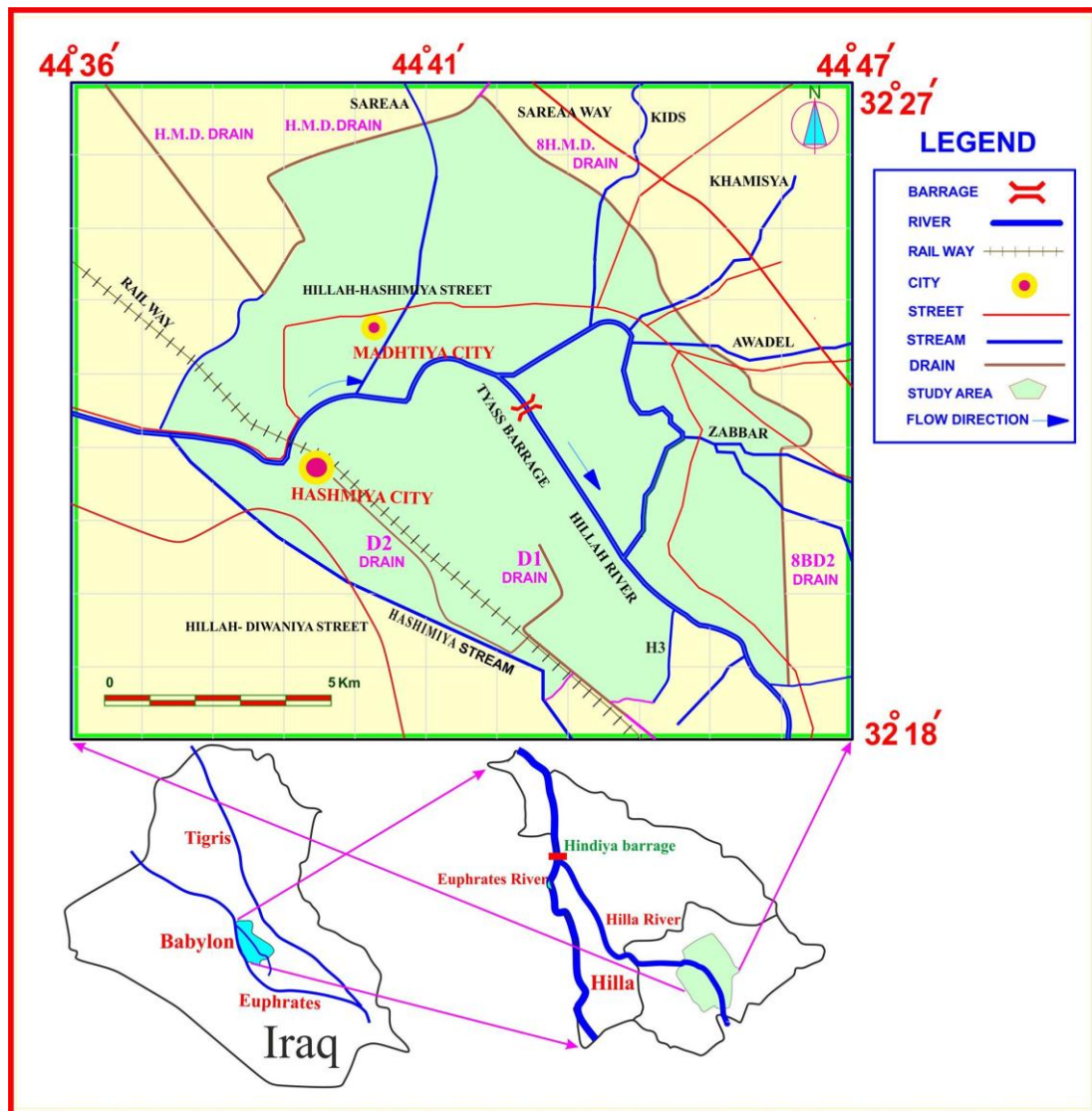
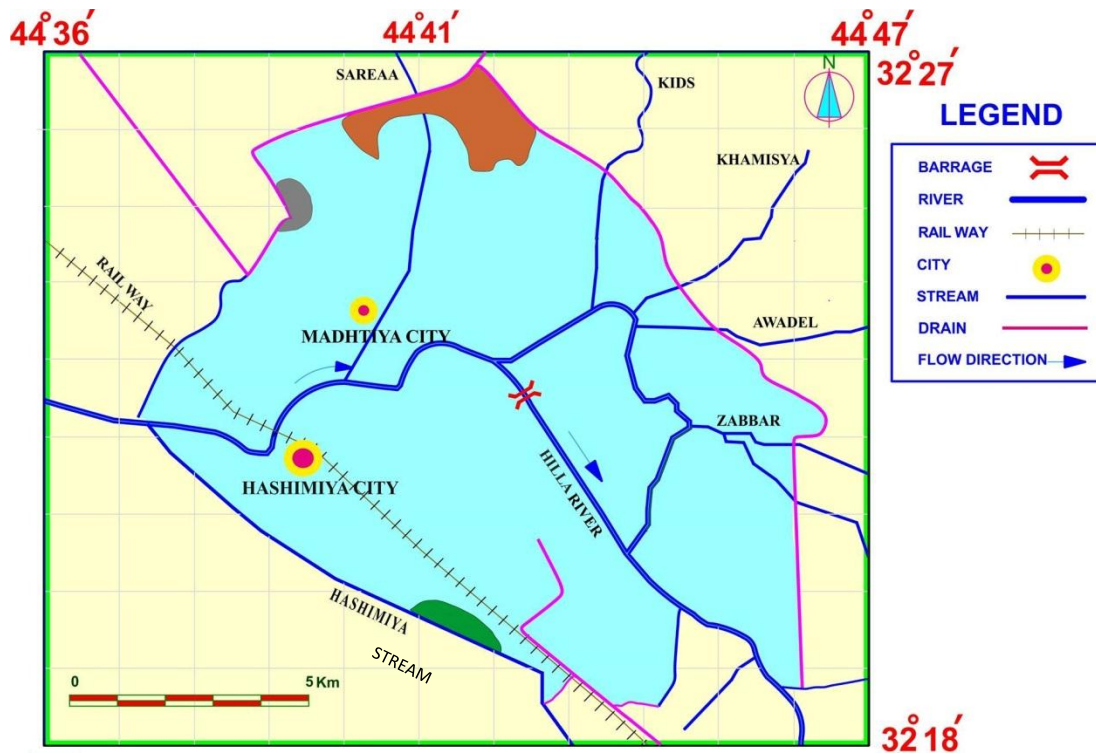


