Measurement of radon gas concentrations in soil gas at AL-Najaf refinery, Al-Najaf province, west of Iraq by using CR-39 detector (cup technique).

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Summary

In this study, CR-39 detectors (cup technique) were used to measure radon gas concentrations in the soil gas for fifty samples collected from different positions at Al-Najaf refinery, where the study area (Al-Najaf refinery) was divided into four sector to achieve a comprehensive study for the area, the resuts showed that radon gas concentrations in the soil gas ranges between (39 ∓ 186 Bq/m^3 - 7341∓ 857 Bq/m^3) with an average (1342 ∓ 123 Bq/m^3), and the Annual Effective Dose for the public due to inhalation of radon gas near the soil in the outdoor air for lung tissue lies between (0.019 mSv/y - 3.608 mSv/y), this study also involved a theorotical estimations for radium content in the soil, uranium concentration in the soil, radon concentration near the soil, area exhalation rate for radon gas from the soil and mass exhalation rate for radon gas from the soil. All the results were within the accepted limit recommended by WHO and ICRP for the Annual Affective Dose due to outdoor inhalation of radon gas, as a result, there is no any health risk for the workers of the refinery due to inhalation of radon gas at outdoor air. Key words: radon, soil gas, Al-Najaf refinery, Al-Najaf province, CR-39, cup technique.

1. Introduction

The soil is derived from rocks, and the mineralogy of the soil depends on two factors, the first one is the natural conditions by which the soil formed and the second one is the minerals in rocks from which the soil derived, where rocks are merely combinations of minerals, and minerals have specific chemical compositions. Rocks in general can be classified into three main types: igneous rocks, sedimentary rocks and metamorphic rocks[1].Radon-222 is a decay product of radium-226 which comes from uranium-238 series, radon-222 can damage the tissues of lungs with alpha particles emitted from its progenies if it is inhaled with high concentrations[2]. High levels of radon-222 are associated with granite igneous rocks, shale and dirty quartz sedimentary rocks, phosphate deposits and some beach sands, which may contain high levels of uranium-238. Radon-222 existence in the soil depends on the degree of bedrock fracture, soil porosity and intervening pathways, where the permeability of rocks is an important factor influence radon-222 availability at the surface, as a result, the rock with low level of uranium content, such as limestone may has high level of radon-222[3]. In general, the concentration of radon-222 increases as the depth from the soil surface increases[4]. The soil of Al-Najaf refinery is a sandy soil, and there are many activities inside the refinery may effect the composition of the soil and radon-222 concentrations in its gas such as digging, transport of oil products, establishing new buildings, maintenances, refining of the crude oil, eliminating wastes... etc, so the soil of the refinery is not stable and varies due to the activities inside the refinery.CR-39 detector is a thermoset polymer with an atomic composition ($C_{12}H_{18}O_7$), can detect alpha particles with an energy range lies between 0.1-100Mev, as a result, it can be used to measure radon concentrations[5], the range of alpha particles energies of CR-39 detector is wide in comparison with other plastic detectors, such as, Lexan, Makrofol E polycarbonates, Cellulose nitrate (CA80-15, CN85, LR115 and Daicel), where each detector has it's own energy range[6].The concentration of radon-222 gas in the air of the space inside the cup (C_{Rn}) can be calculated using cup technique according to Equation (1)[7]**.**

$$\mathbf{C_{Rn}} = \frac{\rho_t}{KT} \qquad (1)$$

Where: ρ_t: is the track density in (Track/cm^2), k: calibration factor (0.2 for our cup) and T: exposure time in (day)**.**

the concentration of radon gas in the sample (C) can calculated according to Equation (2), where the thickness of the sample inside the cup was taken into the consideration [7]:

$$\mathbf{C} = \frac{\lambda_{Rn} \times C_{Rn} \times h \times T}{\textit{thickness of the sample inside the cup}} \qquad (2)$$

$\mathbf{C_{Ra}}$: radium gas concentration in the soil gas in (Bq/kg) is shown in Equation (3) [8]:

$$\mathbf{C_{Ra}} = \frac{\rho_t h A_S}{M_S K T_e} \qquad (3)$$

Where h is the distance between the detector and the surface of the sample in (cm), $A_S$is the sample surface area (cm^2), M_S: sample mass in (Kg), T_e is the effective exposure time in (day) as shown in Equation**(4)** [8]**:**

$$\mathbf{T_e} = T - \frac{1}{\lambda_{Rn}(e^{-\lambda_{Rn}T} - 1)} \qquad (4)$$

Where $\lambda_{Rn}$is the radon decay constant in (day^{-1}). The area exhalation rate (E_A) in (Bq.m^{-2}.h^{-1}), mass exhalation rate (E_M) in (Bq.kg^{-1}.h^{-1}), A_{Rn} is the radon-222 activity in (Bq), and uranium concentration (C_U) in (g/kg) or can be calculated in (mg/kg) and (part per million (ppm)), are given in Equations (5), (6), (7) and (11), respectively [8]**.**

$$\mathbf{E_A} = \frac{C \times V_{cup} \times \lambda_{Rn}}{A_S[T + \lambda_{Rn}^{-1}(e^{-\lambda_{Rn}T} - 1)]} \qquad (5)$$

$$\mathbf{E_M} = \frac{C \times V_{cup} \times \lambda_{Rn}}{M_S[T + \lambda_{Rn}^{-1}(e^{-\lambda_{Rn}T} - 1)]} \qquad (6)$$

$$A_{Rn} = C_{Rn} \times V_{cup} \qquad (7)$$

$$A_{Rn} = \lambda_{Rn} N_{Rn} \qquad (8)$$

$$\lambda_U N_U = \lambda_{Rn} N_{Rn} \quad (9)$$

$$W_U = \frac{N_U \times W_{mol}}{N_{Av}} \quad (10)$$

$$C_U = \frac{W_U}{W_S} \quad (11)$$

Where V_{cup}: an internal cup volume in (m^3), λ_U: uranium-238 decay constant (4.98×10^{-18} S^{-1}), N_U: number of uranium-238 nuclei, W_U: uranium weight in (g),) and W_S: sample weight in (g).

