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Evaluation of the Radioactivity of Radon Gas and Measuring Radium Content and Uranium Concentrations in Soil Samples of Oil Fields in Kirkuk City - Northern Iraq

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Abstract- Used Nuclear track detectors (CR-39 detector) to measured radon concentration and activity in soil sample, Radium content and Uranium concentrations in soil sample in depth (15-30-45-60) cm for four oil fields (Jumbor (15), Khabaz (5), Bai Hassan (96), Bai Hassan (3), Avanah (93)) in Kirkuk city-northern Iraq. The mean of radon concentration in soil sample is (3572.612 Bq. m^{-3}) with range (252.52-8585.58 Bq. m^{-3}) for study area. Mean and range of the activity of radon for the sample in irradiation tube is (0.704 Bq) and (0.321-1.283 Bq) respectively for the soil in study area .The mean of Radium concentration is (4.95) $Bq.Kg^{-1}$ with range (1.78-7.15) $Bq.Kg^{-1}$ in the study area, all the concentration of radium is below the action level (370$Bq.Kg^{-1}$) as recommended by OECD. The mean and range of Uranium concentration (0.945) ppm with range (0.1- 2.2) ppm for soil samples in study area. All results measured are within the safe limit.

Keywords: Radon Gas, CR-39, Radium, Uranium

I. INTRODUCTION

Uranium is one of the radioactive element found in soil , water, air, plants, rocks and people, it is considered a heavy element with a high density (18.9$gm.cm^{-3}$) [1,2] . Heavy natural radionuclide uranium participates in the formation of the Earth crust. Atomic mass of the natural uranium is 238. It was discovered by the German chemist (M.H. Klaproth) in 1789[3]. Uranium is widely spread in nature [4]. Uranium is found in three radioactive isotopes ^{238}U, ^{235}U and ^{234}U with a relative abundance by mass 99.27%, 0.72% and 0.0054%, respectively [5]. Uranium can reach into the human body through different ways. It enters the body in a direct way by inhaling uranium-bearing dust particles or by drinking water which is polluted by uranium, or in an indirect way from the fertile soil layer via the food chain [6].

Radium ^{226}Ra mainly come from naturally occurring Uranium (^{238}U)[7]. The presence of uranium in the soil is related to several factors, including soil porosity, particle size, acidity function, as well as the reduction and oxidation effort. The ability of soil to retain uranium depends on factors the most important of which are chemical adsorption and the ability to ion exchange [8.9].

The Nuclear Track Detector (CR-39) is an organic detector, as it was discovered by Cartwright and Shirk in 1978 [10]. It consists of a polymeric material, which is prepared by a polymerization process of Poly Ally Diglycol Carbonate, which is a random hydrocarbon structure of Amorphous

composition, and the molecular formula of the detector is $O_7H_{18}C_{12}$. . Oil production in the Kirkuk field began in 1934, and its daily production capacity is 470,000 barrels, Within the Kirkuk governorate, there are 6 oil fields, as 4 of these fields are productive and 2 of them are non-productive and are awaiting development [11].

II. STUDY AREA

The four oil fields (Jumbor, Khabaz, Bai Hassan, Avanah) in the study area are located within the oil fields of Kirkuk Governorate - northern Iraq and the areas studied are located in the southeast and northwest part of the city of Kirkuk as shown in Fig.1.

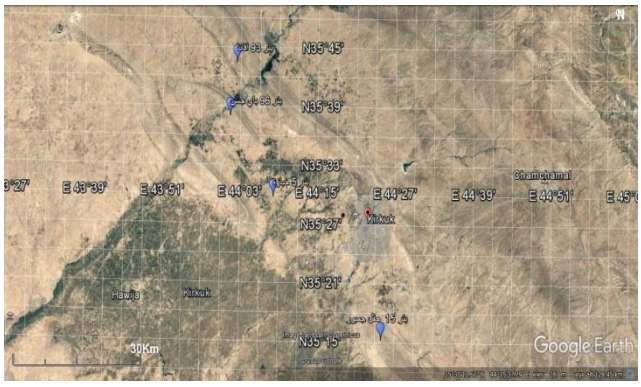

Fig.1 Selected oil fields in the study area

Well 96 in the Bai Hassan field is considered a high-productivity well, while well 3 in the same field has stopped producing oil for many years and is approximately (500) m away from well 96.

All samples were taken at a distance of 30 m from the oil well, because the places near the well contain chemicals used in drilling the well and the soil around the well is contaminated with these materials, and TABLE 1 shows the coordinates of the fields of oil wells in the study area.

