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Advances in Applied Science Research, 2011, 2 (6):336-346

ISSN: 0976-8610
CODEN (USA): AASRFC

The Determination of Radon Exhalation Rate from Water using Active and Passive Techniques

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ABSTRACT

The present article represents two sets of experimental works in the measurements of Radon concentrations in surface, underground water and oil-produced water separated from oil in Basra Governorate in Iraq. The first measurement in by using fast electronic techniques called RAD7 and the second is the passive method by using the solid state nuclear track detectors CR39 and L115-II. Tap water in the governorate, has a very low radon concentration 174Bq/m^3 at Al-Fao port, private wells water did show a radon level as high as 2050Bq/m^3 at Lehais south, while the oil- production waters have radon level ranging between 8464Bq/m^3 and 50926Bq/m^3 .A conclusion has been made, that Basra governorate tap water is safe as far as radon concentration, while oil-production waters should be avoided. Samples of water were collected during March 2011, from all locations in the governorate .From each location three samples were collected at different day time.

Keywords: Radon, RAD7, SSNTD, Drinking water, Underground water, Oil- Production water.

INTRODUCTION

The main concern of environmental monitoring is the measurement of the natural radiation arising from naturally radioactive materials and their progenies. Researches carried out in recent decades show that; under normal conditions, more than 70% of a total annual radioactive dose received by people originates from natural sources of ionizing radiation, whereby 54% is due to the inhalation and the ingestion of natural radioactive gas radon ^{222}Rn and its decay products. Exposure to radon via inhalation in closed rooms is the cause of about 10% of all deaths from lung cancer [1, 2]. Radon is a natural inert radioactive tasteless and odorless gas, whose density is 7.5 times higher than that of air. It dissolves in water and can readily diffuse with gases and water vapor, thus building up significant concentrations. Radon (^{222}Rn), which is one of the daughters of uranium (^{238}U), represents the most important radon isotope, with physical half-life

3.825 days. Decays of the radon nucleus (^{222}Rn) yield short-living daughters; Polonium (^{218}Po), lead ^{214}Pb and Bismuth (^{214}Bi). Thoron, (^{220}Rn), half-life 55.6s is a radioactive noble gas that exists in natural radon gas as well. These isotopes are found in soil-near air , surface water , groundwater and oil-production water. But, due to their presence in different decay chains (^{232}Th and ^{238}U -series, respectively), the geophysical and geochemical behavior (e.g. species, number of precursors, half-life, solubility etc.) of their precursors differs, too. While radon has been a subject to research for a long time, see for example references [3-8], the emanation of thoron from solid material containing the precursor ^{224}Ra and its occurrence in aquatic systems is not well known. Radon is particularly well suited to study groundwater and surface water and their interaction, because of activity in groundwater 1 to100 kBq m^{-3}; depending on the lithology of the area, which is much higher than the surface water 1 to 100 Bq m^{-3} [9]. The sources of Radon in water are; either from radioactive decay of dissolved Raduim in water, or, from a direct release of Radon from minerals containing member of Uranuim and Thoruim decay series. Groundwater that deposit from oil can have unusually high constituents of a dissolved radioactive contribution that build up during the period of water with rack contact for a long time period, or water injected in the well in order to maintain pressure in the well during oil production process[10]. Most of the oil-production waters are particularly rich of soluble radioactive ^{226}Ra and ^{224}Ra isotopes, which brought to the earth surface with oil.