Radon concentration near the soil ($C_{radon\ near\ the\ soil}$) , and the Annual Effective Dose $(AED)_{outdoor\ inhalation}$ in (mSv/y) due to radon gas inhalation in the outdoor air can be calculated due to the following Equations [9]:

$$C_{radon\ near\ the\ soil} = C\sqrt{d/D} \quad (12)$$

$$(AED)_{outdoor\ inhalation} = C_{radon\ near\ the\ soil} \times F \times I \times DCF \quad (13)$$

Where: d: exhalation diffusion constant (0.05 cm^2/s), D: eddy diffusion coefficient (50000 cm^2/s) ,F is the equilibrium factor between radon gas and it's progenies (0.6), I: is the mean of time spent outdoor for a person per year (1760 h/y), and DCF is the dose conversion factor (9000000) mSv.m^3/h for radon exposure).

Many studies conducted in Al-Najaf governorate related to radon gas and it's health risks, here some of them shown in Table (1):

Table (1): Radon gas concentrations in the soil gas obtained from previous studies conducted near our study area (in AL-Najaf province).

Region and year	Radon concentration in the soil gas in (Bq/m^3)	Depth	Technique	Reference
Al-Ameer district, 2011	3260 – 62	25 cm	RAD7 monitor	[10]
Al-Ameer district, 2011	4830 – 123	35 cm	RAD7 monitor	[10]
Al-Ameer district, 2011	1660 – 158	5 cm	RAD7 monitor	[10]
Al-Ansar district, 2012	758 – 56	-	CR-39 SSNTD	[11]
Selected soil samples from Al-Najaf province, 2017	1194 – 506	15 cm	CN-85 SSNTD	[8]

2. Study area

The area of study (Al-Najaf refinery) is located in AL-Najaf province, in the western part of the Republic of Iraq at a latitude 32°13'N and a longitude 44°15', Al-Najaf refinery was built in October 2006, with an area of about 887293 m^2, one hundred miles (160 km) from Baghdad the capital, and it contains three refining units[12]. The area of study (Al-Najaf refinery) was shown in Figure (1) and Figure (2) shows the map for the study area and the positions of soil samples.

The soil of Al–Najaf governorate consists of layers, the layers are silty sand, clayey sand, gypsum, and sand, The order of these layers due to the depth differs from location to another inside Al–Najaf governorate, where they are randomly ditributed according to the depth [13].

Al–Najaf governorate is located in the central part of Iraq with an area of 28824 km2 and an elevation above the sea level 70 m. The surface of it is gradually descends from the southwest to the notheast at a rate of decline equils to 1m per 2 km, the population of Al – Najaf governorate equils to 1500522 according to 2017 census [14].