Table 1. Coordinates of the fields of oil wells in the study area.

Field(well no.)	longitudes	latitudes
Jumbor (15)	44°24'58.92"	35°15'18.84"
Khabbaz (5)	44° 8'9.49"	35°29'48.80"
Bai Hassan (96)	44° 1'24.67"	35°38'2.79"
Avanah (93)	44° 2'23.38"	35°43'28.12"

III. EXPERIMENTAL METHODS

In the present study use CR-39 detector with 500µm British origin from the British company Tasl to measured Uranium and Radium concentrations for the selected oil fields soil and depths (15-30-45-60) cm, all the samples were taken at a distance of 30m from the oil well due to contamination of the soil near the well with the chemicals used in drilling the well. Twenty soil samples were collected, placed in plastic bags, dried, milled, and placed in the irradiation tube. The samples placed at a height of 3 cm were weighed and placed in the closed tube and the nuclear trace detector (2×1)cm² was placed at a height of 25 cm from the surface of the soil sample and the system was tightly closed, and the samples were placed From 2020 - 2-26 to 2020- 4-26, where samples were irradiated for 60 days as shown in Fig.2and the TABLE 2 show the oil fields and sample code and weight of soil samples.

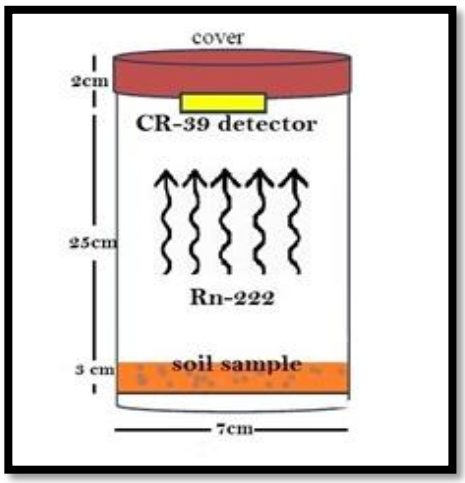

Fig 2. Irradiation tube technique

Table 2. Oil fields and sample code and weight of soil samples.

Field	well no.	Sample Code			
		15 cm	30 cm	45 cm	60 cm
Jumbor	15	SS1	SS2	SS3	SS4
Khabaz	5	SS5	SS6	SS7	SS8
Bai Hassan	96	SS9	SS10	SS11	SS12
Bai Hassan	3	SS13	SS14	SS15	SS16
Avanah	93	SS17	SS18	SS19	SS20

IV. ETCHING SOLUTION (NaOH)

To show the number of track formed, use a sodium hydroxide solution to scrape a light layer of the detector. The detector was placed in the NaOH solution with a N = 6.25 and placed in a water bath at a temperature of 70±1℃ for a continuous period of three hours, and the purity of NaOH was 99%. which is in the form of hemispherical granules.by using an optical microscope (10X.40X) To measure the number of tracks formed on the detector.

The diffusion constant K is calculated according to the used system (the irradiation tube), and because of the difference in the geometric dimensions of the diameter and the length, the diffusion constant K varies according to the design of the irradiation tube, and from the following relationship the diffusion constant K was found [12]:

$$K = \frac{1}{4} r(2\cos\theta c - \frac{r}{R\alpha}) \qquad (1)$$

r : The radius of the irradiation tube is 3.5 cm. θc: the critical angle for the detector and is about 35º . Rα: range of alpha particles produced from radon in air of 4cm.

After substituting the values in the above equation, we notice that the diffusion constant K is equal to K=0.67 $\mathrm{Tr.cm^{-2}d^{-1}/Bq.m^{-3}}$. From the following we find the radon concentration in air space C_{Rn} [13]:

$$C_{Rn} = \frac{\rho}{KT} \qquad (2)$$

ρ: the density of the resulting track effects measured in $\mathrm{Tr.cm^{-2}}$. T: the irradiation time is about 60 days.

Intensity rate of the track (D) measured by the following [12]:

$$D = \frac{\rho}{T} \qquad (3)$$

To measured Radium content in the soil sample for oil fields we must find Radon concentration in soil sample by using the following [14]:

$$C_s = \frac{\lambda_{Rn} C_a h\, t}{L} \qquad (4)$$

C_s : Radon concentration measured by $Bq.m^{-3}$. h: height of the air space (from the surface of the soil sample to the surface of detector) is about 25cm. L: thickness of the soil sample which is about 3cm.