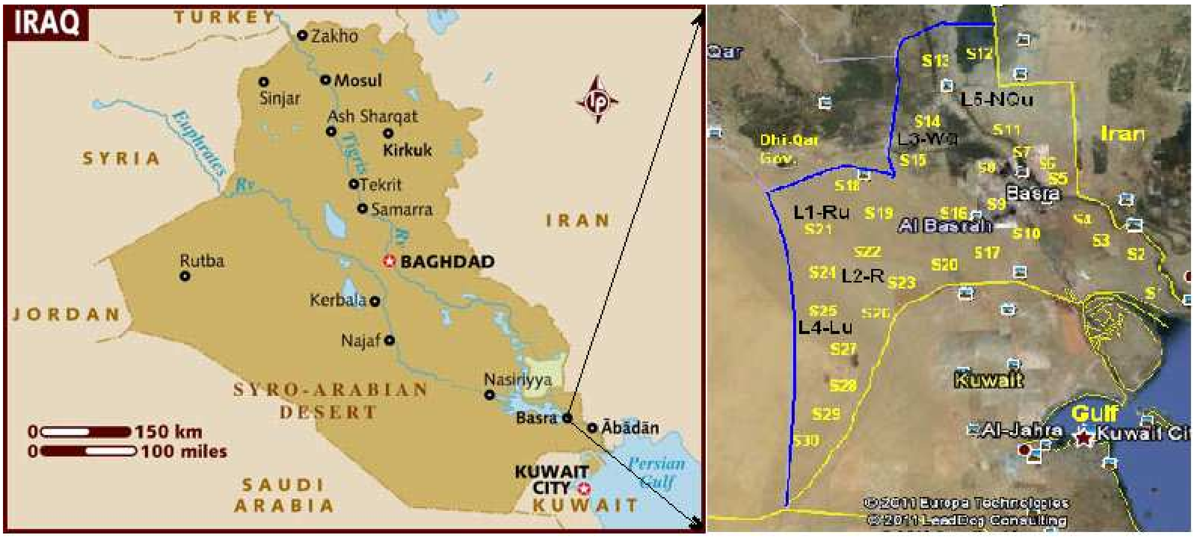


Figure-1. Basra Governorate, dots represent the places where samples taken from, numbering in station number (S) and Location number (L), (Basra map is from Google earth)

This work describes the preliminary findings from Radon-222 activity concentration measurement data collected in. The general aim is to investigate the complex interactions and exchanges between stream flow of water and groundwater, at scales that are imperceptible to standard hydrological and hydraulic analyses, and to estimate how much hazards brought with the oil-production waters. In fact, the study area is located inside Basra Governorate which is located in the extreme south-eastern part of Iraq, see Figure-1. Al-Basra Governorate sited at the northeastern rim of the Gulf, part of the Iraqi Southern Desert in the west and south and relatively short coast on the Gulf. In the northern part of Basra Governorate, Tigris and

Euphrates merge forming Shatt-Al Arab river which flows southward to the Gulf. The total area of Basra is 19070 km^2 and the population exceeds 3 million.

In Basra Governorate, most of the drinking water is obtained from surface water (rivers). These sources of water normally have Radon concentration less than the water obtained from groundwater sources such as private wells

MATERIALS AND METHODS

2. Experimental work

In Basra governorate, the household water is supplied from two sources; one from Bada'a (on Euphrates River) and the other from Shatt-alrab river (formed by the confluence of the Euphrates and the Tigris rivers). Samples from 73 stations and locations were collected during March 2011. The collected, 0.25L, bottles completely filled with water and well sealed to avoid any connection with air. Each water sample was sealed off for 3-4hrs and allowed for ^{222}Rn progeny to reach equilibrium with ^{222}Rn. The measurements of radon concentration water were carried out by two methods:

2.1 Active method

The calibrated alpha spectrometer DURRIDGE *RAD 7 USA*, with special accessories for radon measurement in water was used for measurements of Radon concentration in drinking water and ground water. The detector converts alpha radiation directly to an electric signal and has the possibility of determining electronically the energy of each particle, which allows the identification of the isotopes (^{218}Po, ^{214}Po) produced by radiation, so it is possible to instantaneously distinguish between old and new radon, radon from Thoron, and signal from noise.