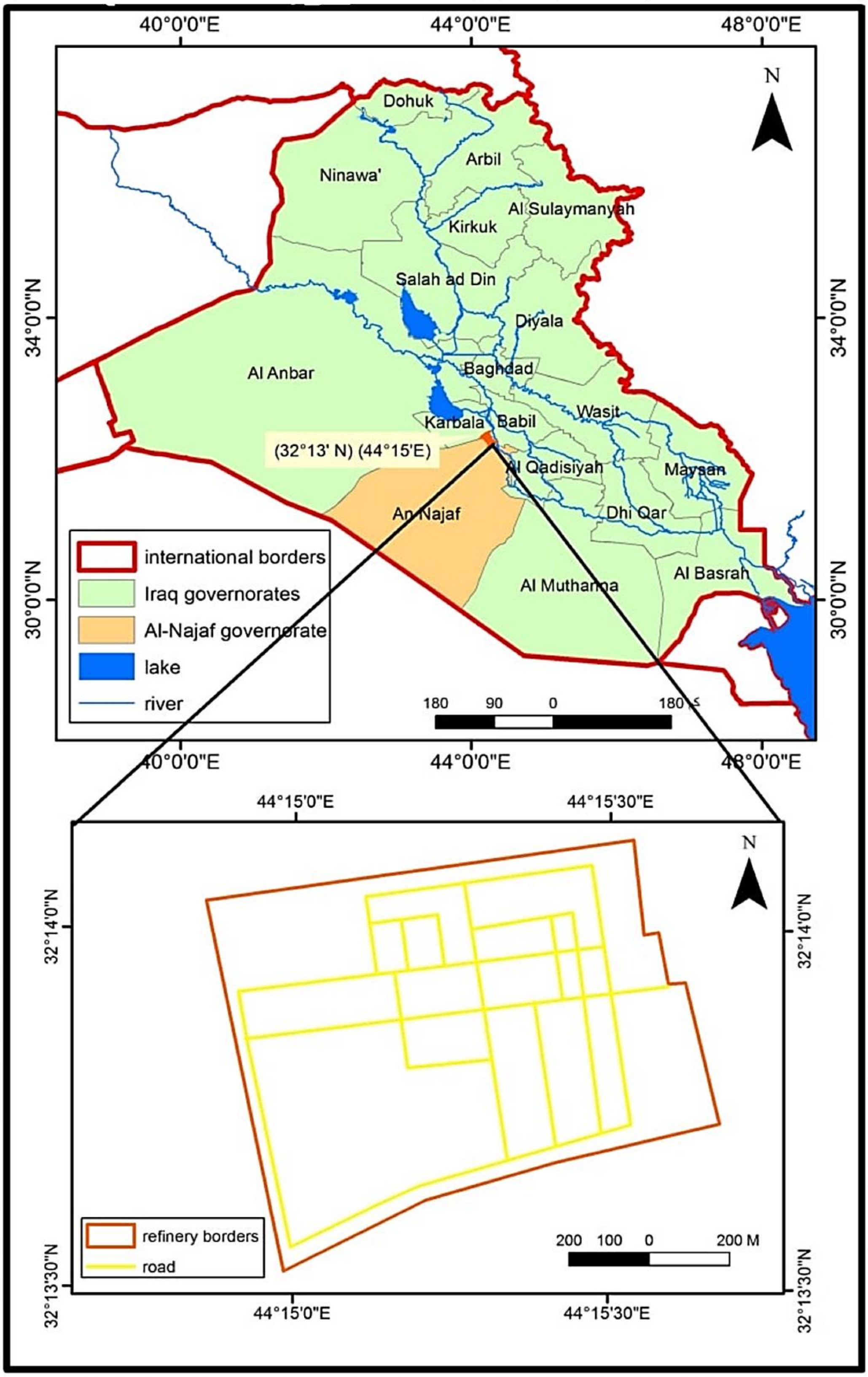
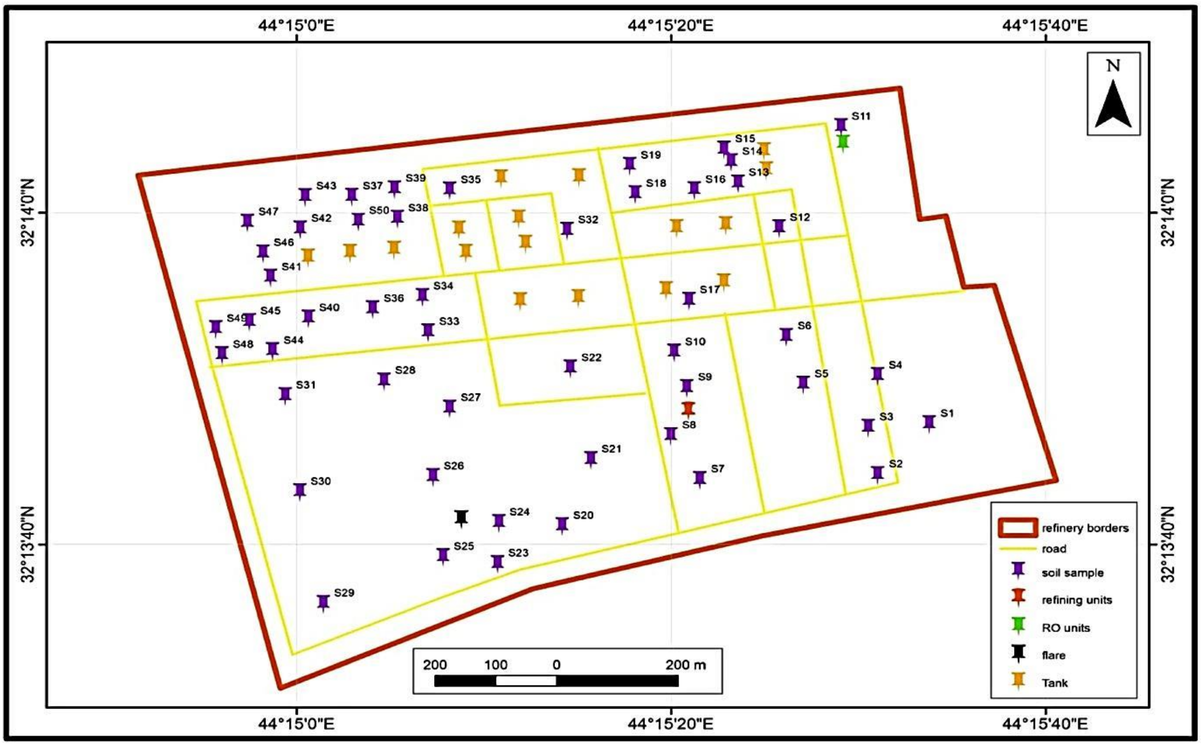


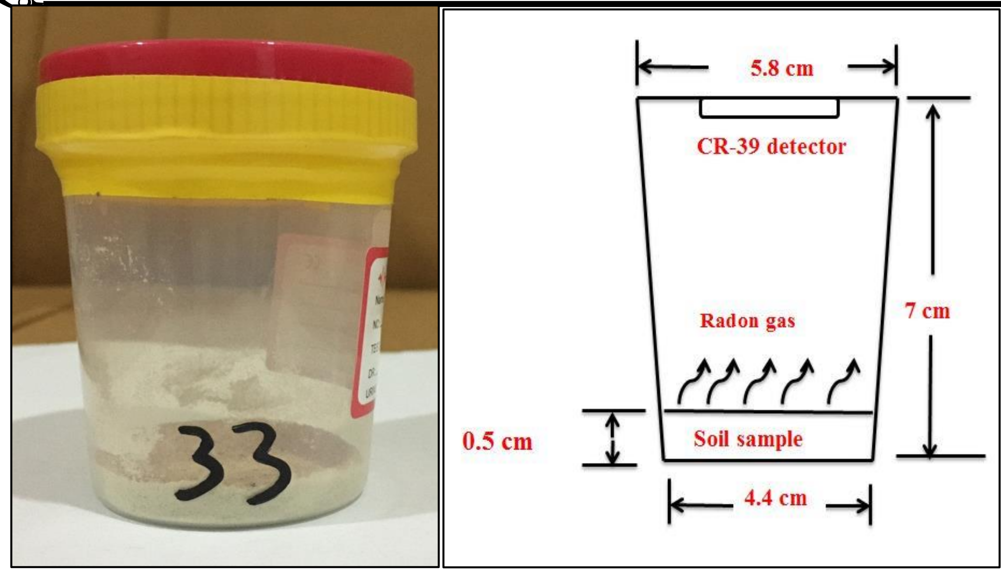
Figure (1): The location of the study area.