Figure 1- Location of the study area.

Geological setting

Most parts of the study area are covered with different types of Quaternary deposits. These deposits are unconsolidated and usually finer grained than the underlying pebbly sandstone [15]. Quaternary deposits are represented by Flood Plain sediments of the Euphrates River. They are made up of deposits of clay, silt and sand with deposits of gypsum and salt resulting from the evaporation of groundwater, in addition to depression fill sediments, these deposits accumulated as a result of the floods of the Euphrates River, consisting generally of fine sand layers, silt and silt loam [16]. Figure -1 represents the geological map of the area. In general, recent sediments within the area are consisting of a succession of layers of mud, sand and shale with a little amount of gravels in deeper layers [17].



- Shallow depression sediments, clay & silty clay
- Marsh deposits (mud and organic materials)
- Flood plain sediments
- Anthropogenesis sediments/hill (rain) ancient settlement and irrigation canals

Figure 2- Geological map of study area

Hydrogeology

The study area is located, according to the hydrogeological partitions of Iraq, within Mesopotamian, and its groundwater water are within Quaternary deposits. Quaternary deposits represent unconfined aquifer which is consisted of alternative stratification sequence of silt and clay where these sediments have low permeability layers, sometimes up to (20m), holding underneath the layers of sand and gravel often are mixed with silt [17]. Twenty wells are drilled in the study area. The Hydraulic properties of the aquifers that are obtained from the process of the pumping tests serve the understanding and evaluating of the hydrogeological conditions of the area. The nature and movement of groundwater and the amount of storage coefficient are determined (Table-1).

Table-1 Represent locations, depth and elevation of wells and water level during wet and dry periods

No. Well	Coordinate		Well Depth (m)	elevation a.s.l (m)	Bed level (m)	Static level Depth(m) from Ground Surface		Static level Depth a.s.l (m)
	latitude	longitude				Wet	Dry	
1	32°21'38"	44°38'32"	6.45	25.75	19.3	0.35	0.47	25.4
2	32°22'14"	44°40'21"	7	26.40	19.4	1.00	1.30	25.4
3	32°22'37"	44°41'36"	7	26.20	19.2	1.00	1.42	25.2
4	32°20'46"	44°39'44"	7.2	25.00	17.8	1.30	1.64	23.7
5	32°21'06"	44°40'54"	7.3	25.30	18	1.31	1.55	24
6	32°21'30"	44°41'45"	7.2	25.2	18	1.00	1.20	24.2
7	32°21'18"	44°42'42"	7.6	25.2	17.6	1.32	1.60	23.9
8	32°19'42"	44°41'43"	7.8	24.8	17	1.50	1.73	23.3
9	32°20'25"	44°42'26"	7.9	25.2	17.3	1.82	1.88	23.4
10	32°19'20"	44°43'12"	8.2	25.2	17	2.00	2.45	23.2
11	32°19'24"	44°44'54"	8.4	25.4	17	2.01	2.15	23.4
12	32°20'01"	44°45'26"	8.2	25.5	17.3	2.00	2.37	23.5
13	32°20'56"	44°45'30"	7.55	24.95	17.4	1.45	1.68	23.5
14	32°20'58"	44°44'18"	7.4	25.6	18.2	1.20	1.51	24.4
15	32°24'36"	44°42'42"	8.9	26.4	17.5	2.52	2.96	23.9
16	32°23'39"	44°41'50"	8	26.5	18.5	1.75	1.97	24.75
17	32°25'26"	44°41'30"	8.4	26.2	17.8	2.20	2.29	24
18	32°22'42"	44°38'43"	6.4	25.4	19	1.40	1.62	24
19	32°23'37"	44°39'47"	7.1	25.6	18.5	1.35	1.43	24.25
20	32°25'24"	44°40'18"	8.1	26	17.9	2.25	2.45	23.75

Measurements and methodology

Twenty wells are drilled in the area. They are used to monitor the concentration of radon and investigate the hydrogeological conditions. The depth of wells are reached to the brown clay bed within the Quaternary deposits. The distribution of wells and their locations are determined using GPS (Figure-3) (Table-1). Radon concentrations are measured using Alpha Guard according to a procedure that is listed in the manual of the device (Germany).

