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Estimation of radon-gas, uranium-238 concentrations in the surface soil samples and alpha risk index at many Northern Iraqi Refineries

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***Abstract*---**Radon gas concentrations and uranium-238 concentration in soil samples at the surface (depth = 15 cm) were determined for fifteen samples in three refineries (Al-Kasak, Al-Qayarah, Baiji) using “sealed can technique”. CR-39 solid-state nuclear pathway detectors. The concentration of radon in surface samples at the Al-kasak refinery ranged from (1509 to 8323.2 Bq/m^3) with an average of (4916.1 Bq/m^3), and the surface exhalation rate in surface soil samples ranged from (1110.3 to 612.7 Bq/m^3) with an average (861.5 Bq/m^3). The effective activity of radon ($A_{Rn}{}^{222}$) are (15.82 to 8.84 Bq/kg) with an average (12.33 Bq/kg).In a Al-Qayara refinery, the radon concentration in surface samples ranged from (11709.6 to 10281 Bq/m^3) with an average of (10995.3 Bq/m^3), and the surface exhalation rate in surface soil samples ranged from (861.5 to 756.3 Bq/m^3) with an average of (808.9 Bq/m^3), the effective activity of radon gas ($A_{Rn}{}^{222}$) are (12.28 to 10.756 Bq/kg) averaged (11.518 Bq/kg).In the Baiji refinery, the radon concentration in surface samples ranged from (13501 to 5062 Bq/m^3) with an average of (9281.5 Bq/m^3), and the surface exhalation rate in the surface soil samples ranged from (992 to 372 Bq/m^3) with an average of (682 Bq/m^3), the activity of radon gas ($A_{Rn}{}^{222}$) are (14.13 to 5.427 Bq/kg) with an average (9.778 Bq/kg), while the uranium-238 concentrations

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were in the samples (1.254 to 0.711 ppm) with an average (0.9825 ppm) in the Al- Kasak refinery, (0.988 to 0.863 ppm) in an average of (0.9255 ppm) at the Qayara refinery and (1.12 to 0.419 ppm) in an average (0.77 ppm) at the Baiji refinery.By comparison, we found all results in the limitations of ICRP regulations. The alpha risk index for radon was also determined because it was highest in Al-Kasak refinery, (S9-Site) (0.0791), and lowest in Baiji refinery, (S15-Site)(0.0365). When compared with the European Commission's(EC), Waste Control NORM, it was found that had no effect 0n refinery workers.

***Keywords*---**Radon Concentration, Soil Sample, CR-39 Track Detector, Radon Exhalation Rate, alpha risk index.

1. Introduction

In general, radiation is either natural or man-made. Humans serve a variety of roles, and they are exposed to a variety of sources. Different proportions of radiation. The environment is affected by radiation. Its effects last for years, changing the genetic composition of those who are exposed to it. For both people and animals, this results in a genetic flaw that manifests itself. As a result, we must not overlook a crucial issue that will affect future generations. It is the arrival of pollution's influence on water and soil, and from there, on the food chain for both people and animals. High quantities of radiation are emitted by several sources. Ionization and nuclear explosions that damage the environment and their impacts on humanity, the Hiroshima and Nagasaki disasters in 1945, processes arising from resource consumption in the pharmaceutical sector, agriculture, and other industries, all demonstrated the necessity to investigate the impact. Several approaches have arisen to detect radiation and determine the level of environmental contamination as well as strategies to remedy it, including the CR-39 effect detector technology, which was utilized to compute the concentrations in this study. Uranium and radon levels in soil samples from three refineries: Baiji, Kasak, and Qayara. Soil samples were obtained from the surface of the soil. During oil extraction activities, refineries are one of the most vulnerable regions to radioactive contamination. These natural elements, uranium and radon, are dissolved in the soil and interfere with the oil and water processes connected with oil production. The mechanical activities that take place in oil production and oil refining processes in petroleum products are an industrial source of radiation. Uranium, thorium, and radium are radioactive heavy metals(Sujo, L. C., Cabrera et al.,2004) . In the soil, radioactive heavy metals arise naturally (such as uranium, thorium and radium), as uranium represents the most abundant percentage in the earth's crust and tends to spread through the soil because the rocks in the outer crust have suffered from erosion and atmospheric processes. Soil depends on information related to the processes of accumulation and movement away from the site of pollution, as the accumulation and movement of radioactive materials depends on the interaction of materials and compounds with the solid part of the soil, and the type of this interaction reflects the ability of the soil to retain radioactive materials (Souad, N. Al-Azzawi,2020) (UNSCEAR,2017) (Washington, C. M et al.,2015). In one of the studies, a measurement of the uranium concentration was conducted for soil samples taken from different areas located on the international highway linking Baghdad to the Anbar Governorate, which witnessed many military actions during and after the 2003 war, and to show the percentage of contamination of these areas, which include rural residential and agricultural areas as well as being areas for breeding Livestock of all kinds has been determined ,Concentrations by calculations based on comparison with standard models. The trace detector (CR-39) was used as a method for detecting uranium for soil samples for the six study areas, and the Fallujah region showed the largest percentage of uranium among other samples (1.7ppm) and this applies to the reality where this area witnessed More extensive military operations than other areas (UNSCEAR.,2011). Whereas, the results of our study were compared with internationally permissible limits. The value of uranium in soils is set by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) at 32 Becquerels, while the International Atomic Energy Agency has set the value of uranium in soils. Uranium in slag (8-5 x 105) Bq/kg and therefore does not pose any danger to the lives of refinery workers, the field and the environment (UNSCEAR.,2011)(UNSCEAR.,2017).

