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Radon Concentration in oily Sludge Produced from Oil Refineries in the Southern oil plant at Basra Governorate - Iraq

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ABSTRACT

Oily sludge waste could be included Naturally Radioactive Materials (NORM) from the three natural radioactive series and Potassium. The sludge usually returned to the environment and could cause a serious hazard. The aim of the present work is to investigate the radioactive Radon gas (^{222}Rn) concentration, which reflects the present of NORM concentration in the sludge waste produced from crude oil refinery. The selected samples of oil sludge, had been grinded and dried then a passive method for measuring radon concentration performed using solid State Nuclear Detectors (SSNTD) technique. Two detectors were used in this work; CR39 and L115-II. Samples of sludge were collected, from different locations and from each location three samples collected in different depth. The arithmetic average radon concentration was 26089 Bq/m^3, with maximum131618 Bq/m^3 and minimum 2523 Bq/m^3.

Keyword: Oily-sludge, Natural radioactivity, Radon, Solid State Nuclear Track Detectors.

INTRODUCTION

One of the major challenges faced by oil refineries is safe disposal of oil sludge, scales and production water generated during the processing of crude oil, as improper disposal would lead to environmental pollution, particularly soil and ground water contamination with toxic hydrocarbons, salt, trace of heavy elements. These contamination effect soil, groundwater, and surface water [1]. Radioactive waste contains NORM, mainly from the natural radioactive series (238U, 235U and 232Th), together with potassium (40K), or production water, which contains mainly Radium isotopes which are alpha and gamma emitter. These radioactive isotopes are part of natural radioactive series, ^{226}Ra a daughter of ^{238}U and ^{228}Ra is a daughter of ^{232}Th, and the dissolved in production water when it conduct the rocks surrounding the oil-well during crude oil production. Sludge, oily sediment that is produced during cleaning operation of oil separators storage tanks and other surface equipment, is considered as the main source of sludge contaminations. Sludge a mixture of residues left in the process of oil refinery. It's made up of sand pump up during extraction of the oil, heavy Hydrocarbons like paraffin, scale duct surface, pump and valves. The presents of radioactive elements in the oily sludge has been recorded in several works [2-8]

The present of radioactivity in the soil environment originated from Uranium and Thorium natural series and their progeny. The important progeny are Radon (^{222}Rn) gas and Thoron (^{220}Rn) gas. The existences of these isotopes in a certain sample reflect how much uranium or thorium in it.

The aim of the present work is to measure alpha particle concentration due to Radon exhalation form in the oily sludge using the Solid State Nuclear Track Detectors (SSNTDs). Nuclear track detection technique based on radon measurement with the passive detectors CR39 and LR115-II [9]. These detectors are commonly used because of their simplicity, availability, ease of handling and low cost. The principle of this technique based on the production of track in the detector due to alpha particle emitted from radon and its progeny. After exposure the track made visible by chemical etching using NaOH or KOH. Counting the visible tracks which leads to track density is then converted to radon concentration [10].

MATERIALS AND METHODS

2. Measuring Procedure

The sludge samples were collected from different location of the Oil Refineries in the Southern oil assembly at Basra Governorate – Iraq. Each sample is dried in the oven at 110 ^oC, milled in grinder, and sieved. Each dried sample of oil sludge placed in the bottom (1 cm height) of closed plastic cylinder container of high 10cm and diameter 4cm as shown in Figure-1[11]. The 1.5x1.5cm^2 SSNTDs films are placed on the bottom of the cover of the containers, to allow irradiation process with Radon only to take place. After 90 days of irradiation the detectors was removed carefully from the cylinder and etched with NaOH solution with condition 2.5N at 70 ^oC for 7 hour for CR39 film and 2.5N at 60 ^oC for 2 hour in the case of LR115-II film [11]. The detectors then washed many times by distill water and dried with tissue papers. The numbers of tracks due to alpha particles interaction are counted by the means of optical microscope 400X.