Continuous pumping for 3 hours duration is carried out and samples are taken from all wells each half an hour in wet period. Twelve surface water samples and 140 samples of groundwater in wet period are collected to be use in the measurement of radon concentrations in continuous pumping, and twenty samples of groundwater are collected, one sample for each well that represented the dry period.

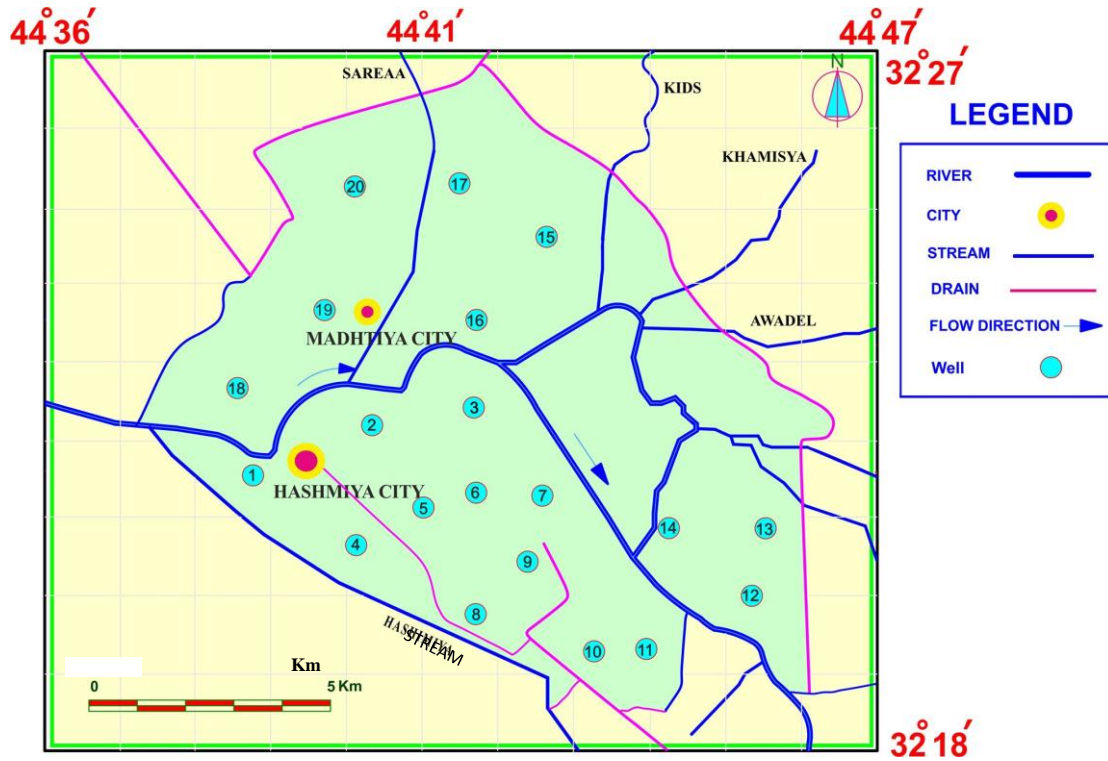


Figure 3- Locations of wells drilled in study area.

These samples are taken and kept in a closed cool-box to prevent releasing of radon into atmosphere in order to determine its concentrations in groundwater. Radon concentration estimating basically depends upon the indicated radon concentration in the monitor. This value is not the required value of radon concentration, since some of the radon is diluted by air within the measurements setup and small part of it remains diluted in a watery phase. To quantify the diluted radon, the interior volume of the measurement set-up (V_{system}) is required. The remaining quantity of radon can be determined by introducing the coefficient k . Eq. (1) is the basic form for radon estimation.

$$C_{water} = \frac{C_{air} * \left(\frac{V_{system} - V_{sample}}{V_{sample}} + k \right) - C_o}{1000} \dots\dots\dots 1$$

Where: C_{water} : Radon concentration in water sample ($Bq L^{-1}$), C_{air} : Radon concentration in the measuring setup after spelling the radon ($Bq m^{-3}$), C_o : Initial concentration in the measuring setup before sampling ($Bq m^{-3}$), V_{system} : Interior volume of the measuring setup (mL), V_{sample} : is the measuring water sample volume (mL), and k : is the radon distribution coefficient.

In order to estimate radon concentration, the measuring set-up parameters are as: $V_{system}=1102$ ml, $V_{sample}=100$ ml, $k=0.26$ and $C_o=0$ $Bq m^{-3}$. Accordingly, Eq. (1) may be abbreviated to:

$$C_{water} = \frac{C_{air} * 10.28}{1000} \dots\dots\dots 2$$

Result and discussion
Concentration of Radon

The radon concentrations found in the groundwater are about many folds over its concentrations in surface water. The minimum concentrations in groundwater are about (7) Bq/L and (5) Bq/L while the maximum concentrations are about (31) Bq/L and (19) Bq/L in wet and dry seasons respectively (Table-2). In comparison, the range of radon concentrations in river water is between (1.06) Bq/L and (1.21) Bq/L (Table-3). Figures-4 and 5 represent the distribution of radon in groundwater and river water in wet and dry seasons respectively.