Figure 2 shows RAD7 and schematic diagram of RADH2O. The radon detector was used for measuring radon in water by connecting it with a bubbling kit which enables to degas radon from a water sample into the air in a closed loop. A sample of water was taken in a radon-tight reagent bottle of 250ml capacity. This bottle was connected in a close circuit with a zinc Sulphide coated detection chamber which acts as Scintillatore to detect alpha activity and a glass bulb containing calcium chloride to absorb the moisture. Air was then circulated in a closed circuit for a period of 5-10 min until the radon was uniformly mixed with the air and the resulting alpha activity was recorded and it directly gives the radon concentration. RAD7 is used in the measurements of radon concentration in the surface and ground water, but not for oil-production water, due to oil gauming with water ,which has hard effect on the detector desiccants.

2.2. Passive Method

We used the Solid State Nuclear Track Detectors (SSNTD), for the measurements of Radon concentration in drinking water, ground water and oil-production water. The water samples collected from the locations sited in Figur-1. A 15ml water samples were poured into 10x4 cm cylindrical containers (Fig-2). Two SSNTD, CR39 and LR115-II 1.5x1.5cm films, were glued to the bottom of container's cover. The containers then covered, sealed and left for 90 days to irradiate the films with alpha particles emitted from radon.

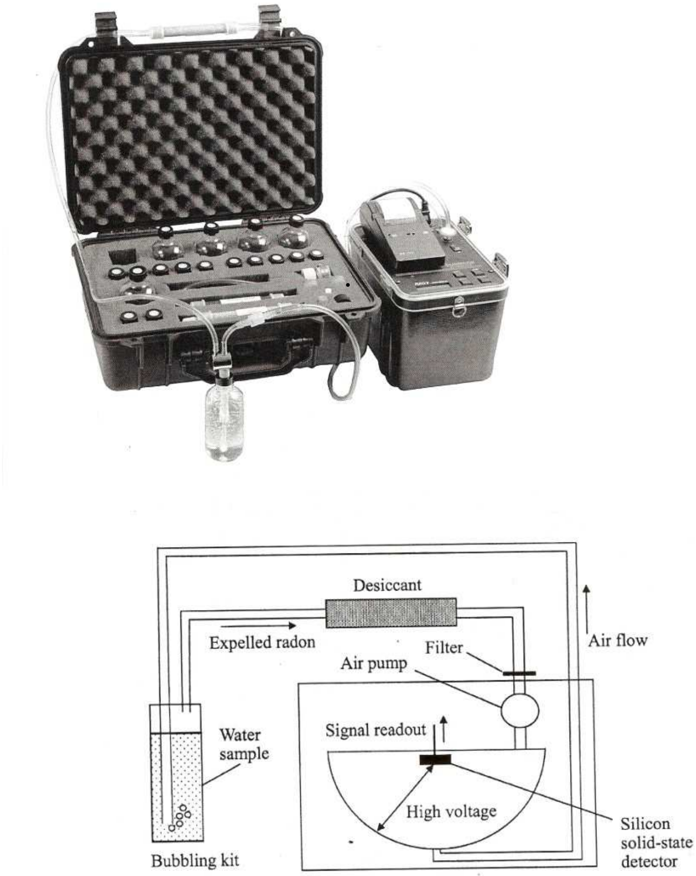


Figure-2a. The RAD7 connected with RAD7H2O accessories.

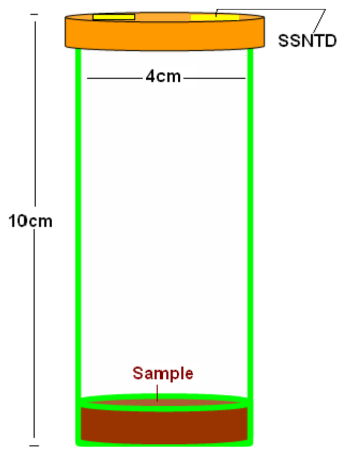


Figure-2b. The cylindrical plastic containers used to measure Radon concentration in water samples [11]. The amount of water samples is 15ml.