Figure(2): Map of samples positionss at the study area.

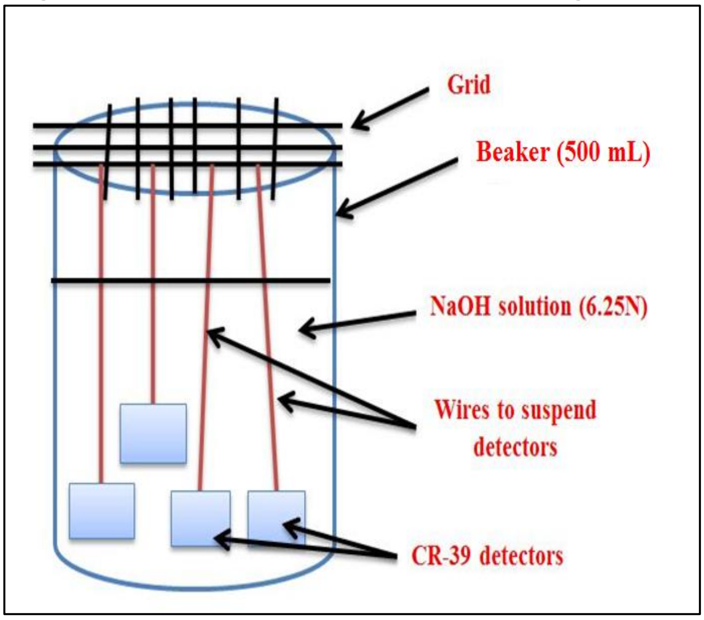
3. Sample Collection and Test Procedures:

Fifty samples of soil were collected from selected positions of the Al-Najaf refinery as shown in Figure(2) with a depth of 20 cm from the surface durring the period (22/11/2021 – 6/12/2021). After sample collection, the samples were sieved to eliminate the impurities (such as plant pieces, plastic pieces, insects ... etc) and then dried in an oven under the following conditions: time = 3 hours, temperature = 80°C, sample surface area = (30 cm × 30 cm) and sample thickness = 5mm. After the drying stage, the dried samples were milled with a grinder (100 g capacity) to get a homogeneous powder, and then the powder of the dried samples was poured into plastic cups, the mass of the sample inside the cup was 10 g, and the dimensions of the cup and sample is shown in Figure (3). The sample was left for 30 days for a radiological equilibrium, after this period, the detectors were swiftly planted at the upper cover of the cup as shown in Figure (3). After this step, the cups were left in a steady state for 60 days for detectors exposure. After this stage, the detectors were carefully removed from the cups and etched with a chemical solution of NaOH with normality of 6.25N for 5 hours by using a water bath under 70°C, the scheme of the beaker used in the water bath and the water bath used in the etching process is shown in Figure 4) and Figure (5), respectively. And then, the detectors were washed with a distilled water to make the tracks visible. After the etching process, the tracks (pits) on the surface of the detectors were counted by using the TASL system as shown in Figure (6) (at the nuclear laboratory, college of science, Kufa university). The calibration factors for the cup of soil was taken from a previous research which involved many cups with heights ranging from 5 cm to 45 cm, and from the reults of that research, the calibration factor for the cups with heights varying between 5 cm to 25 cm equals to 0.3, and for other heights equals to 0.2, as a result, the calibration factor for our cup of soil is considered to be equal to (0.3) [2].

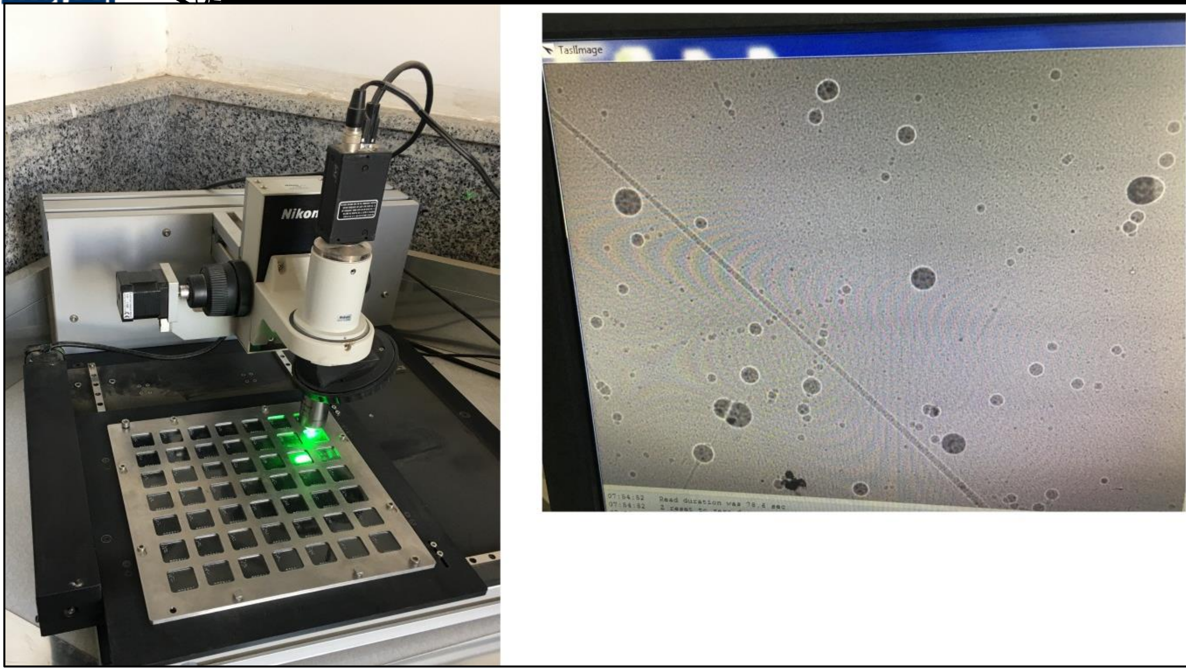


Figure(3): A schematic diagram for the cup used in this work.