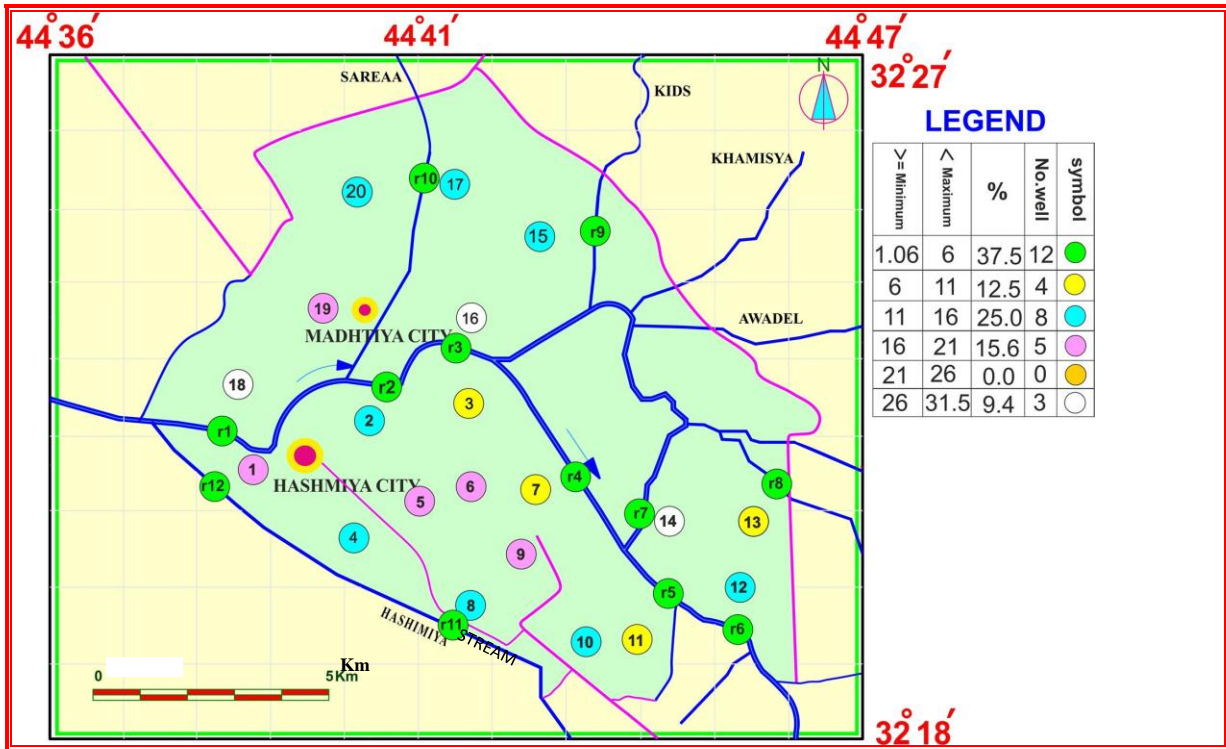


Figure 4- Exhibited colored classes post map overlying base map of study area indicating the distribution of radon (Bq/L) in groundwater and river water in wet season.

Surface water is low in radon concentration. However, by measuring various spots in rivers or streams, areas with high radon concentrations could indicate groundwater seepage points [18-21]. Figures-4 and 5 showed that radon concentrations in wells water in study area are higher than that of radon concentrations in the adjacent sampling points of river water in the two seasons. However, four wells (3, 7, 11 and 14) in wet season have radon concentrations within class (6-11) Bq/L which may be influenced by the dilution of the river water. The concentrations of radon increase as the groundwater moves away from the river.

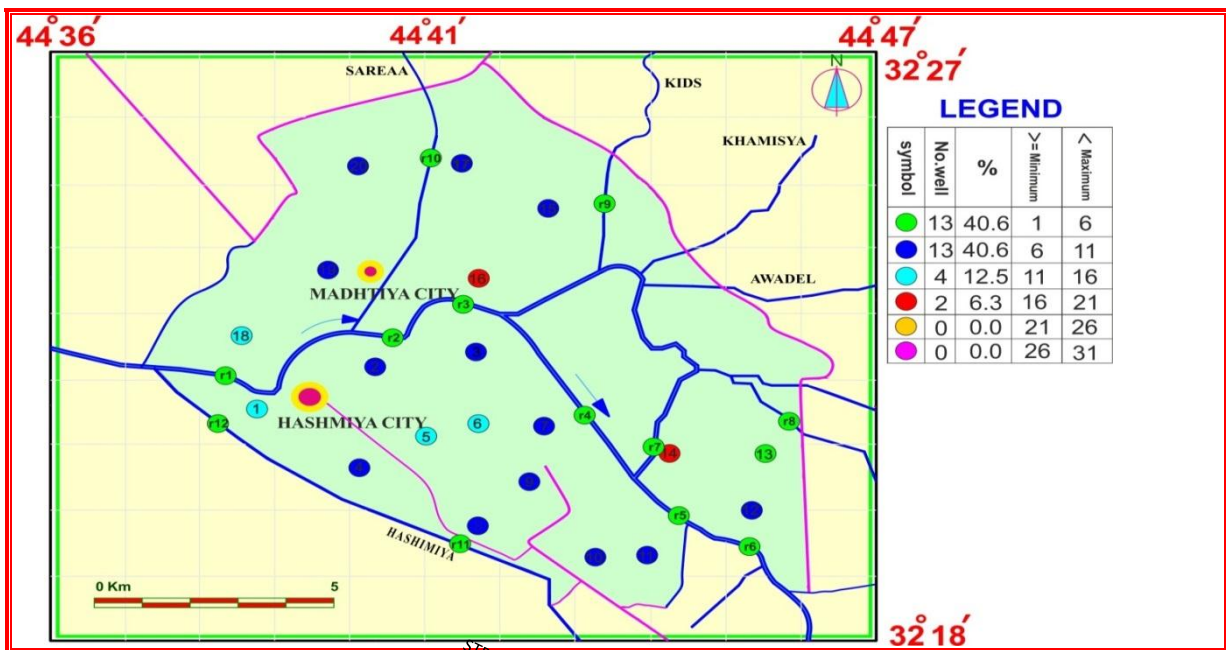


Figure 5- Colored classes post map overlying base map of study area indicating the distribution of radon in well water and river water in dry season.