2. Materials and Methods

2.1 Location of study

This study included collecting samples as shown in Figures (1,2,3) from three refineries in the northern region, namely Qayara and Al-Kasak refineries within Nineveh Governorate and Baiji refinery in Tikrit Governorate, where it is located (Al-Qayara refinery is located 60 km south of the city of Mosul), It is located (Al-Kasak Refinery, 70 km northwest of Mosul), and (Bayji Refinery, which is located in Salah al-Din Governorate, about 130 km north of Baghdad in the middle of the road leading from Baghdad to Mosul).

Figure 1. A map shows the Location of AL.Kasik refinery.

Figure 2. A map shows the Location of AL.qayara refinery

Figure**. 3.** A map shows the Location of Baiji refinery.

2.2 Sample collection

(Fifteen) different samples were collected from (Al-Kasak, Qayara and Baiji) refineries. It included samples of soil in the oil product area. It was taken from the soil surface with a weight of one kilogram. And put them in plastic bags with a label. Each sample contains the sample type, location, and date it was taken.

2.3 Sample preparation

The samples were taken to the University of Mosul, where they were prepared at the College of Environmental Science and Technology for study. Samples were prepared by placing them in tightly closed 4 cm high plastic cans, weighing the sample with a sensitive balance with a sensitivity of 0.001 g and leaving it for 30 days to obtain the optimal balance between radium and radon, which was calculated as 98% from an ideal equilibrium. Through the radioactivity balance relationship (Cowie, M et al., 2009) (Saeed, S. H et al.,2015) (RAND O. Farqad et al.,2022).

$$A_{Rn=}A_{Ra}(1-e^{-\lambda Rn\, \boldsymbol{t}})$$

Where A_{Rn} is the effective radon and $A_{R\propto}$ represents the effectiveness of radium and λ_{Rn} represents the decay constant of radon 0.1814 d^{-1}, t which is the time required to reach equilibrium.

3. Measurements

The uranium concentration and specific radioactivity of radon gas were measured for all samples using a solid-state CR-39 nuclear track detector. The samples were placed in an irradiation system consisting of two plastic cups identical in shape and size, and filter paper was placed between them and closed with adhesive tape tightly. CR-39 detectors were left inside the chambers for 30 days (Hameed, T. K et al., 2016) (Mohammad, A. M. 2011). then we prepared the corrosion solution, the chemical substance 6.25 N NaOH. After the irradiation period was completed, the reagents were removed and placed inside the chemical etching solution, and then they were placed in a water bath at a temperature of 70 degrees Celsius for 4 hours. The radiation background was 130 $Tr.Cm^{-2}$ and it was removed from the intensity of particulate effects. Alpha came from the samples under study, and the effect of Alpha was measured (using an optical microscope with a magnification of 400x). Figure 2 shows the irradiation system that was used (Kadhim, N. F et al.,2014).