Figure (4): The water bath used in the etching process.



Figure(5): A schematic diagram for the beaker with detectors suspended inside it which was used in the etching process.



Figure(6): TASL system and an image taken by TASL system.

4. Results and discussion

The concentrations of radon gas in the soil gas samples are presented in Table (2), these results were calculated according to Equation (1) and Equation (2) after calculating the density of tracks on the detectors and knowing the value of the calibration factor (K=0.3). Figure(7) illustrates that maximum value was recorded in S20 was 7341 ∓ 857 Bq/m^3, while the minimum value occurred in S30 (39 ∓ 186 m^3), with an average 1342 ∓ 123 Bq/ m^3.

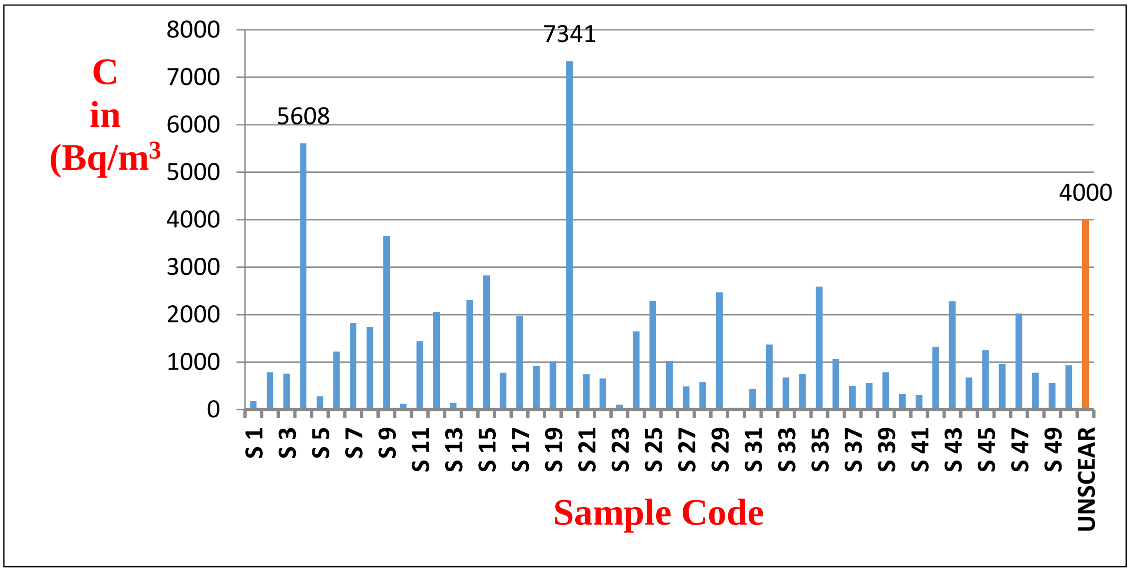
Table (2): The coordinates of soil samples and the concentration of radon gas in the soil gas for each sample (C).

sample code	Coordinates		detector number	in ρ_t (track/cm^3)	C in (Bq/m^3)
	Longitude	Latitude			
S1	32°13'47.20"N	44°15'33.81"E	2839	23	180 ∓ 166
S2	32°13'44.14"N	44°15'31.06"E	2840	100	784 ∓ 80
S3	32°13'46.99"N	44°15'30.55"E	2841	97	761 ∓ 83
S4	32°13'50.11"N	44°15'31.05"E	2842	715	5608 ∓ 609
S5	32°13'49.59"N	44°15'27.08"E	2843	36	282 ∓ 151
S6	32°13'52.47"N	44°15'26.18"E	2844	156	1224 ∓ 17
S7	32°13'43.85"N	44°15'21.55"E	2845	232	1820 ∓ 68
S8	32°13'46.50"N	44°15'20.01"E	2846	222	1741 ∓ 57

S9	32°13'49.39"N	44°15'20.86"E	2847	467	3663 ∓ 332
S10	32°13'51.52"N	44°15'20.19"E	2848	16	125 ∓ 174
S11	32°14'5.12"N	44°15'29.11"E	2849	183	1435 ∓ 13
S12	32°13'59.02"N	44°15'25.79"E	2850	262	2055 ∓ 102
S13	32°14'1.73"N	44°15'23.59"E	2851	18	141 ∓ 172
S14	32°14'3.01"N	44°15'23.23"E	2852	294	2306 ∓ 138
S15	32°14'3.76"N	44°15'22.85"E	2853	360	2824 ∓ 212
S16	32°14'1.32"N	44°15'21.27"E	2854	99	776 ∓ 81
S17	32°13'54.65"N	44°15'20.98"E	2855	251	1969 ∓ 90
S18	32°14'1.07"N	44°15'18.10"E	2856	117	918 ∓ 61
S19	32°14'2.78"N	44°15'17.81"E	2857	129	1012 ∓ 47
S20	32°13'41.05"N	44°15'14.21"E	2858	936	7341 ∓ 857
S21	32°13'45.05"N	44°15'15.73"E	2859	95	745 ∓ 85
S22	32°13'50.57"N	44°15'14.64"E	2860	84	659 ∓ 98
S23	32°13'38.78"N	44°15'10.75"E	2861	13	102 ∓ 177
S24	32°13'41.25"N	44°15'10.82"E	2862	210	1647 ∓ 44
S25	32°13'39.19"N	44°15'7.84"E	2863	292	2290 ∓ 135
S26	32°13'44.02"N	44°15'7.30"E	2864	130	1020 ∓ 46
S27	32°13'48.15"N	44°15'8.20"E	2865	62	486 ∓ 122