In dry season, 40.6% of the wells have radon concentrations range between 6 and 11 Bq/L. It appears clearly that there is groundwater flow originated from the river which causes a dilution to the groundwater with river water having low concentrations of radon (Table-3). The same case was noted by Hoehn and von Gunten, (1989) [8]. The non-homogeneity influence of dilution by river water is due to non-uniformity distribution of radium concentration in the sediments comprising the beds of the aquifer. No clear relationship was found between radon in groundwater and radium in sediment of the well (Figure-6). The difference in the lithology of the wells in the area in addition to well's location relative to the river and the direction of water flow are others reasons for the inhomogeneity this dilution.

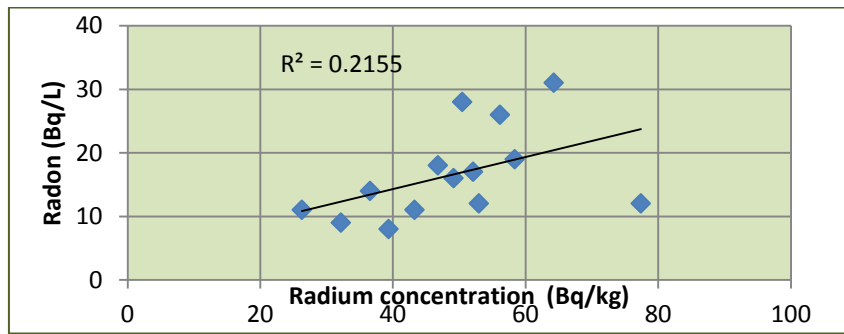


Figure 6- Correlation between radium concentration in well's sediment and radon in the well water

Table 2- Concentration of radon in wet and dry seasons in groundwater and concentration of radon in groundwater with continuous sampling.

Well No.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20
²²²Rn Concentration with time (Bq/ L)																				
Time of measuring (Min)	0	16	11	9	13	19	17	10	15	18	12	8	7	26	11	31	12	28	17	14
	30	32	16	21	35	41	22	27	24	30	15	30	18	34	28	40	17	36	25	27
	60	47	27	35	60	49	31	42	56	47	58	33	40	46	52	19	39	39	44	44
	90	66	34	42	78	61	58	55	71	58	72	54	73	63	70	49	43	46	49	49
	120	95	48	56	96	77	64	72	83	66	89	65	90	71	100	67	84	63	52	52
	150	101	50	61	109	103	69	84	88	79	109	67	99	88	110	92	102	70	65	65
	180	103	52	64	110	105	75	88	95	80	113	68	108	93	113	107	104	76	68	68
²²²Rn Concentration for the two seasons (Bq/ L)																				
Well No.	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20
Wet season	16	11	9	13	19	17	10	15	18	12	8	7	26	11	31	12	28	17	14	
Dry season	12	7	6	9	13	11	7	8	10	8	6	9	5	17	7	19	9	15	10	8

Table 3- Concentration of ^{222}Rn in surface water. (Sample No. as shown in Figure-4)

Sample No.	^{222}R Concentration (Bq/L)
r1	1.20
r2	1.11
r3	1.14
r4	1.17
r5	1.09
r6	1.06
r7	1.10
r8	1.16
r9	1.21
r10	1.13
r11	1.08
r12	1.10

Many researchers have been mentioned that radon concentration in water varies with sampling time due to dilution by recharge [22]. In the current study variation in concentrations of radon is noticed in the two seasons (Figure-3), (Figure-4) and (Table -2). The average value of radon concentrations in wet season is (15.3 ± 6.5) Bq/L while the average value of radon concentrations in dry season is (9.8 ± 3.8) Bq/L. As mentioned above, dilution with low concentrations of radon from river water caused these variations. Climate changes (temperature) may be another reason. Change in the contributing aquifer due to pumping and fluctuation in the water table may lead to the variation.

Continuous Pumping Process Technique

An efficient pump was setup at each well separately and the pumping process is exactly continued to (3hrs) with a constant pumping discharge of (5L/s). Seven samples are taken for each well. The radon concentration is estimated according to Eq.(2) for the 140 samples. Figure-7 shows the distribution of radon in well water after pumping for 180 minutes. It seems obviously that the concentrations of radon in groundwater at the end of the pumping are much higher than that of the river (Table-2).