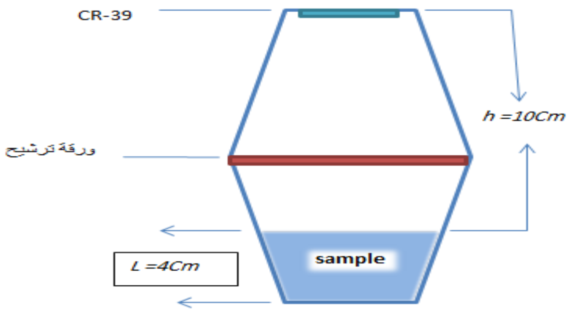

Figure 4. Explains the irradiation system.

3.1 The following equations numbered (1) to (7) were used for calculation (M Al-Badrani, M., 2006) (I Hassan, H., 2014):

1-Measure the concentration of radon gas through the following relationship

$P = KC_a T$ ....(1)

Where P (Tr. cm^{-2}) represents track density, K (Tr. $\frac{Cm^{-2}}{h\ Bq.m^{-3}}$) represents the diffusion constant, T represents the irradiation time in hours and C_a represents the radioactivity of radon in the air space in units (Bq. m^{-3}).

2- Determination of the propagation constant k

$K = \frac{1}{4} r (2Cos\,\theta - \frac{r}{R\alpha})$..........(2)

Where r represents the radius of the cup and is equal to 2.58 cm and Rα represents the range of alpha particles in the air produced by radon and is equal to 4.16 cm and represents the θ angle of 35 °

3- Determining radon concentration in samples (C_s) :

The radon concentration in the samples was calculated by the following relationship:

$C_S = C_a\, T\, h\, \lambda Rn / L$ ....(3)

Where C_s represents the radon activity within the samples in units (Bq. m^{-3}),h represents the height of the detector from the sample surface in cm units, T represents the irradiation time in days, λ_{Rn} represents the radon decay constant and L represents the thickness of the sample in cm.

4- The radioactivity of radon gas produced from samples in the (Bq) unit:

$A_{Rn} = C_s V$................. (4)

$V = L\,\pi\, r^2$...................... (5)

where A_{Rn} is the radioactivity of radon gas in the samples (Bq) unit and V volumesamples (m^3) and r radius equal 2.55 (cm).

5- Counting the number of radon atoms N_{Rn} in the samples:

$N_u \lambda_u = N_{Rn} \lambda_{Rn}$ .... (6)

After determining the number of radon atoms, the number of uranium atoms is found through the law of radiation balance.

$N_{Th}\, \lambda_{Th} = N_{Rn} \lambda_{Rn}$ .... (7)

6- Determining the mass of uranium in the samples:

$W_{U=N_u A_u / A_{av}}$ .... (8)

Where represents the mass of uranium in the samples, A_U represents the mass number of uranium, and N_{av} represents the number of Avogadro, N_U number of uranium atoms in the samples.

7- Determining the uranium concentration (C_u) in samples in ppm units:

$pp_m = \frac{W_U}{W_S}$ .... (9)

Where W_S represents the mass of the sample in (gm).

8- Alpha gravity index calculator

$I\alpha = \frac{A\ Rn}{200\ Bq/kg} \leq 1$........(10)

4. Results and Discussion

The results show a high values of radioactivity (concentrations of radon-gas) and u-238 in the surface soil samples (S9-site) as in table (3) at Al-Kasik refinery (15.82 Bq/kg) and (1.254 ppm) respectively, while the lower radioactivity and concentration of u-238 (S6-site) (8.84 Bq/kg) and (0.711 ppm) respectively.

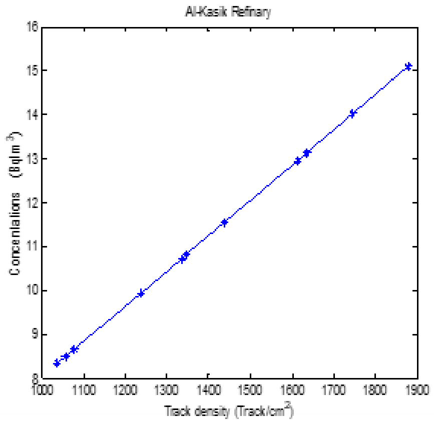

Figure 5. Radon gas concentration as a function of path density in Al-kasik Refinery samples