After 90 days of irradiation the detectors removed carefully from the cylinder and etched with NaOH solution with conditions 2.5N at 70 ^{o}C for 7 hours for CR39 film and 2.5N at 60 ^{o}C for 2 hours in the case of LR115-II film . The detectors then washed many times by distilled water and dried with tissue papers. The numbers of tracks due to alpha particles interaction are counted by the means of optical microscope 400X.

2.2.1. Theoretical aspect

The alpha particle densities that registered on CR39 and LR115 written as;

$$\rho_G = \frac{Trak\ number}{Global\ view\ area\,(cm^2)\,x\,Global\ erradiation\ time\,(\sec)} \quad (Tr/cm^2.Sec) \quad (1)$$

This density can be given by [11-12]

$$\rho_G^{CR} = A_c^{222}(Bq.cm^{-3})\left[\sum_{i=1}^{3} k_i\, P_i^{CR}\, R_i + \frac{A_c^{220}}{A_c^{222}}\sum_{i=1}^{4} k_i\, P_i^{CR}\, R_i\right] \quad (2)$$

where A^{222} and A^{220} are radon and thoron concentration, P_i^{CR} the probability of alpha-particle of energy E_α (tabulated), R_i is the range of alpha particle in the detector material, and k_i is the branching ratio for alpha decay of radon (tabulated)

$$\rho_G^{LR} = A_c^{222}(Bq.cm^{-3})\left[3P^{LR}\,\Delta R + 4P^{LR}\,\Delta R\,\frac{A_c^{220}}{A_c^{222}}\right] \quad (3)$$

Dividing equation (2) on (3) we get

$$\frac{\rho_G^{CR}}{\rho_G^{LR}} = \frac{\sum_{i=1}^{3} k_i\, P_i^{CR}\, R_i + \frac{A_c^{220}}{A_c^{222}}\sum_{i=1}^{4} k_i\, P_i^{CR}\, R_i}{3P^{LR}\,\Delta R + 4P^{LR}\,\frac{A_c^{220}}{A_c^{222}}} \quad (4)$$

Measuring ρ_G^{CR} , ρ_G^{LR} and making use P_i^{CR}, P_i^{LR} values, one can calculate the ^{220}A/^{222}A ratio from the following;

$$\frac{A_c^{220}}{A_c^{222}} = \frac{\sum_{i=1}^{3} k_i\, P_i^{CR}\, R_i - 3P^{LR}\Delta R\,\frac{\rho_G^{CR}}{\rho_G^{LR}}}{4P^{LR}\Delta R\,\frac{\rho_G^{CR}}{\rho_G^{LR}} - \sum_{i=1}^{4} k_i P_i^{CR} R_i} \quad (5)$$

Using relations (5), (2) and (3) one can evaluate the activity of Radon ^{222}A and thoron ^{220}A

RESULTS AND DISCUSSION

Short term measurements using the RAD7 accessories for drinking and washing surface water and a long term measurement using the SSNTDs were done for 12 samples, S1 to S12, in different locations in Basra. The results are shown in table-1. Eighteen vials of water, S13 to S30, were collected from private well drilled in private farms and used for drinking and plantations in the south of Basra near the border of Kuwait (Zubair), were tested by the two methods as well. The results are presented in Table-1.

By looking at the details of the results, for both kinds of tests, one can recognize that there are differences in the results between the RAD7 and SSNTDs. However, a correlation between the active and passive detectors has been made and shown in figure-3. These differences are due to long and short time measurements. For SSNTDs the measurements were for 90days, while RAD7 measurements were for 1h.