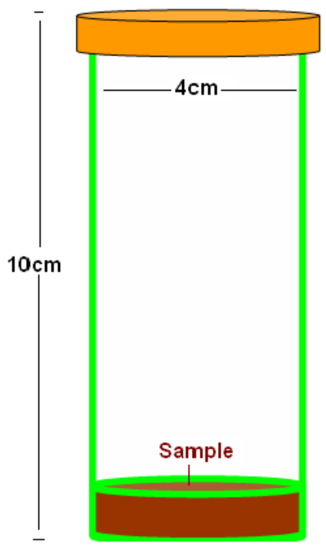

Figure-1.Schematic diagram showing the geometry of the CR39 and LR115-II Solid state Nuclear Track Detectors based radon passive dosimeter.

2. Theoretical aspect

The alpha particle densities that was 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 [10]

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

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 make use P_i^{CR}, P_i^{LR} values, one can calculate the $^{220}A/^{222}A$ ratio from the fallowing;

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

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

RESULTS AND DISCUSSION

The activity of Radon ^{222}A and thoron ^{220}A together with their ratios are listed in Tables-1-11. Eleven locations in the plant (from L1 to L11), have been tested for Radon concentration, and from each location 10 samples have been taken. In table-1 shows the average Radon concentration in Central Gas Separation Station in North Rumaila oil field, which is equal to 37800 Bq/m^3 with maximum at stations S1 and S2, which is equal to 65809 Bq/m^3. It must be noted that, this concentration is high compared with the recommended working levels. However, these samples are originated from depth of 3000m to 4500m from the ground surface, and due to the geological structure of the oil wells, the uranium and thorium and their progenies are highly concentrated in these depths. According to EPA and CEC [12], radon concentration of 1Bq/m^3 equivalent to the effective dose of 0.025mSv y^{-1}.

Table-1 Radon gas concentration in sludge samples from the Central Gas Separation Station of the Southern- Rumaila oil field (L1)

Station No.	Station 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	1st stage B	4.54242	2.79034	0.17751	65809
S2	Entrance site C	1.36273	8.43591	0.41017	15906
S3	1st stage C	5.19133	3.24458	0.92695	42324
S4	Beginning of C	4.28285	2.66055	0.55759	44509
S5	2nd Stage C	4.54244	2.79034	0.17751	65809
S6	3rd Stage C	4.67220	2.92012	0.92695	38091
S7	2nd Stage B	2.31977	1.43986	0.51878	24821
S8	Near Castor D2	3.24458	2.01164	0.47085	36044
S9	Near Sechemer A	2.07979	1.27988	0.22280	28780
S10	1st Stage B –V2	1.36272	8.43591	0.41017	15906

Table-2. Radon gas concentration in sludge samples from the Degassing Station in the Southern-Rumaila oil field (L2)

Station No.	Station 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	Third Stage Bank B	0.63994	3.99961	0.92695	5217
S2	Dehydrator	5.84024	3.63393	0.69571	57206
S3	First stage Bank B	1.91981	1.19988	0.92695	15651
S4	First stage Bank C	0.39991	0.17777	0.12376	9173
S5	Desolate	2.27120	1.47616	1.15260	13721
S6	Second Stage Bank B	9.08483	5.58069	0.17751	131618
S7	Second stage Bank A	1.94675	1.23294	3.54484	6279
S8	Second stage Bank A	2.92012	1.81696	0.92695	21162
S9	Third Stage Bank A	2.07979	1.27988	0.22281	28780
S10	Desolater(2)	2.92012	1.81696	0.63957	28603

Table-3.The radon gas concentration in the sludge samples from Qurenit Degassing Station of the southern Rumaila oil field (L3)

Station No.	Station 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	Tank D1	3.37436	2.07653	0.22280	46694
S2	Tank D1 Desolater	no tracks	no tracks	-	-
S3	Tank Bank 2	3.56904	2.20631	0.35987	43485
S4	First stage Bank B En	no tracks	no tracks	-	-
S5	Scammer under flange	4.52415	2.85523	1.55260	27441
S6	First stage Bank A	0.79992	0.47995	0.17770	18345
S7	Second Stage Bank B	2.55975	1.59984	0.92695	20869
S8	First stage Bank C	2.55975	1.35991	1.79321	11696.
S9	First stage Bank C(2)	2.07970	1.27988	0.22280	28780
S10	Dehydrator	2.85523	1.75207	0.15207	42461