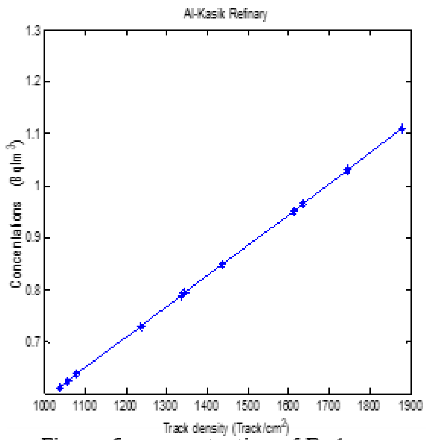

Figure 6. concentration of Radon gas as afunction of trak density at Al-kasik refinery

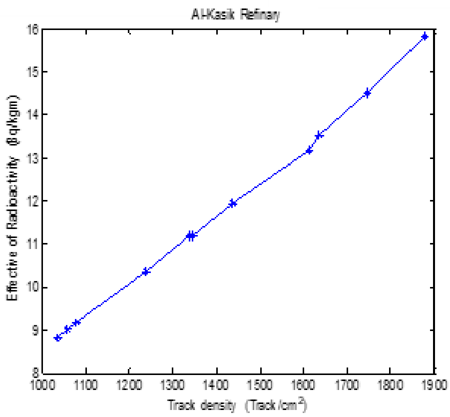

Figure 7. efective radioactivity of radon as function of trak density at Alkasik refinery

At Al-Qayara refinery the higher levels of radioactivity and concentration of u-238 (S1-site) are (12.28 Bq/kg) and (0.988 ppm) respectively in the soil samples, where the rate of radioactivity and low concentration of u-238 (S3-site) are (10.756 Bq/kg) and (0.863 ppm) respectively. Soil texture and composition are very important in determining radon gas concentration, so the difference in concentration values is a function of exact density, porosity of samples and soil type. The lower value of concentrations is attributed to the type of soil that is characterized by soft soil and low porosity (low diffusion coefficient) which leads to low radon concentration, and finally is a function of uranium concentration. For the same reason in all soil samples,

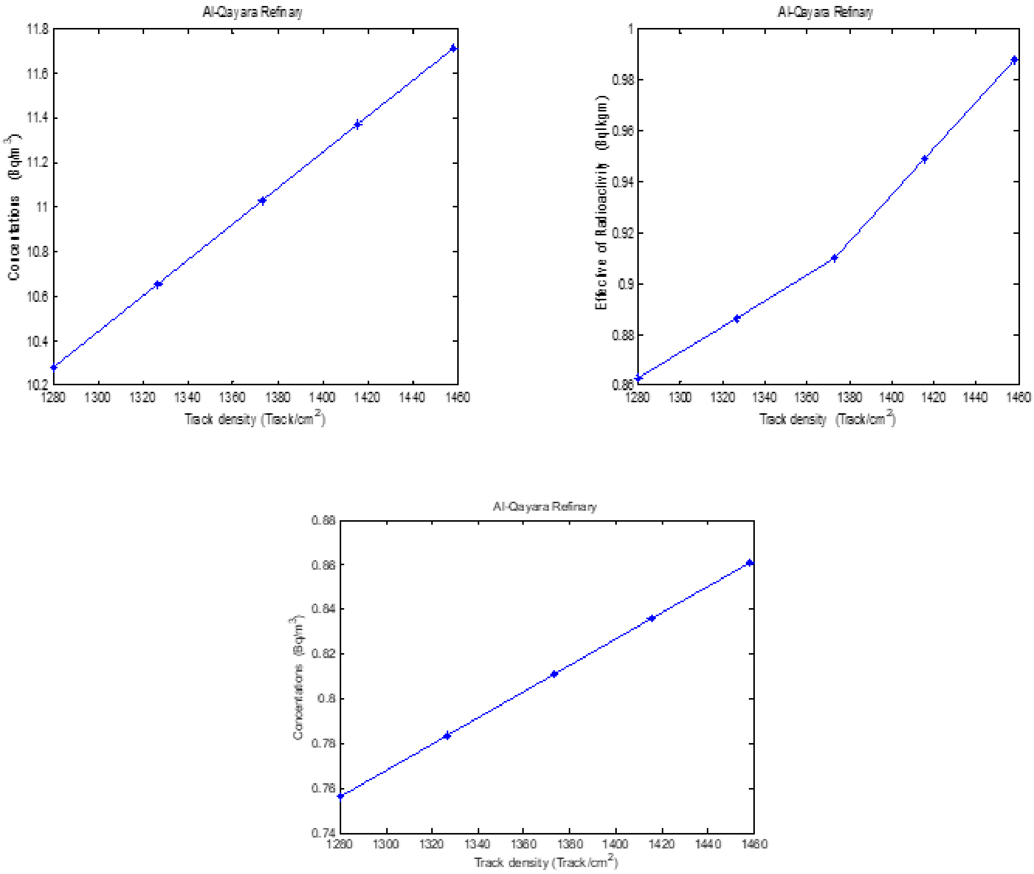

Figure **10.** Radon gas concentration as a function of path density in Al-qayara Refinery