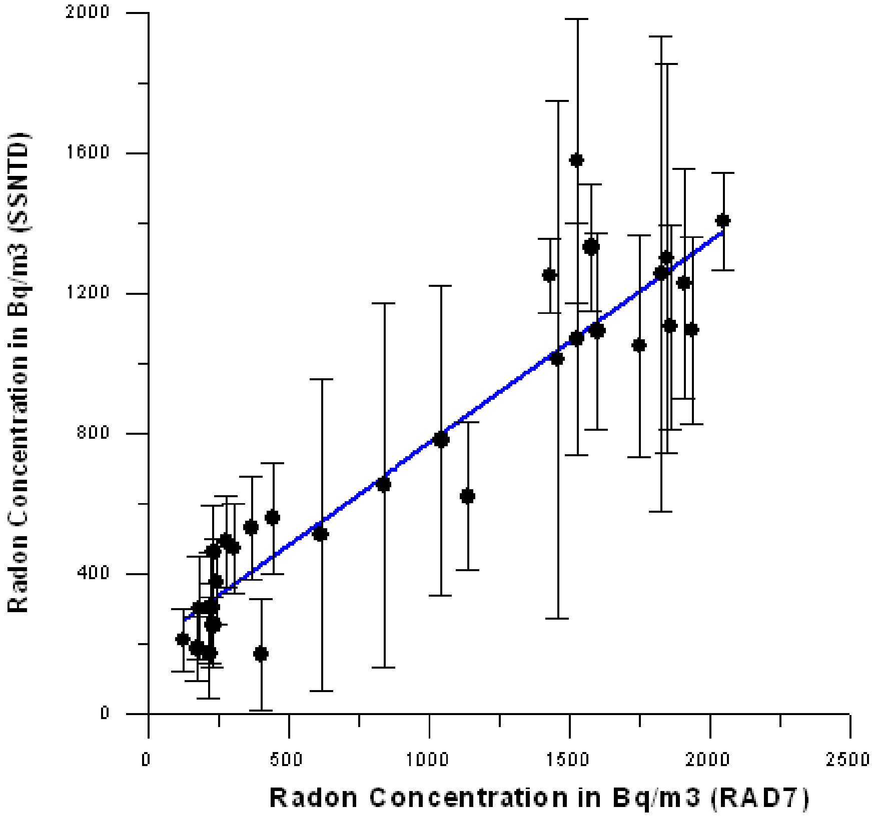


Figure-3a. The linearity between the results of two detectors; active (RAD7) and passive (SSNTDs), in the measurements of radioactive Radon gas annihilates from water.

The radon concentration in the drinking water and groundwater of Basra governorate, in places shown in Figure-1, shows that the average of radon concentration is relativity low, where the source of those samples is surface water. It is found that the Radon concentration is very small when compared with EPA recommended levels (500 Bq/m^3) drinking water and WHO Guidelines for drinking water quality [13]. The arithmetic mean value found to be 308Bq m^{-3}, which is an expected result since the water moves more than 1000km from Turkey to reach Basra (Iraq). In the case of underground water the average is 1151Bq/m^3 and the maximum radon concentration is in the private well near the border of Kuwait 1728 Bq m^{-3}(this was wars zone during 1991 and 2003). These wells are closely associated with homes and were sometimes used as drinking water supplies as well as for irrigation.

Table-1. Measurement) results for water samples (rivers, tap, and groundwater in Basra Governorate

Site Number	Site Name	Radon concentration in Bq/m^3 using RAD7)	Radon concentration in Bq/m^3 using(SSNTD)
S1	Al-Fao	174(90)	185(13)
S2	Abu Floos	216(130)	173(12)
S3	Abu Alkhsieb river	223 (161)	302(21)
S4	Mahalla	230(130)	462(32)
S5	Al- Khora	239(120)	377(26)
S6	Ashaar river	230(120)	252(18)
S7	Alrubat river	274(130)	492(34)
S8	Makal river	302(130)	471(33)
S9	Jubaila river	365(150)	530(37)
S10	Dakier	442(160)	558(39)
S11	Shut Al-basra	400(160)	169(12)
S12	Qurna river	122(90)	212(15)
S13	Tigers	1140(211)	620(43)
S14	Lehais north	1530(329)	1070(75)
S15	Lehais two	1044(444)	781(55)
S16	Lehais one	177(148)	301(21)
S17	Lehais three	614(444)	511(36)
S18	Lehais four	838(518)	652(46)
S19	Lahais six	1460(740)	1011(71)
S20	Lehais nine	1830(678)	1256(88)
S21	Lehais south	2050(140)	1405(98)
S22	Rafithia	1940(266)	1094(77)
S23	Safwan-1	1910(329)	1230(87)
S24	Safwan-2	1860(292)	1105(77)
S25	Safwan-3	1850(555)	1299(91)
S26	Alnajmi	1750(318)	1050(74)
S27	Alnajmi-1	1430(107)	1251(88)
S28	Alnajmi-2	1530(407)	1580(110)
S29	Alnajmi-3	1600(281)	1092(76)
S30	Alnajmi-4	1580(181)	1332(93)