S28	32°13'49.79"N	44°15'4.69"E	2866	73	573 ∓ 110
S29	32°13'36.37"N	44°15'1.44"E	2867	315	2471 ∓ 161
S30	32°13'43.11"N	44°15'0.19"E	2868	5	39 ∓ 186
S31	32°13'48.90"N	44°14'59.40"E	2869	55	431 ∓ 130
S32	32°13'58.87"N	44°15'14.47"E	2870	175	1373 ∓ 4
S33	32°13'52.74"N	44°15'7.05"E	2871	86	675 ∓ 95
S34	32°13'54.88"N	44°15'6.74"E	2872	96	753 ∓ 84
S35	32°14'1.30"N	44°15'8.19"E	2873	330	2588 ∓ 178
S36	32°13'54.14"N	44°15'4.08"E	2874	135	1059 ∓ 40
S37	32°14'0.92"N	44°15'2.97"E	2875	63	494 ∓ 121
S38	32°13'59.59"N	44°15'5.40"E	2876	71	557 ∓ 112
S39	32°14'1.38"N	44°15'5.25"E	2877	100	784 ∓ 80
S40	32°13'53.58"N	44°15'0.64"E	2878	42	329 ∓ 145
S41	32°13'56.04"N	44°14'58.63"E	2879	39	306 ∓ 148
S42	32°13'58.96"N	44°15'0.21"E	2880	169	1326 ∓ 2
S43	32°14'0.91"N	44°15'0.47"E	2881	291	2282 ∓ 134
S44	32°13'51.61"N	44°14'58.73"E	2882	86	675 ∓ 95
S45	32°13'53.38"N	44°14'57.49"E	2883	159	1247 ∓ 14
S46	32°13'57.50"N	44°14'58.22"E	2884	122	957 ∓ 55
S47	32°13'59.36"N	44°14'57.39"E	2885	258	2024 ∓ 97
S48	32°13'51.37"N	44°14'56.03"E	2886	99	776 ∓ 81

S49	**32°13'52.94"N**	**44°14'55.70"E**	**2887**	**71**	557 ∓ 112
S50	**32°13'59.43"N**	**44°15'3.31"E**	**2888**	**119**	933 ∓ 58
UNSCEAR (2000)					**4000**
Maximum value					**7341 ∓ 857**
Minimum value					**39 ∓** 186
The average					**1342 ∓ 123**



Figure(7): Radon gas concentrations in the soil gas at Al-Najaf refinery in (Bq/m^3). In Table (3), radium concentrations (C_{Ra}) were calculated according to Equation (3), uranium concentrations (C_U) were calculated according to Equation (11), area exhalation rate for radon gas, mass exhalation rate for radon gas, radon gas concentration near the soil and the Annual Effective Dose (AED) $_{\text{outdoor inhalation}}$ were calculated according to Equation (5), Equation (6), Equation (12) and Equation (13), respectively. The maximum value of radium concentration (C_{Ra}), uranium concentration (C_U) and Annual Effective Dose (AED) $_{\text{outdoor inhalation}}$ estimated at S20 were 0.730 Bq/kg, 8.157 mg/kg, 0.070 ∓ 0.0083 mSv/y and the minimum value at S30 were 0.004 Bq/kg, 0.044 mg/kg, 0.0004∓0.0017 mSv/y respectively, as explained in Figure (8), Figure(9) and Figure (10).

Table (3): Concentration of radium (C_{Ra}), concentration of uranium (C_U), area exhalation rate (E_A) and mass exhalation rate calculated for each sample.