The radioactivity of the Radon element is found in the samples by using the following relationship [15]:

$$A_{Rn} = C_s\, V \qquad (5)$$

A_{Rn}: radioactivity of radon in samples is measured in unit Bq. V: volume of the soil sample measured in m^3, and the volume found by the following:

$$V = \pi r^2 L \qquad (6)$$

r: radius of the irradiation tube which is equal to 3.5cm. L: thickness of the sample inside the tube is 3cm.

To find the Radium concentration in soil samples by using the following [16]:

$$C_{Ra} = \frac{\rho h A}{K M T_e} \qquad (7)$$

C_{Ra} : Radium concentration and measured by $Bq.gm^{-1}$. A: Sample surface area, measured in m^2 . M: soil sample weight in gm. Te: Radiation exposure time [17]:

$$T_e = \frac{T-1}{\lambda(1-e^{-\lambda t})} \qquad (8)$$

Calculating the concentration of Uranium for soil samples requires finding the number of radon atoms N_{Rn} in the samples through the following [17]:

$$A_{Rn} = \lambda_{Rn}.N_{Rn} \qquad (9)$$

Using the Ideal radiation equilibrium to find the number of uranium atoms [17]

$$\lambda_u N_u = \lambda_{Rn} N_{Rn} \qquad (10)$$

λ_u : Uranium decay constant is equal $4.2 \times 10^{-13} d^{-1}$. N_u : the number of uranium atoms.

Thus, the weight of uranium W_U is measured in units (gm) and is calculated from the following relationship [17]:

$$W_U = \frac{N_U M_U}{N_A} \qquad (11)$$

W_U : The mass number of Uranium, which is 238.03. N_A : The number of Avcadro which is $6.02\times10^{23}\,\mathrm{mol^{-1}}$.

Thus, the uranium concentration in (ppm) unit is found through the following relationship [18]:

$$C_U = \frac{W_U}{M_S} \qquad (12)$$

M_S : Weight of samples in gm.

V. RESULTS

A. Radon Concentration and activity in the soil sample.

Table 3 show the radon activity and Concentration in air and soil sample and the intensity rate of the track. The mean and range of radon Concentration in air for study area is (67.216 Bqm^{-3}) and (30.62-122.5 Bqm^{-3}) respectively and it is below the action level (200-600 Bqm^{-3}) as recommended by ICRP (1993) [19]. The mean of radon concentration in soil sample measuring by nuclear track detector was (3572.612 Bq. m^{-3}) with range (252.52-8585.58 Bq. m^{-3}) for study area. Mean of the activity of radon for the sample in irradiation tube is (0.704 Bq) with range (0.321-1.283 Bq) for the soil in study area, Because the soil surrounding the oil well is contaminated with chemical fertilizer used for agriculture, Fig.2 shows the linear relationship between the average intensity of track Tr. $cm^{-2}d^{-1}$and sample concentration in unit Bq. m^{-3} .

Table 3. Radon activity and Concentration in air and soil sample

Sample Code	C_{Rn} (Bqm^{-3})	D ($\mathbf{Tr.cm^{-2}d^{-1}}$)	C_s (Bq. m^{-3})	A_{Rn} (Bq)
SS1	44.34	29.40	1515.10	0.467
SS2	50.11	33.07	2020.14	0.525
SS3	66.81	44.10	3535.24	0.699
SS4	86.3	56.96	5302.86	0.904
SS5	30.62	20.21	252.52	0.321
SS6	36.08	23.81	747.45	0.378
SS7	64.03	42.26	3282.72	0.671
SS8	66.82	44.10	3535.24	0.699
SS9	75.17	49.61	4292.79	0.788
SS10	77.95	51.45	4545.31	0.817
SS11	108.57	71.66	7322.99	1.138
SS12	122.5	80.85	8585.58	1.283
SS13	41.76	27.56	1262.59	0.438
SS14	47.33	31.23	1767.62	0.496
SS15	50.11	33.07	2020.14	0.525
SS16	52.89	34.92	2272.65	0.554
SS17	58.47	38.58	2777.69	0.613
SS18	83.52	55.12	5050.34	0.875
SS19	86.3	56.96	5302.86	0.904
SS20	94.65	62.47	6060.41	0.992
Mean	67.216	44.369	3572.612	0.704
Range	30.62-122.5	20.21-80.85	252.52-8585.58	0.321-1.283