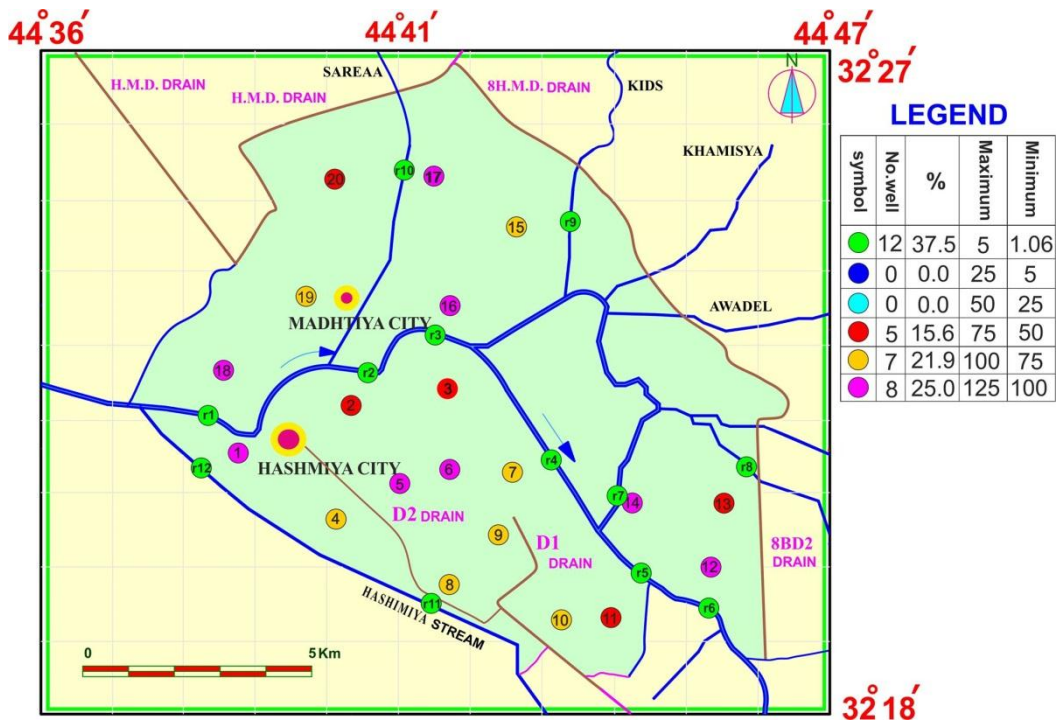


Figure 7- Colored classes post map overlying base map of study area indicating the distribution of radon in well water and river water after 180 minutes pumping.

Increasing of radon in groundwater occurs due to recharging of wells with new deep water which have more concentration of radon. Since this new water is being confined and in touch for longer time with host rocks and sediments. Sloto, (2000) [22] indicated that radon concentration changes with time as a result of dilution by natural recharge. It is also noticed that there is a direct change with depths.

The results of a short period continuous pumping for the twenty scattered wells in the study area are shown in Table-2. Some of these wells (well No.1, 5, 6, 12, 14, 16, 17 and 18) have radon concentration exceeds the allowable limit of radon concentration in drinking water [100 Bq L^{-1}] recommended by WHO, (2008)[23] at the end of the pumping period (3hrs) in groundwater. The radon concentrations with pumping time for the 140 samples are represented graphically in Figures- 8, 9, 10 and 11. In these figures, the concentrations of radon increase rapidly during pumping, in an ‘S’ shape curve. After reaching the maximum, the radon then decreases or be in steady state. The rapid increasing is due to the initial measurements of old stagnant water in the well that have low concentration because it releases its radon to the atmosphere before reaching the fresh deep groundwater. Freyer et al., (1997) [24] and Guisepepe, (2006) [25] achieved studies with similar results. They also found ‘S’ shape curve and they attribute that to the effect of an initial mixing of old water with rich- radon fresh groundwater. Behavior of radon concentration during continuous pumping of polluted groundwater has been noted by Fukui, (1985)[26]; Hightower and Watson (1995) [27] and Freyer et al., (1997) [24] have indicated that it increases continuously and rapidly during purge as "S" curve and radon concentration is depending on recent amounts of pumped groundwater. It is agreed by many researchers that local geology with a specified pumping conditions during sampling reflects temporal variation. All researchers observed that early hours offered lowest radon concentrations and in the later day the concentration increased by about 58%. However, they agreed to use the stability of radon concentration periods as representative values to be evaluated and treated as it was undertaken in this research.

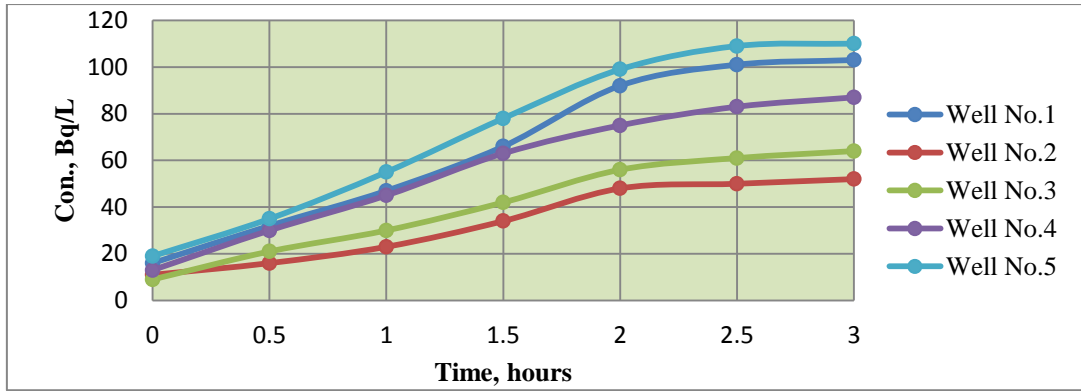


Figure 8- S-curve of radon concentration for wells (1, 2, 3, 4 & 5)