Table-4. The radon gas concentration in the sludge samples from Shamei Degassing station in the of southern Rumaila (L4)

Station No.	Station 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	First Stage Bank C	1.35986	0.79993	0.31950	40646
S2	Point Near tank 2	6.81362	4.21795	0.41017	79529
S3	Scammer Tank	2.59567	1.62229	0.92695	21162
S4	2nd Stage Bank C	2.40100	1.49251	0.58966	24370
S5	Dehydrator	3.24461	2.01164	0.47089	36044
S6	Out Side station	2.92012	1.81696	0.63951	8603
S7	Desolater	2.92012	1.81697	0.63951	8603
S8	West nearby	3.51966	2.15979	0.15261	52342
S9	West near Fence	2.71973	1.67893	0.33086	33977
S10	West near the door	1.03989	0.63994	0.22281	14390

Table-5. The radon gas concentration in the Sludge samples from Ratka Degassing Station in the southern Rumaila oil field (L5).

Station No.	Station 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	First stage Bank D1	4.2179571	2.5956659	0.2228030	58367
S2	Second Stage D1	2.2712077	1.4276162	1.526029	13721
S3	D1 Desolater	1.3627246	8.4359141	0.4101703	15905
S4	AL Scammer Tank	1.1680496	7.1380811	6.688020	19114
S5	First stage Bank	3.2445823	2.0116412	0.4708539	36044
S6	2nd Stage Bank C	no tracks	no tracks	-	-
S7	2nd Stage Bank C2	2.55975	1.59984	0.926950	20869
S8	3rd Stage Bank C	2.31977	1.43986	0.51878	24824
S9	West from ground	1.43987	0.87991	0.06689	23562
S10	Old west in Baril	3.51966	2.15979	0.15261	52342

Table-6. The radon gas concentration in the Sludge samples from Central Degassing Station in the Northern Rumaila oil field (L6).

Station No.	Station 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	Dehydrator	3.56904	2.20631	0.35986	43485
S2	First Bank A	1.16050	0.71381	0.06689	19115
S3	2nd Stage Bank A	1.75981	1.11999	8.95662	2523
S4	3rd Stage Bank A	1.27987	0.79992	0.92695	10434
S5	4th Stage Bank A	1.91981	1.19988	0.92695	15651
S6	1st stage Bank B	1.16050	0.713808	0.06689	19115
S7	2nd Stage Bank B	0.63993	0.39996	0.92673	5217
S8	3rd Stage Bank B	0.79991	0.47995	0.17770	18345
S9	4th Stage Bank B	1.19988	0.71993	0.17770	27518
S10	1ststageB2Bank	1.03989	0.63994	0.22280	14390

This means that; $1Bq/m^3=2.854x10^{-3}\mu Sv\ h^{-1}$, in other word, maximum radon concentration $65809Bq/m^3$ equivalent to 174 $\mu Sv\ h^{-1}$.If we consider that the maximum exposure rate in the USA, is 50 $\mu Sv\ h^{-1}$for crude oil equipments contaminated with NORM [13], then this result highly exceeded the allowed level. Rabee et al.[14], reported that the limit of the effective dose for exposed workers shall be 100mSv in a consecutive five years period, which is the same limitation of EPA mentioned above. Table-12, contain the radon concentration in location L2, with average 31741Bq/m^3les than location (L1), but this location contains the highest radioactive station (131618Bq/m^3) in all fields of study. This Radon concentration is equivalent to 376 µSv h^{-1}, effective dose, which in very high.