For the measurement of radon concentration level in oil-production water, table-2 and figure-3, reflect the fact that, there was some high level of radon concentration in this water higher than the most of public tap and ground waters in the governorate. The results for these 43 samples categorized into five locations, from L1 to L5, shown in Figure-1. The data ranging from 8468Bq/m^3 in S16at L2-R-51 to 50926 Bq/m^3 at L1-Rumaila-310 sample No. 10. Samples optical images of the tracks recorder on CR39 on the right and LR115-II on the left are shown in figure-4.

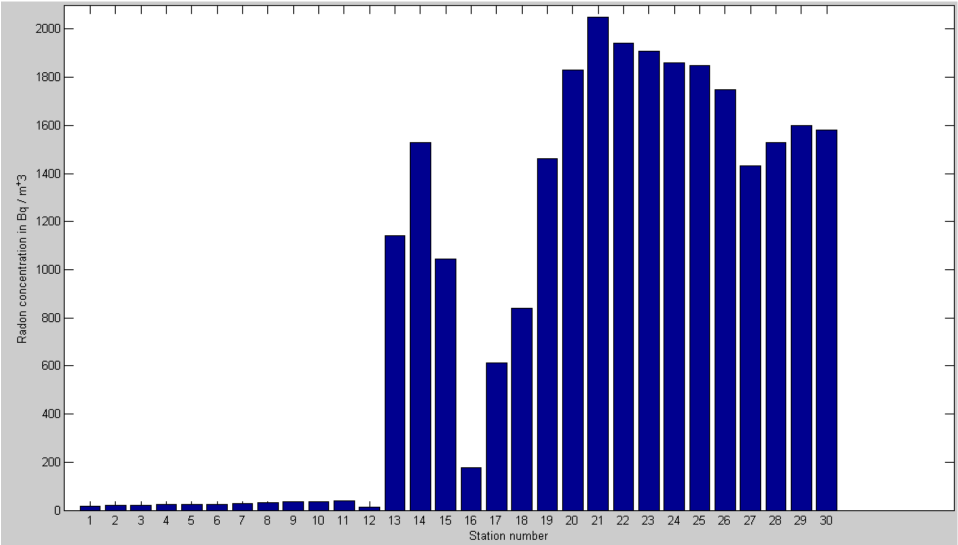


Figure-3b. Radon concentrations in drinking water and groundwater at Basra Governorate