we found in Baiji refinery, the radioactivity and u-238 concentrations are (14.13 Bq/kg) and (1.254 ppm) respectively, in the (S10-site), and the lower radioactivity and u-238 concentrations are (5.427 Bq/kg) and (0.419 ppm) respectively, in (S13-site). Our results are compare with the permissible limits of radon gas in the soil, as the permissible limit of Worldwide average (soil)(32 Bq/kg) with researchers (Husain, H. A et al., 2017), Iraq (Surface soil)(3.7-43.3 Bq/kg) with researcher (UNSCEAR, 2008), Iraq (Soil) (49.8-97.6) with researcher (Ali, K. K., Shafik, S. S et al.,2017),and the permissible concentration of uranium in the soil according to the International Atomic Energy Agency(IAEA) (50 Bq/kg) as in ref] (UNSCEAR,.1994). We find that the results which obtained in our study within the maximum permissible limits globally.

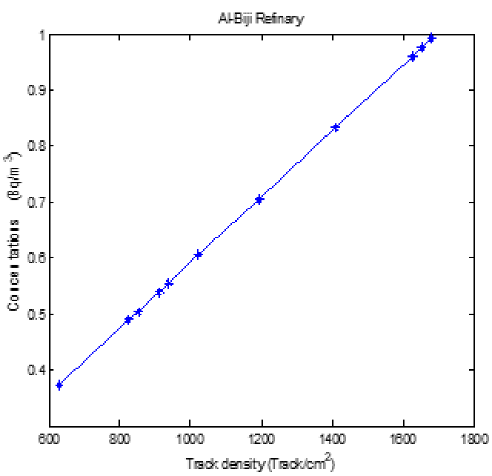

Figure **11**. concentration of Radon gas as afunction of trak density at Al-Baiji refinery

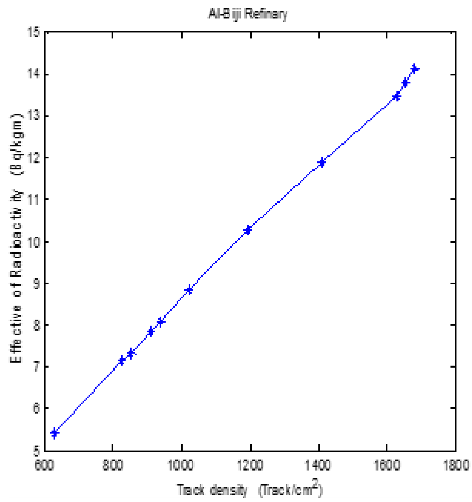

Figure.**12** .efective radioactivity of radon as function of trak density at Al-Baiji refinery

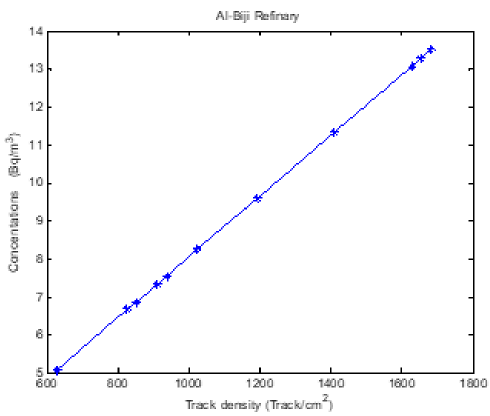

Figure.**13** .Radon gas concentration as a function of path density in Al-Baiji Refinery samples

The alpha risks of radon were calculate for all samples (equ. 10), and compare with the limits regulated by the European Commission(ec.europa.eu was first indexed by Google more than 10 years ago.,2011). for Waste Control in Industry NORM, which is equal to (≤1)(Zhukovsky, M. Et al.,2019).it was found that the results were within the maximum permissible limits and had no effect on a refinery workers.