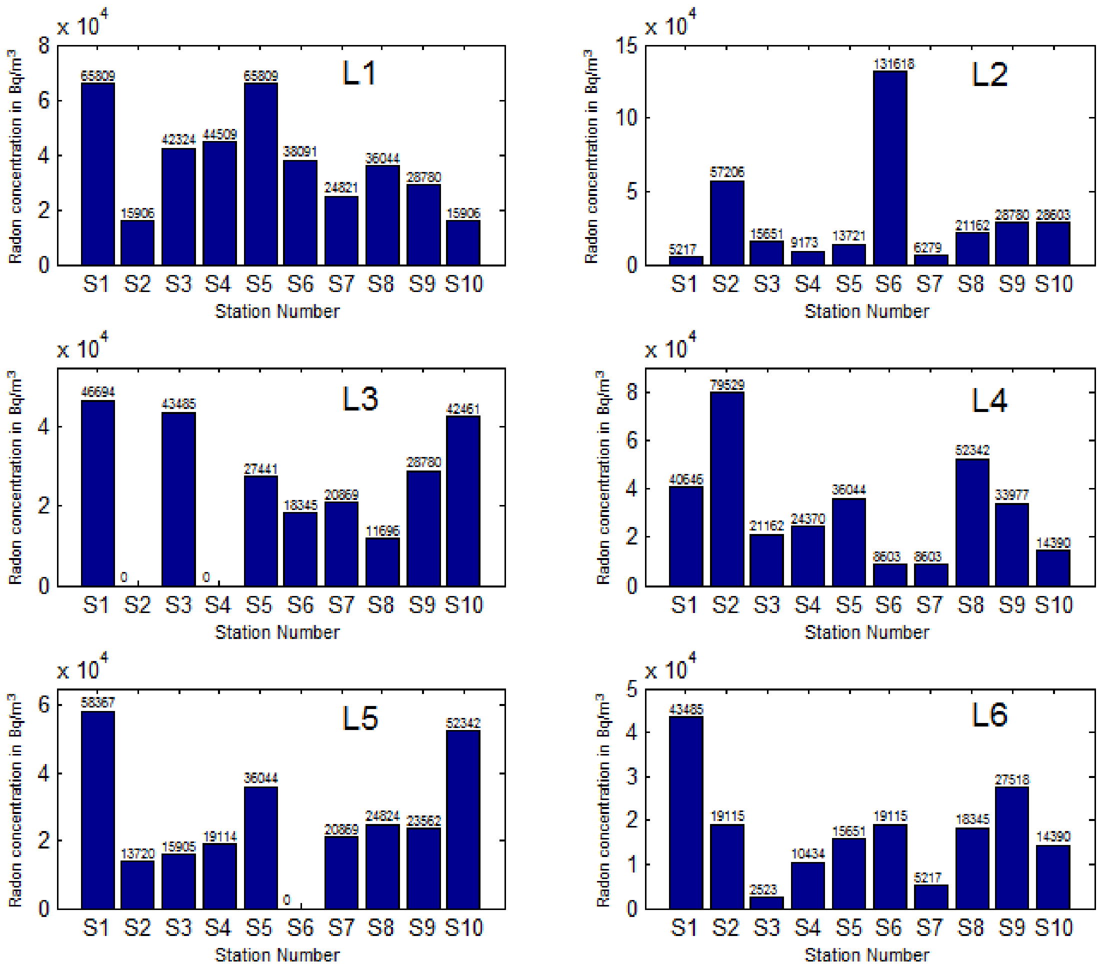

Figure -2.The Radon concentrations (Bq/m^3) in six locations from L1 to L6, as a function of station number

Table-7. The radon gas concentration in the sludge samples from Second Degassing Station in the Northern Rumaila oil field (L7).

Station No.	Station 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	First stage Bank	1.36272	0.84359	0.41017	15906
S2	2nd stage Bank A	2.07653	1.29783	0.92569	16929
S3	3rd stage Bank A	1.16805	0.71380	0.06889	19114
S4	1st stage Bank B	1.36272	0.84351	0.41017	15906
S5	2nd Stage Bank B	2.27121	1.42761	1.52602	13721
S6	3rd Stage Bank B	1.36272	0.84359	0.41017	15906
S7	Tank2	1.03989	0.63994	0.22280	14390
S8	Tank2	0.80000	0.47995	0.17770	18345
S9	Wells Assembly	0.63993	0.39996	0.92695	5217
S10	Old Scammer sys.	6.03492	3.76372	0.77472	53997

Table-8. The radon concentration in the sludge samples from the Degassing Station in the Northern Rumaila oil field (L8).