Sample code	C_{Ra} (Bq/kg)	C_u (ppm) or (mg/kg)	E_A (Bq/m^2.h)	E_M (× 10^{-4}) (Bq/kg.h)	$C_{radon\ near\ the\ soil}$ in (Bq/m^3)	(AED) *outdoor inhalation*
S1	0.018	0.200	0.043	0.008	0.180	0.002 ∓ 0.0014
S2	0.078	0.871	0.186	0.037	0.784	0.007∓ 0.0007
S3	0.076	0.845	0.181	0.035	0.761	0.007 ∓ 0.0007
S4	0.558	6.230	1.332	0.261	5.608	0.053 ∓ 0.0059
S5	0.028	0.314	0.067	0.013	0.282	0.003 ∓ 0.0013
S6	0.122	1.359	0.291	0.057	1.224	0.012 ∓ 0
S7	0.181	2.022	0.432	0.085	1.820	0.017 ∓ 0.0007
S8	0.173	1.934	0.414	0.081	1.741	0.017 ∓ 0.0007
S9	0.364	4.069	0.870	0.170	3.663	0.035 ∓ 0.0033
S10	0.012	0.139	0.030	0.006	0.125	0.001 ∓ 0.0016
S11	0.143	1.595	0.341	0.067	1.435	0.014 ∓ 0.0003
S12	0.204	2.283	0.488	0.096	2.055	0.020 ∓ 0.0011
S13	0.014	0.157	0.034	0.007	0.141	0.001 ∓ 0.0016
S14	0.229	2.562	0.548	0.107	2.306	0.022 ∓ 0.0014
S15	0.281	3.137	0.671	0.131	2.824	0.027 ∓ 0.0021
S16	0.077	0.863	0.184	0.036	0.776	0.007 ∓ 0.0007
S17	0.196	2.187	0.468	0.092	1.969	0.019 ∓ 0.0010
S18	0.091	1.020	0.218	0.043	0.918	0.009 ∓ 0.0004
S19	0.101	1.124	0.240	0.047	1.012	0.010 ∓ 0.0003
S20	0.730	8.157	1.743	0.342	7.341	0.070 ∓ 0.0083
S21	0.074	0.828	0.177	0.035	0.745	0.007 ∓ 0.0007
S22	0.066	0.732	0.156	0.031	0.659	0.006 ∓ 0.0009
S23	0.010	0.113	0.024	0.005	0.102	0.001 ∓ 0.0016
S24	0.164	1.830	0.391	0.077	1.647	0.016 ∓ 0.0006
S25	0.228	2.544	0.544	0.107	2.290	0.022 ∓ 0.0014
S26	0.101	1.133	0.242	0.047	1.020	0.010 ∓ 0.0003
S27	0.048	0.540	0.115	0.023	0.486	0.005 ∓ 0.0010
S28	0.057	0.636	0.136	1.40	0.573	0.005 ∓ 0.001
S29	0.246	2.745	0.587	6.03	2.471	0.023 ∓ 0.0016
S30	0.004	0.044	0.009	0.0957	0.039	0.0004∓0.0017
S31	0.043	0.479	0.102	1.05	0.431	0.004∓ 0.0011
S32	0.137	1.525	0.326	3.35	1.373	0.013∓ 0.0001
S33	0.067	0.749	0.160	1.65	0.675	0.006 ∓0.0009
S34	0.075	0.837	0.179	1.84	0.753	0.007 ∓ 0.0007
S35	0.257	2.876	0.615	6.32	2.588	0.025 ∓ 0.0019
S36	0.105	1.176	0.251	2.58	1.059	0.010 ∓ 0.0003
S37	0.049	0.549	0.117	1.21	0.494	0.005 ∓ 0.001
S38	0.055	0.619	0.132	1.36	0.557	0.005 ∓ 0.001
S39	0.078	0.871	0.186	1.91	0.784	0.007 ∓ 0.0007
S40	0.033	0.366	0.078	0.804	0.329	0.003 ∓ 0.0013
S41	0.030	0.340	0.073	0.746	0.306	0.003 ∓ 0.0013
S42	0.132	1.473	0.315	3.230	1.326	0.013 ∓ 0.0001

S43	0.227	2.536	0.542	5.570	2.282	0.022 ∓ 0.0014
S44	0.067	0.749	0.160	1.650	0.675	0.006 ∓ 0.0009
S45	0.124	1.385	0.296	3.040	1.247	0.012 ∓ 0
S46	0.095	1.063	0.227	2.340	0.957	0.009 ∓ 0.0004
S47	0.201	2.248	0.481	4.940	2.024	0.019 ∓ 0.001
S48	0.077	0.863	0.184	1.890	0.776	0.007 ∓ 0.0007
S49	0.055	0.619	0.132	1.360	0.557	0.005 ∓ 0.001
S50	0.093	1.037	0.222	2.280	0.933	0.009 ∓ 0.0004
Maximum value	**0.730**	**8.16**	**16.570**	**0.3417**	**7.341**	**0.070 ∓ 0.0083**
Minimum value	**0.004**	**0.04**	**0.089**	**0.0018**	**0.039**	**0.0004 ∓ 0.001**
The average	**0.134**	**1.49**	**3.030**	**0.0625**	**1.342**	**0.013 ∓ 0.001**

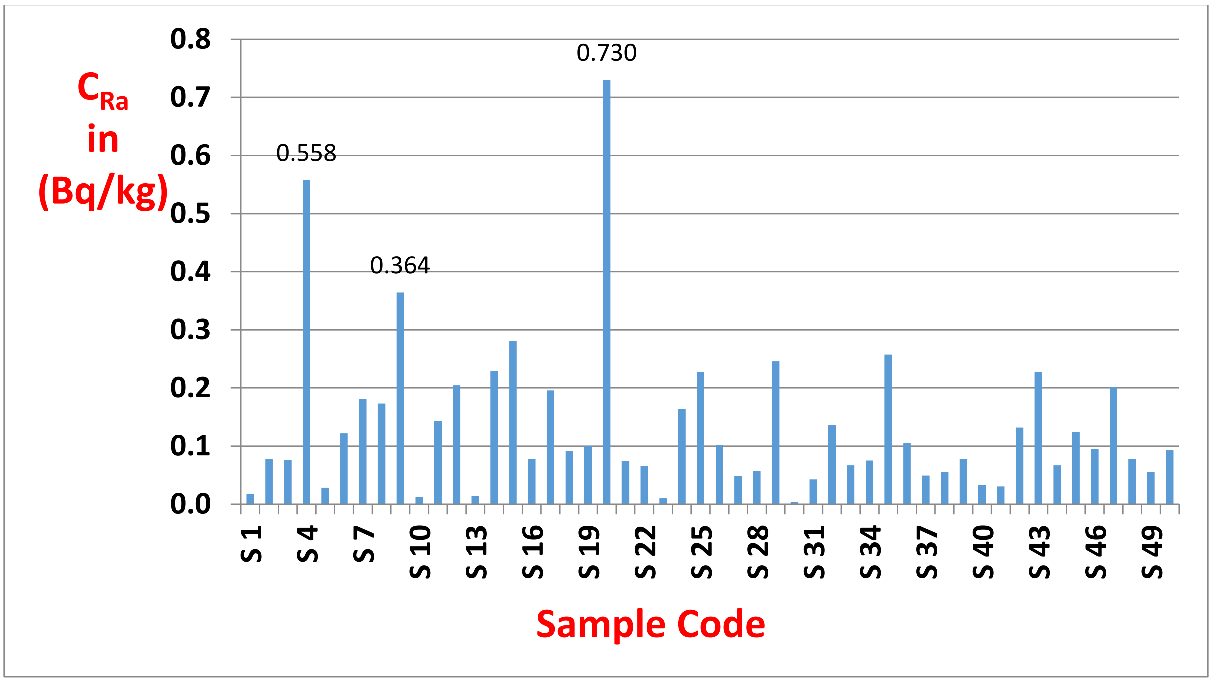


Figure (8): Radium concentration for the soil samples.