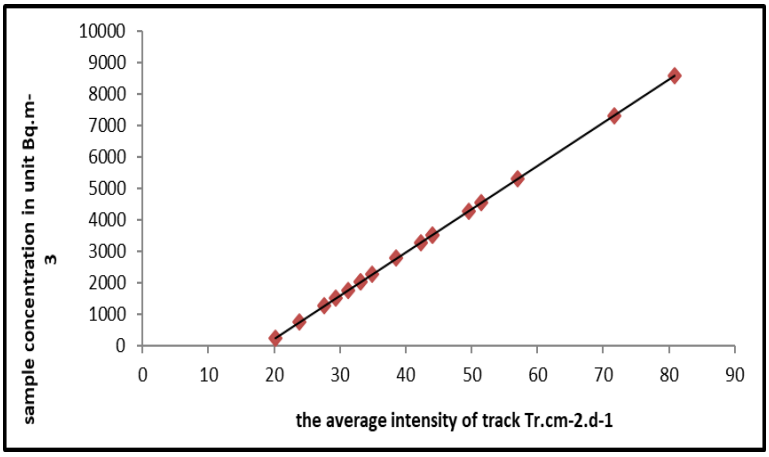

Fig 3. The linear relationship between the average intensity of track Tr. $cm^{-2}d^{-1}$and sample concentration in unit Bq. m^{-3}.

B. Radium Concentration for Soil Sample.

Table 4 shows the values of radium concentrations for different depths in the soil, starting from the surface of the earth, reaching the depth (60cm), with an increase of (15cm) each time using the CR-39 detector. The highest radium content ^{226}Ra was measured for sample SS12 in the Bay Hassan well 96 field for depth of 60cm, it was (7.15$Bq.Kg^{-1}$), and the lowest content was measured in the Khabaza oil field well 5 for a depth of about 15cm , it was (1.78 $Bq.Kg^{-1}$). The rate and range of radium concentrations in the selected oil fields are shown in Table 5.The rate of radium concentration in the soil of the study area using CR-39 detector reached (4.095 $Bq.Kg^{-1}$)and with a range (1.78-7.15) $Bq.Kg^{-1}$. According to the recommendations of the Organization for Economic Cooperation and Development (OECD) the permissible value is (370 $Bq.Kg^{-1}$) [20].

Table 4. Radium concentrations for different depths in the Soil.

Field	well no.	Radium Concentration $Bq.Kg^{-1}$			
		150cm	30cm	45cm	60 cm
Jumbor	15	2.04	2.59	3.57	5.17

Khabaz	5	1.78	2.12	4.02	4.10
Bai Hassan	96	4.82	4.70	6.53	7.15
Bai Hassan	3	2.83	3.16	3.27	3.75
Avanah	93	3.55	5.49	5.46	5.80

Table 5. The mean and Range of Radium concentration in the soil of the study area.

Field	Well no.	Mean $(Bq.Kg^{-1})$	Range $(Bq.Kg^{-1})$
Jumbor	15	3.34	2.04-5.17
Khabaz	5	3.00	1.78-4.1
Bai Hassan	96	5.8	4.7-7.15
Bai Hassan	3	3.25	2.83-3.75
Avanah	93	5.08	3.55-5.8
Overall mean	-	4.095	-
Total range	-	-	1.78-7.15

Fig 4. Shows the radium concentrations measured by the CR-39 detector for all soil samples and Fig 5. shows the Radium concentration selected oil fields

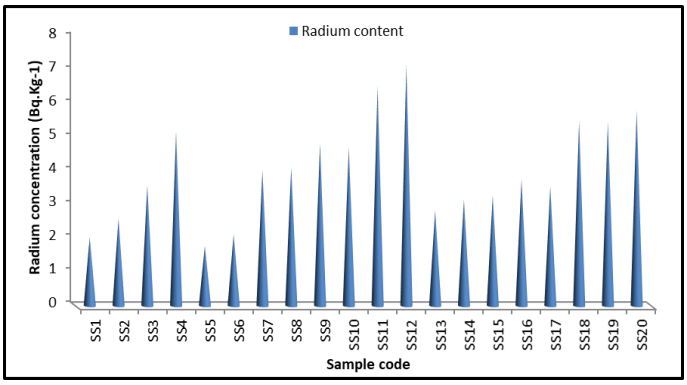

Fig 4. mean of radium concentrations measured by the CR-39 detector for soil samples.