Station No.	Station 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	1st stage Bank A	3.24458	2.01164	0.47085	36044
S2	2nd stage Bank A	2.59567	1.62229	0.92695	21162
S3	3rd Stage Bank A	2.07653	1.29783	0.92567	16929
S4	1st stage Bank B	2.27121	1.42761	1.52605	13721
S5	2nd Stage Bank B	3.56904	2.20616	0.35987	43485
S6	3rd Stage Bank B	4.52416	2.85523	1.55261	27442
S7	1st stage Bank C	1.36272	8.43591	0.41017	15906
S8	2nd Stage Bank	2.92012	1.81696	0.92695	21162
S9	2nd Bank A	2.40099	1.49250	0.58966	24378
S10	Tank No.1	No Tracks	No tracks		

Table-9. The radon concentration in the sludge samples from the Fourth Degassing Station in the Northern Rumaila oil field (L9)

Station No.	Station 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		3.89350	2.40099	0..28132	50926
S2	Dehydrator	3.24458	2.01164	0.47085	36044
S3	2nd Stage Bank B	1.16805	7.13808	0.06889	19114
S4	1st stage Bank B	1.43986	0.87991	0.06889	23562
S5	1st stage Bank A	1.16805	7.13808	0.06889	19114
S6	2nd stage Bank A	1.43986	0.87991	0.06781	23562
S7	3rd Stage Bank A	1.43986	0.87991	0.06781	23562
S8	3rd Stage Bank B	1.36272	0.84359	0.41017	15906
S9	Wells Assembly	1.36272	0.84359	0.41017	15906
S10	Dehydrator	2.31977	1.43986	0.51878	24824

Table-10. The radon concentration in the sludge samples from the Fifth Degassing Station in the Northern Rumaila oil field (L10)

Station No.	Station 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	1st stage Bank A	1.51985	0.95990	2.4908	6479
S2	2nd stage Bank	1.439873	0.87981	0.06778	23562
S3	3rd stage Bank A	2.27120	1.42761	1.52603	13720
S4	1st Bank B	1.36272	0.84359	0.41017	15906
S5	2nd Bank B	2.55975	1.59984	0.92695	20869
S6	3rd Stage Bank B	3.56904	2.20617	0.35987	43485
S7	Residual sludge	3.63393	2.27121	0.92695	29626
S8	Dehydrator	3.89350	2.40091	0.28132	50926
S9	Near st. out	5.71046	3.50415	0.15261	84923
S10	Desolater	3.89350	2.40091	0.15261	50926

The rest of the tables contain data of average Radon concentration in the rest of locations. Fuger-2 and 3, represent a summary of Radon concentration in six locations. One should notice that the minimum radon concentration in sludge is $2523 Bq/m^3$, which equivalent to effective dose 7.2 $\mu Sv\ h^{-1}$. Even though this is not high dose but based on the ALARA Principle [15], it is to reduce effective dose and minimize the radiation exposure.

Table-11. The radon concentration in the sludge samples from Central Pump Station-1 in the Northern Rumaila oil field (L11)

Station No.	Station 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	Point under tank 1	1.03826	6.48916	0.92695	8465
S2	Tank No.2	1.36272	0.84359	0.41017	15906
S3	Sludge accumulation area	1.36272	0.84359	0.41017	15906
S4	Input tank No.1	1.03826	6.48916	0.92695	8465
S5	Tank No.3	0.47991	0.23991	0.61671	3384
S6	Tank No.4	0.40000	0.23991	0.1777	9172
S7	Tank No.5	0.47991	0.23991	0.61671	3384
S8	Tank No.6	0.06399	0.03100	0.92695	5217