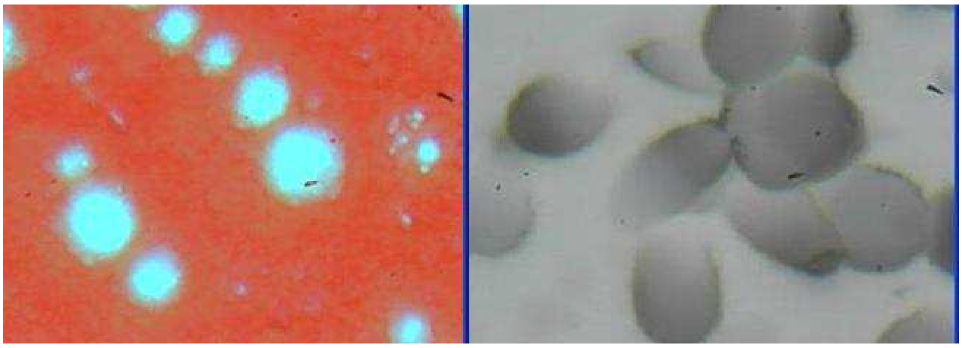
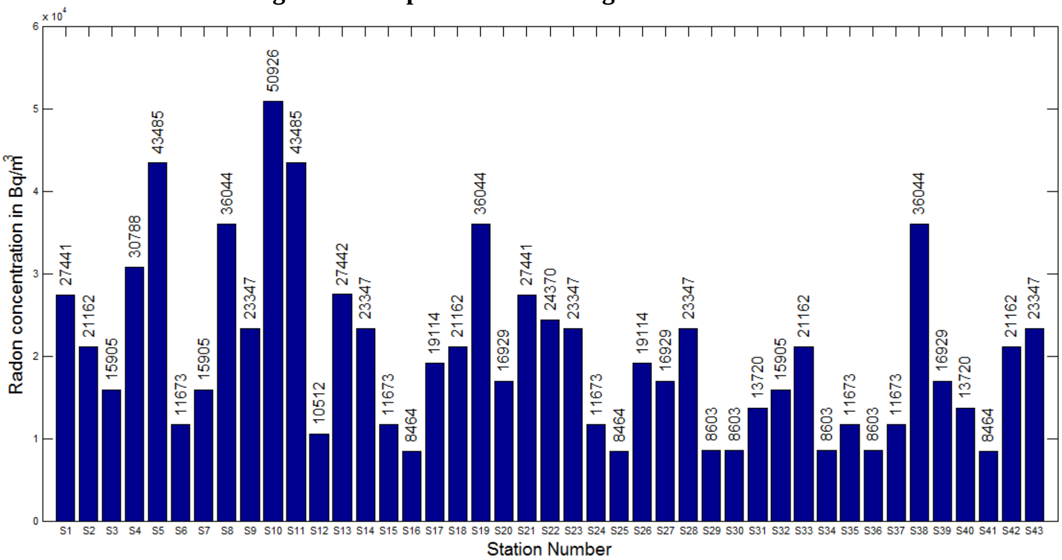