Table1- shows the (ID & type) of samples and their locations in the refineries and coordinates for each location

Samples ID	Type of samples	Location	the coordinates of the location
S1	Soil/ Light gas oil production pumps area	Qayara refinery	(35° 47.409'N 43° 16.908'E)
S2	Soil/Asphalt production pumps area	Qayara refinery	(35° 47.327'N 43° 16.687'E)
S3	Soil/Naphtha and Light gas oil cooling exchangers	Qayara refinery	(35° 47.260'N 43° 16.788'E)
S4	Soil/dram naphtha pot	Alkasik refinery	(36° 27.563'N 42° 40.347'E)
S5	Soil/Crode oil	Alkasik refinery	(36° 27.491'N 42° 40.376'E)
S6	Soil/Naphtha tanks	Alkasik refinery	(36° 27.434'N 42° 40.110'E)
S7	Soil/diesel tanks	Alkasik refinery	(36° 27.489'N 42° 40.168'E)
S8	Soil/fueloil tanks	Alkasik refinery	(36° 27.394'N 42° 40.266'E)
S9	Soil/loading pumps	Alkasik refinery	(36° 27.623'N 42° 40.311'E)
S10	Soil/RCR	Baiji refirenery	(35° 0.122' N, 43°29.859'E)
S11	Soil/ gasoline	Baiji refirenery	(35° 0.158' N, 43° 29.896'E)
S12	Soil/gas	Baiji refirenery	(35° 0.291' N, 43°30 .007'E)
S13	Soil/ kerosen	Baiji refirenery	(35° 0.378' N, 43°29.866' E)
S14	Soil/Crode oil	Baiji refirenery	(35°0.250' N, 43° 29 .745'E)
S15	Soil/sweet naphtha	Baiji refirenery	(35° 0.220' N, 43° 29 .701'E)

Table 2- illustrates the sample location, number of impacts, and radon concentration in the air and samples

Sample ID	Location	Samples Type	ρ (Tr.cm^{-2})	Cα (Bq.m^{-3}10^{3})	Cs (Bq.m^{-3} 10^{3})
S1	Qayara refinery	Soil	1458	0.861	11.713±0.31
S2	Qayara refinery	Soil	1373	0.811	11.029±0.298
S3	Qayara	Soil	1280	0.756	10.281±0.287

	refinery				
S4	Alkasik refinery	Soil	1237	0.730	9.941±0.282
S5	Alkasik refinery	Soil	1635	0.966	13.139±0.32
S6	Alkasik refinery	Soil	1037	0.612	8.333±0.25
S7	Alkasik refinery	Soil	1078	0.637	8.663±0.263
S8	Alkasik refinery	Soil	1612	0.952	12.954±0.322
S9	Alkasik refinery	Soil	1879	1.110	15.1±0.35
S10	Baiji refirenery	Soil	1680	0.992	13.501±0.33
S11	Baiji refirenery	Soil	1626	0.960	13.067±0.0.32
S12	Baiji refirenery	Soil	1194	0.705	9.595±0.28
S13	Baiji refirenery	Soil	630	0.372	5.062±0.20
S14	Baiji refirenery	Soil	1028	0.607	8.255±0.26
S15	Baiji refirenery	Soil	852	0.503	6.847±0.23

Table 3- the radioactivity of radon (Bq), the radioactivity of radon (Bq/kg), the number of uranium atoms, the mass of uranium in the samples, the concentration of uranium(ppm)in the samples.