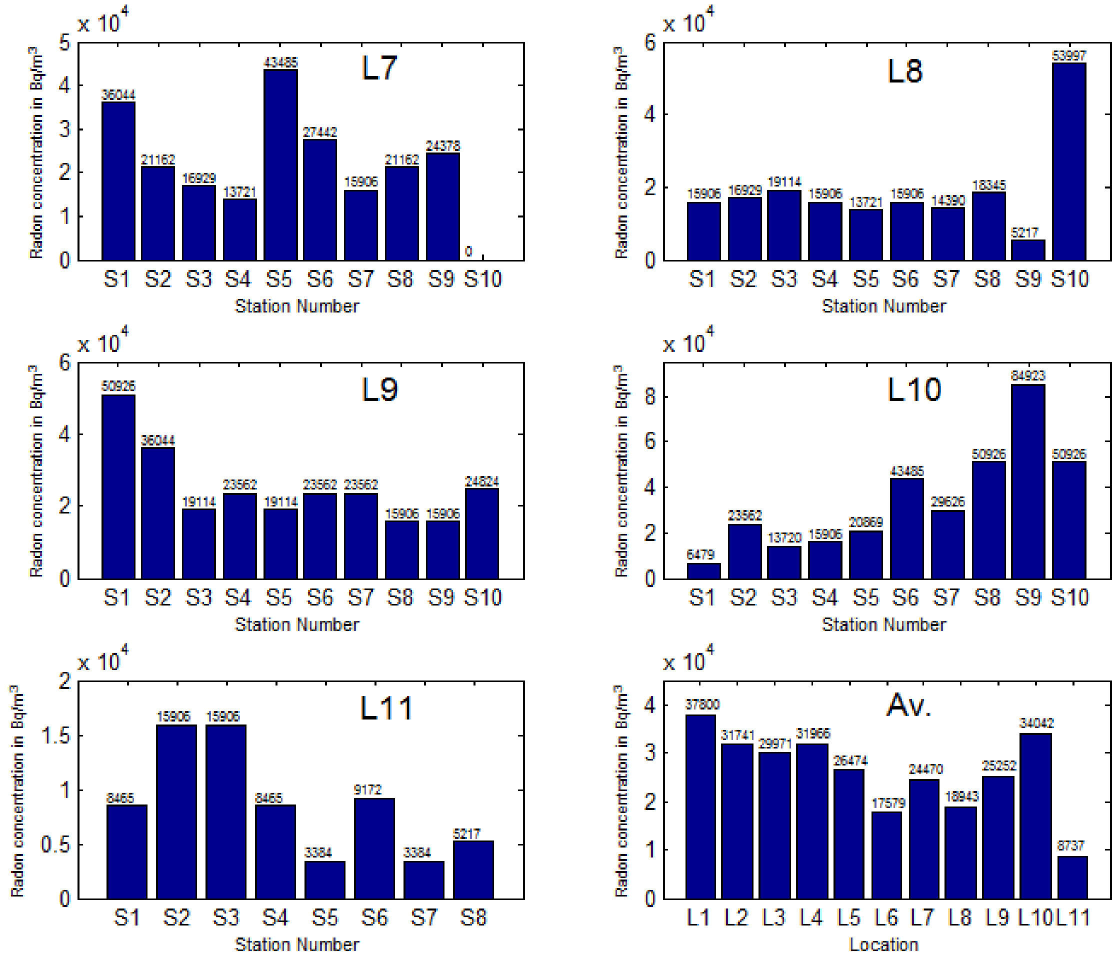

Figure -3.The Radon concentrations (Bq/m^3) in five locations from L7 to L11, as a function of station number, and the average radon concentration in each location as a function of location number.

Talking about the arithmetic average of radon concentration for each location, we can see it is always high except location L11. The average radon concentration for all locations is 26089 Bq/m^3.

Table-12. The average Radon concentrations in the eleven locations in Basrah plant oil fields

location	Average Radon concentration Bq/m^3
L1	37800
L2	31741
L3	29971
L4	31966
L5	26474
L6	17579
L7	24470
L8	18943
L9	25252
L10	34042
L11	8737

CONCLUSION

In conclusion, one can report that; we have tested 108 samples, 78 samples with concentration less than 30000Bq/m^3(72%), 16 with concentration ≥30000Bq/m^3 and ≤ 50000Bq/m^3(15%) , 10 with concentration ≥50000Bq/m^3 and ≤ 70000Bq/m^3(9.2%), 3 with concentration ≥70000Bq/m^3 and ≤ 90000Bq/m^3 (2.8%) and finally one has concentration 131618Bq/m^3.Most of the radon concentration in sludge west are high. The arithmetic average for all samples was 26089Bq/m^3 (equivalent to 60 μSv h^{-1}) and this exceed the recommended limit of worker exposure and increases the probability of lung and stomach cancers. Cross[16], estimated the probability of getting cancer to be 1.6x10^{-3} for each 37Bq/m^3. In our average value, the probability increases to 705 multiple.

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