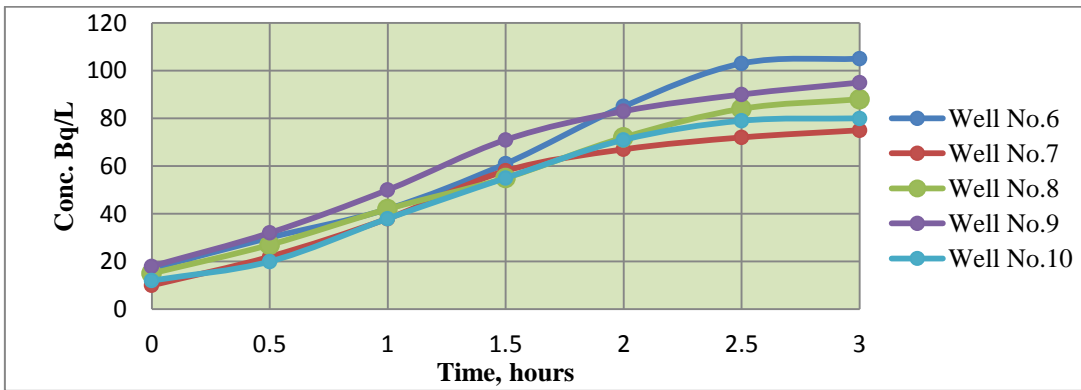


Figure 9- S-curve of radon concentration for wells (No.6, 7, 8, 9 & 10)

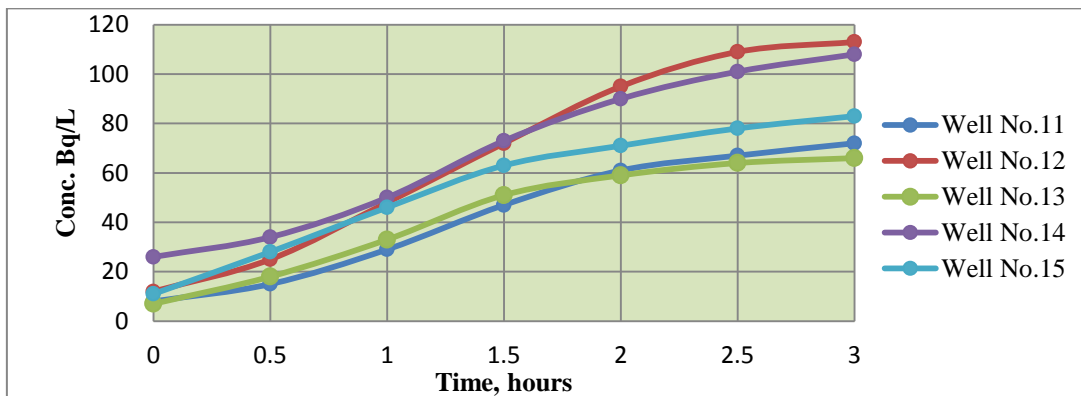


Figure 10- S-curve of radon concentration for wells (No.11, 12, 13, 14, 15)

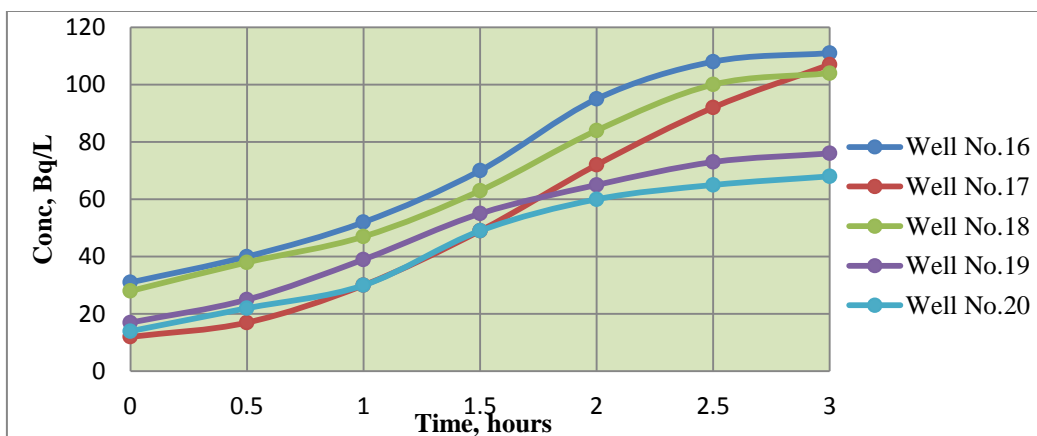


Figure 11- S-Curve of Radon concentration for wells (No.16, 17, 18, 19 & 20)

The physical interpretation for the ascending concentration of radon with time during pumping process is due to releasing process of radon come from new confined groundwater in deep water bearing beds. Figures-8, 9, 10 and 11 revealed that around the vicinity of the pumping wells the initial concentrations are in their minimum values since the polluted water lost most of its radon concentration due to the exposure of the aquifer water to the atmosphere. But as a pumping process is continued, the confined groundwater in deep water bearing stratum and fractures flowing toward the centers of the pumping wells and immediately the tested specimens are taken corresponding to a standard process before groundwater releasing its natural radon concentration into atmosphere. This is interpreted why radon concentrations increase with time to exceed $[100 \text{ Bq L}^{-1}]$. It is expected that radon concentrations are exceeded the presenting values shown in Figures-8, 9, 10 and 11, if the uppermost of pumping discharges increase to (10 or 20L/s).