Sample ID	Location	Samples type	ρ (Tr.cm^{-2})	A_{Rn}^{222} (Bq)	A_{Rn}^{222} Bq/kg)(	$N_U \times 10^{17}$	W_U $*10^{-6}$	W Sample (gm)	C_U PPm
S1	Qayara refinery	Soil	1458	0.956	12.28	1.95	77.05	78	0.988±0.026
S2	Qayara refinery	Soil	1373	0.901	11.45	1.834	72.31	79.5	0.91±0.025
S3	Qayara refinery	Soil	1280	0.839	10.756	1.71	67.57	78	0.863±0.024
S4	Alkasik refinery	Soil	1237	0.811	10.36	1.654	65.35	78.23	0.835±0.024
S5	Alkasik refinery	Soil	1635	1.073	13.54	2.187	86.41	79.23	1.091±0.027
1.	Alkasik refinery	Soil	1037	0.680	8.84	1.384	54.68	76.9	0.711±0.022
S7	Alkasik refinery	Soil	1078	0.707	9.19	1.44	56.90	76.9	0.739±0.022
S8	Alkasik refinery	Soil	1612	1.057	13.19	2.155	85.15	80.33	1.06±0.026
S9	Alkasik refinery	Soil	1879	1.233	15.82	2.515	99.38	79.23	1.254±0.029
S10	Baiji refirenery	Soil	1680	1.102	14.13	2.245	88.71	79.23	1.12±0.027
S11	Baiji refirenery	Soil	1626	1.067	13.467	2.177	86.024	79.23	1.085±0.027
S12	Baiji refirenery	Soil	1194	0.783	10.289	1.594	62.98	76.1	0.827±0.024
S13	Baiji refirenery	Soil	630	0.413	5.427	0.84	33.19	76.1	0.419±0.020
S14	Baiji refirenery	Soil	1028	0.674	8.85	1.371	54.17	76.1	0.711±0.022
S15	Baiji refirenery	Soil	852	0.559	7.316	1.14	45.04	76.4	0.589±0.020
									12.079 | 0.197

Table 4: Alpha risk index

Sample ID	Type Sample	Location	Alpha risk index
S1	Soil/ Light gas oil production pumps area	Qayara refinery	0.0614
S2	Soil/Asphalt production pumps area	Qayara refinery	0.057
S3	Soil/Naphtha and Light gas oil cooling exchangers	Qayara refinery	0.0537
S4	Soil/dram naphtha pot	Alkasik refinery	0.0518
S5	Soil/Crode oil	Alkasik refinery	0.0677
S6	Soil/Naphtha tanks	Alkasik refinery	0.044
S7	Soil/diesel tanks	Alkasik refinery	0.045
S8	Soil/fueloil tanks	Alkasik refinery	0.065
S9	Soil/loading pumps	Alkasik refinery	0.0791
S10	Soil/RCR	Baiji refirenery	0.071
S11	Soil/ gasoline	Baiji refirenery	0.0673
S12	Soil/gas	Baiji refirenery	0.0514

S13	Soil/ kerosen	Baiji refirenery	0.0271
S14	Soil/Crode oil	Baiji refirenery	0.044
S15	Soil/sweet naphtha	Baiji refirenery	0.0365

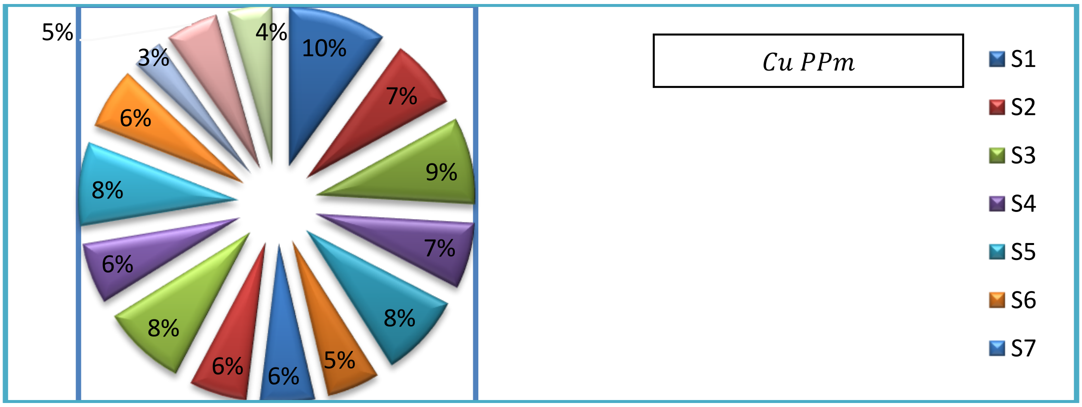

Figure 14.The concentration of uranium in the sample.

Conclusions

The radioactivity of radon, uranium-238 concentration and alpha risk index were achieve in the soil surface at many of the Iraqi northern refineries(Al-Qayarah, Al-Kasik and Al-Baiji refineries) using the CR-39 technique, the results show that within the maximum permissible limits, in spite of small difference in concentrations between the sites of refineries . However, maintaining the quantities of naturally occurring radioactive materials needs continuous monitoring in order to preserve the workers, public and environment.

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