Figure-4. Samples of tracks images on the SSNTDs



Figur-5. Radon concentration for the oil-production water measured by SSNTDs

Table-2.The Radon Concentration in the oil- production water measured by SSNTDs

Sample. No	Location Name	$\rho_G^{CR} x10^{-3}$ $Tr/cm^2.Sec$	$\rho_G^{LR} x10^{-3}$ $Tr/cm^2.Sec$	$^{220}A_c/^{222}A_c$	$^{222}A_c$ in Bq/m^3
S1	L1-Ru-7	4.54241	2.85523	1.52682	27441*
S2	L1-Ru-64	2.59566	1.62229	0.92695	21162
S3	L1- Ru-207	1.36272	0.43591	0.417017	15905
S4	L1-Ru-252	2.01164	1.23294	0.12600	30788
S5	L1-Ru-270	3.56904	2.20631	0.35986	43485
S6	L1-Ru-158	0.84359	0.51913	0.22280	11673
S7	L1-Ru-138	1.36272	0.43591	0.41707	15905
S8	L1-Ru-204	3.24458	2.01164	0.47085	36044
S9	L1-Ru-269	1.68718	1.03826	0.22280	23347
S10	L1-Ru-304	3.89349	2.4009	0.28132	50926
S11	L1-Ru-314	3.56904	2.20631	0.35986	43485
S12	L1-Ru-35	2.46588	1.55739	2.49083	10512
S13	L1-Ru-76	4.54241	2.85521	1.52673	27442
S14	L1-Ru-313	1.68718	1.03826	0.22280	23347
S15	L2-R-11	0.84359	0.51913	0.22280	11673
S16	L2-R-51	1.03826	6.48916	0.92695	8464
S17	L2-R-86	1.16804	7.13808	0.06889	19114
S18	L2-R-177	2.59566	1.62229	0.92695	21162
S19	L2-R-194	3.24458	2.01164	0.47085	36044
S20	L2-R-231	2.07653	1.29783	0.92566	16929
S21	L2-R-238	4.52415	2.85523	1.55260	27441
S22	L2-R-447	2.40099	1.49250	0.58966	24370
S23	L2-R-502	1.68718	1.03826	0.22280	23347
S24	L3-WQ-58	0.84359	0.51913	0.22280	11673
S25	L3-WQ-66	1.03826	6.48916	0.92695	8464
S26	L3-WQ-83	1.16804	7.13808	0.06889	19114
S27	L3-WQ-152	2.07653	1.29783	0.92566	16929
S28	L3-WQ-184	1.68718	1.03826	0.22280	23347
S29	L3-WQ-185	2.92012	1.81696	0.63951	8603
S30	L3-WQ-209	2.92012	1.81696	0.63951	8603
S31	L3-WQ-212	2.27120	1.42761	1.52602	13720
S32	L3-WQ-224	1.36272	0.43591	0.41707	15905
S33	L3-WQ-243	2.59566	1.62229	0.92695	21162
S34	L4-Lu-3	2.92012	1.81696	0.63951	8603
S35	L4-Lu-8	0.84359	0.51913	0.22280	11673
S36	L4-Lu-12	2.92012	1.81696	0.63951	8603
S37	L4-Lu-13	0.84359	0.51913	0.22280	11673
S38	L4-Lu-14	3.24458	2.01164	0.47085	36044
S39	L4-Lu-20	2.07653	1.29783	0.92566	16929
S40	L4-Lu-22	2.27120	1.42761	1.52602	13720
S41	L5-NQ-9	1.03826	6.48916	0.92695	8464
S42	L5-NQ-11	2.59566	1.62229	0.92695	21162
S43	L5-NQ-14	1.68718	1.03826	0.22280	23347

**The error in the results for all radon concentration is 7%.*

All the results summarized in figure-3 as well. In the lack of radon map for Iraq, one may compare these data with the surrounding area. The work of Misconi and Navi summarized the average radon concentration in different Middle East countries [14]; in Iran (Hamadan) 364 Bq m^{-3}, Syria (Damascus 45 Bq m^{-3} , Jordan (north) 144 Bq m^{-3}, Israil (national) 47 Bq m^{-3}, Saudia Arabia (Al- Jauf) 30 Bq m^{-3} and Yemen (Hodeidah) 42 Bq m^{-3}. However those are mainly for doweling, which is affected by the water used.

CONCLUSION

The Radon concentrations in public drinking water were analyzed for 73 samples in Basra governorate (southern Iraq) covered most of the residential area plus the oil fields locations. The concentration for 12 points, where surface water is used, range from 180 Bq m^{-3} in an average to 500 Bq m^{-3} with an average 308Bq m^{-3}, which is in the low concentration range. The concentration in the 18 points (groundwater), ranging from 239 Bq m^{-3} to 1728 Bq m^{-3} with an average 1452 Bq m^{-3}, these concentrations could be categorized into medium level of Radon concentration. Most of the high level concentrations are located near the border of Kuwait and this may related to the penetration of the deplete uranium used in the Gulf wars. Our presented data for radon concentration in oil produced water (43 samples) clearly shows that level of radioactivity exceed limits set by US EPA in average. There will be long term exposure risk to the health of community, if this water recycled back to the river or mixed with underground water. The radioactivity of produced water is mainly from ^{226}Ra and its decay product (^{222}Rn). It is worth underlying health hazard on the workers in the oil fields, because of high radon emanation rate from produced water, which has maximum effective dose 1.629mSv/h, if they close enough to radon gas continuously.

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