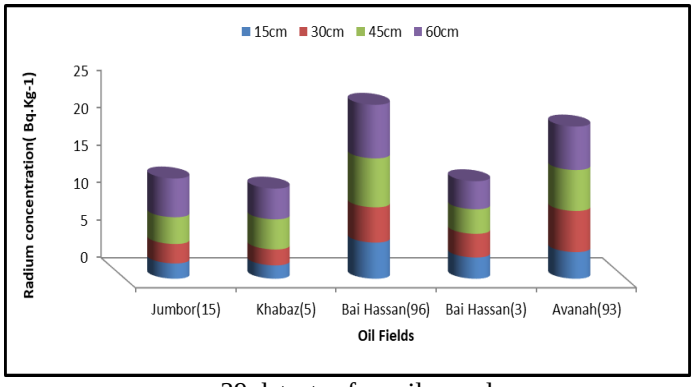

Fig 5. Radium concentration in (Jumbor (15), Khabaz (5), Bai Hassan (96), Bai Hassan (3), Avanah (93)) oil fields.

C. Uranium concentration in Soil sample

Table 6 show Uranium concentration for different depth in soil sample. The highest concentration of uranium was (2.2 ppm) in the SS12 sample for the Bai Hassan field well 96 at a depth of 60cm, and the lowest concentration of uranium in the sample SS5 in the Khabaza oil field well (5) depth of 15cm, which is (0.1 ppm). The mean of Uranium concentration in study area is (0.945ppm) and with range (0.1-2.2ppm).

Table 6. Uranium concentration for different depth in soil sample

Field	well no.	Uranium Concentration (ppm)			
		15cm	30cm	45cm	60cm
Jumbor	15	0.3	0.5	0.8	1.4
Khabaz	5	0.1	0.2	0.9	0.9
Bai Hassan	96	1.2	1.2	1.9	2.2
Bai Hassan	3	0.4	0.5	0.6	0.7
Avanah	93	0.7	1.4	1.4	1.6

Fig 6. shows the Uranium concentrations measured by the CR-39 detector for soil samples in oil field and. Table 7 shows the mean and Range of Uranium concentration(ppm) in the soil of the study area.

Fig 6. Uranium concentrations measured by the CR-39 detector for soil samples

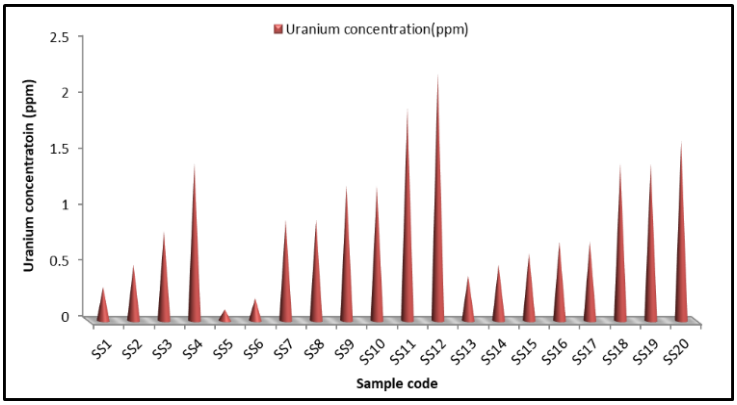

TABLE 7. Mean and Range of Uranium concentration(ppm) in the soil of the study area.

Field	Well no.	Mean (ppm)	Range (ppm)
Jumbor	15	0.75	0.3-1.4
Khabaz	5	0.53	0.1-0.9
Bai Hassan	96	1.63	1.2-2.2
Bai Hassan	3	0.55	0.4-0.7
Avanah	93	1.28	0.7-1.6
Overall mean	-	0.945	-
Total range	-	-	0.1-2.2

VI. CONCLUSION

The results of radioactivity of radon gas Radium and Uranium concentration in soil sample for oil field in Kirkuk city measured by nuclear track detector CR-39. The mean of radon concentration in soil sample is (3572.612 Bq. m^{-3}) with range (252.52-8585.58 Bq. m^{-3}) for study area. The Mean and range of the activity for radon gas in irradiation tube is (0.704 Bq) and (0.321-1.283 Bq) respectively for the soil in study area. The mean of Radium concentration to different depth for the four oil fields (Jumbor(15), Khabaz(5), Bai Hassan(96), Bai Hassan(3), Avanah(93)) are (3.34, 3.00, 5.8, 3.25, 5.08) $Bq.m^{-3}$ respectively, the total mean and range (4.095) $Bq.m^{-3}$ and (1.78-7.15$Bq.m^{-3}$) respectively. All the concentration of radium is below the action level (370$Bq.Kg^{-1}$) as recommended by OECD[20]. The mean of Uranium concentration in the study area is (0.945ppm) for the different depth in soil sample with range (0.1-2.2ppm). The result of this work show by increasing the depth of soil uranium concentration is increase and all the Uranium concentration less than the maximum permissible limit (11.7 ppm) and worldwide average value (2.8 ppm) as the recommended by UNSCEAR.

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