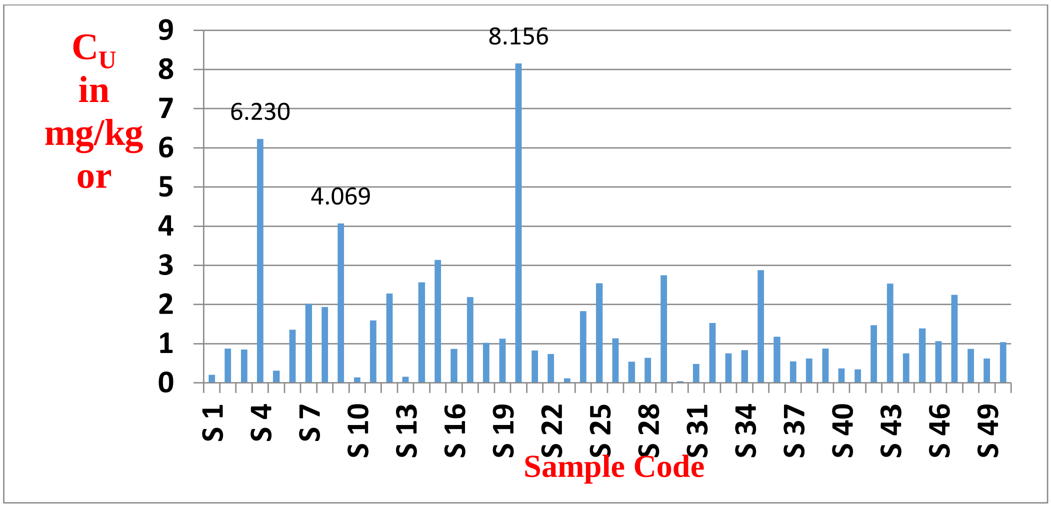


Figure (9): Uranium-238 concentrations for the soil samples.

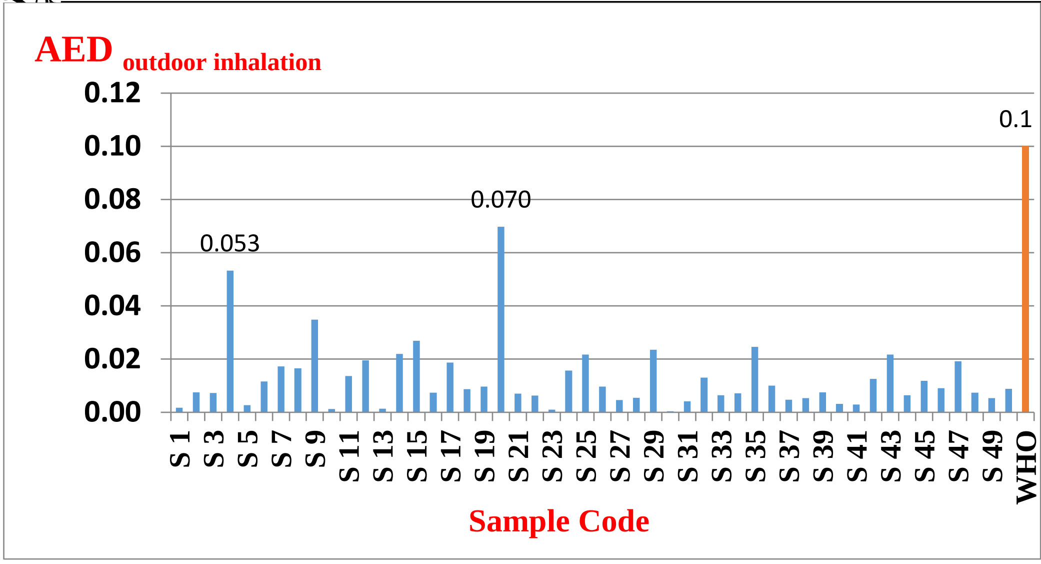


Figure (10) The annual effective dose AED due to inhalation in the outdoor air in (mSv/y) for lung tissue.

5. Conclusions

From the results, it is clear that some samples were with high concentrations of radon gas in comparison with the results obtained from the previous studies mentioned in Table (1), this may be due to industrial operations conducted in the study area (AL-Najaf refinery) which may cause an increase of radium concentration in the soil which comes from oil wastes (sludge and scale), oil wastes have complex, variable composition, or may be due to geological reasons such as a fracture in the bed rock under the soil in the area, the porosity of the soil and rocks from which the soil is derived. The concentrations of radon–222 gas in the soil gas for the soil samples with codes (S4 and S20) excede the action level recommended by UNSCEAR (2000).

The Annual Effective Doses due to radon–222 gas inhalation in the outdoor air which is resulted from the soil contributions are all less than the value recommended by WHO, and there is no any radiological risk resulted from radon gas inhalation to the health of the workers of the refinery if they existed in these positions for a long period (long-term exposure).

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