Spatial Variations of Radon concentration in Hashimiya

A spatial distribution contour map of radon concentrations in aquifer in Hashimiya area is shown in Figure-12 indicated that radon concentrations exceeding the allowable limits of $[100 \text{ BqL}^{-1}]$ occurred in three locations overall the study domain.

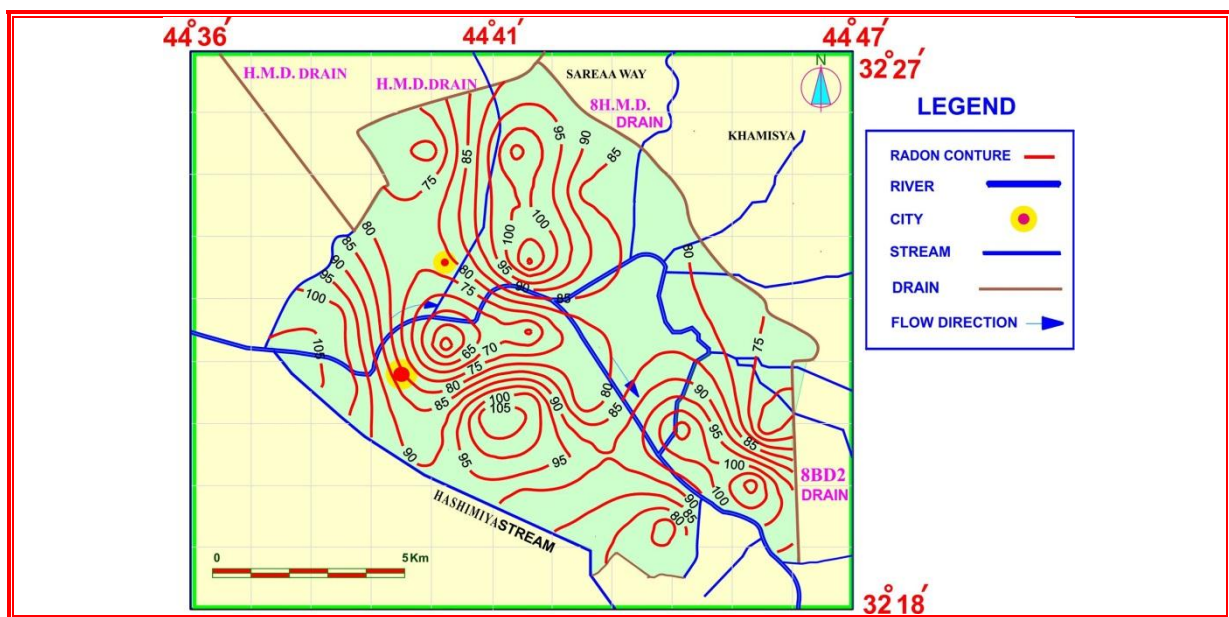


Figure 12- Spatial distribution of radon in Hashimiya aquifer $[\text{Bq L}^{-1}]$ Radiological aspect

The annual effective dose D_w (Svy^{-1}) to the individual consumer due to intake of radon from drinking water is calculated by using the following formula [28-29]

$$D_w = C_w CR_w D_{cw}$$

Where C_w is concentration of radon (Bq/L) in water, CR_w is the annual intake of water (730 liters [23] and D_{cw} is the dose conversion factor $= (3.5 \times 10^{-9} \text{ Sv/Bq})$ [30]. The average measured concentration of radon in groundwater in continuous pumping for 3 hours is used in estimation of the annual effective dose.

The result of annual effective dose rate is found to be ranged between (0.1- 0.21) mSv. Consequently, all the wells offered annual effective dose greater than 0.1 mSv the recommended value by UNSCEAR[30]. So they are considered as radon contaminated wells and need for remediation before their water is used or consumed. Moreover, if the recommendation suggested by WHO is considered, some of the tested wells (No. 1, 5, 6, 12, 14, 16, 17 and 18) will have values exceed the 100 Bq/L. The average global dose for ingestion of radon in drinking water is 0.002 mSv/y. About 90% of the dose of radon in drinking water caused from inhalation of radon released from drinking water (0.025 mSv/y) [14, 23, 30]. According to these values the annual effective dose by inhalation of radon released from uses of groundwater in study area might exceed 1.1 mSv/y (the global average dose caused from inhalation of radon and its decay products from all sources [14].

Thus, the population of Al-Hashimiya city at some locations vulnerable to the health risk because of the higher concentrations of radon in the groundwater if it is used it as drinking water or even for domestic uses.

Conclusion

The current study indicated that radon is a good environmental tracer to figure the interconnection between surface water and shallow groundwater and also indicated the recharge of deep groundwater into shallow groundwater. The water flow from river water to shallow groundwater in closed wells especially in wet period diluted groundwater by river water of low concentration of radon. On the other hand, there is a flow of deep groundwater with high concentration of radon to the shallow groundwater caused the increases of the concentration of radon in these waters. Water sampling from continuous pumping have radon concentration in groundwater exceed the level of 100 (Bq/L), the value recommended by the WHO [23] for drinking water. The annual effective dose caused by consuming this water as drinking water or even for domestic uses will be greater than the global average dose caused by ingestion or by inhalation or radon releases from this water. So remedy of water with high radon concentration must be done before consuming of the groundwater in the study area.

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