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Original Research Paper

NORM in Soil and Sludge Samples in Dukhan Oil Field, Qatar State

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The main objective of this work is to measure the activity concentrations of Naturally Occurring radioactive Materials (NORM) produced as by-products in oil production. The analysis of NORM gives available information for guidelines concerning radiation protection. Recently NORM was subjected to restricted regulation issued by high legal authority in Qatar state. Twenty five samples of soil from Dukhan onshore oil field and 10 sludge samples were collected from 2 offshore fields in Qatar state. High resolution low-level gamma-ray spectrometry is used to measure gamma emitters of NORM. The activity concentrations of natural radionuclide in 22 samples from Dukhan oil field, were with average worldwide values. Only three soil samples have high activity concentration of Ra-226 which is more than 185 Bq/kg, the exempted level for NORM in the Quatrain regulation. The natural radionuclide activity concentrations of 10 sludge samples from offshore oil fields was greater than 1100Bq/kg, the exempted values of NORM set by Quatrain regulation so the sludge need special treatments. The average hazards indices (Hex, D, and Raeq), for the 22 samples were below the word permissible values. This means that the human exposure to such material does not impose any radiation risk. The average hazard indices (Hex, D, and Raeq), for 3 soil samples and sludge samples were higer than the published maximum permissible values. Thus, human exposure to such material impose radiation risk.

Keywords: NORM, Gamma spectroscopy, By–products, Oil and gas industry, Risk, Radiation, Dukhan, Sampling.

INTRODUCTION

Series of naturally occurring radionuclides are found in varying concentrations in the Earth's crust depending on the geological formation. Various industrial processes of oil and gas extracting and processing operations lead to enhance the natural activity. This "enhanced'' NORM, is called TENORM by some states. (Technologically-Enhanced Naturally Occurring Radioactive Materials). Sludge, are examples of materials that can contain high levels of NORM, in equipment and materials, can be created when industrial activity increases the concentrations of uncontrolled activities such as leak of produced water or scale from pipe and valves can contaminate the soil and environment and pose a risk to human health.

Various industrial processes of oil and gas extracting and processing operations lead to enhance the natural activity. Since Ra-226 is slightly soluble, it is mobilized from subsurface formation in the liquid phases and transported to the surface in the produced water. As the produced water is brought to the surface, some of the dissolved radium precipitates come out in solid. The primary radionuclide of concern in oil and gas stream are Ra-226, Th-232, and K-40 which are responsible for most of the external exposure in such facilities (1).

Radium solubility and mobility depend on the salinity of the formation water; higher salinity is aligned with a greater solubility (2).

MATERIAL AND EXPERIMENTAL ARRANGEMENTS

Samples locations

Twenty five samples of soil from Dukhan at north west of the coast, which is a big onshore oil field in the state of Qatar is comprise of three reservoirs, the oldest one was AL- Khatiyah which started the oil production at 1947 till now. The other two sectors are Fahahil which started producing oil in 1954 followed by Jaleha in 1955.

The most important producers of oil in the state of Qatar are the offshore oil fields which are operated by QP, sharing with other international oil companies according to exploration and development sharing agreements (DPSA). Ten samples of sludge collected from separation tanks from two offshore, the first one from the field PS and the second location for sludge collection was from Al Shaheen field.

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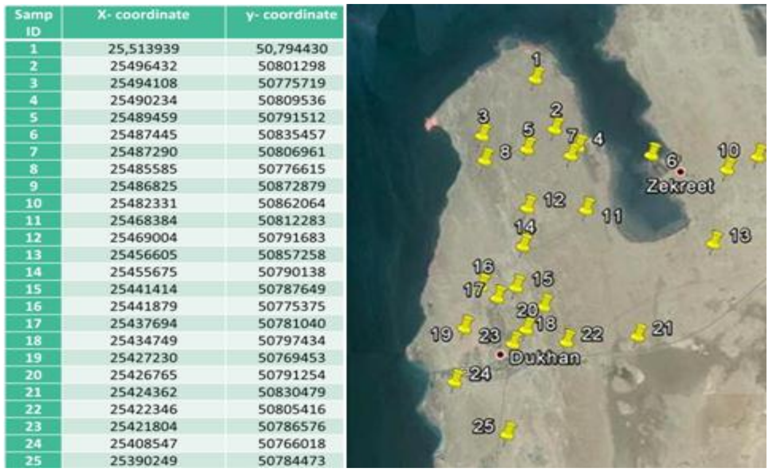

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8	25485585	50776615
9	25486825	50872879
10	25482331	50862064
11	25468384	50812283
12	25469004	50791683
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15	25441414	50787649
16	25441879	50775375
17	25437694	50781040
18	25434749	50797434
19	25427230	50769453
20	25426765	50791254
21	25424362	50830479
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25	25390249	50784473

FIG (1). Samples location on Qatar map

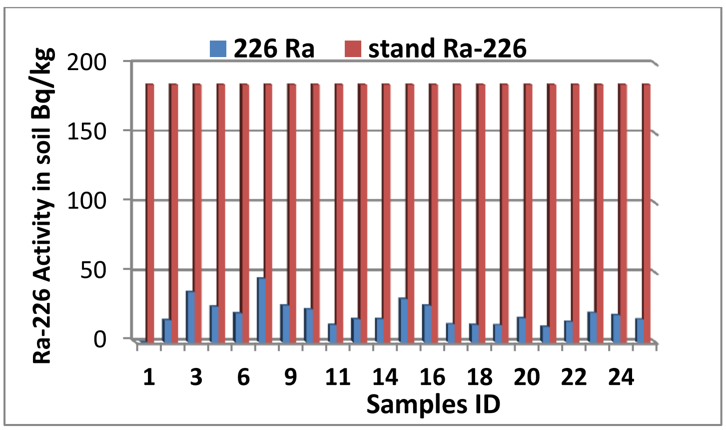

Fig (2). ^{226}Ra concentration of soil samples not contaminated with NORM at Dukhan oil field in Qatar

Samples preparation

Twenty five soil samples from Dukhan field were collected randomly near the oil well head as seen in Fig. 1 using Back-Back detector as radiation survey meters and tools for positioning. Sample of about 1 kg was taken after removal of stones and biological parts, then transferred into sealed labeled polyethylene bags.

The samples were dried to get rid of any significant moisture using oven set at 353 K for 12 hours. Each soil sample is packed and sealed in an airtight PVC Marinelli beaker. The beakers were stored for 30 days before its counting radioactivity to achieve the secular equilibrium between the daughter products of radon (222Ra), (220Ra) and their short lived decay products.

Gamma ray system calibration

Measurements were conducted by Gamma ray spectroscopy system equipped with Canberra n-type detector of high-purity germanium (HPGe). Each sample was counted for 24h to reduce the statistical uncertainty. Minimum detectable activity was determined from the background radiation spectrum
The detector has a resolution of 2.5 keV and relative efficiency of 30% for 1.332 MeV gamma energy of 60Co. The output of the detector is connected to PC. The spectral data was analyzed using the "Genie 2000 Gamma Analysis Software package". The detector had a graded shielding made of lead of 10 cm thick shield to reduce the background radiation level of the system, and lined inside with 1mm copper sheets to minimize the X-rays emitted due to interaction of cosmic radiation with lead.

The absolute photo-peak efficiency calibration of the system were carried out using standard source of 152Eu in a Marinelli beaker because of its suitable half-life and the wide range of gamma ray energies produced during its decay process. The specific radioactivity of 226Ra under the peak energy of 186.21 keV is the sum of 235U under the peak energy of 185.7 keV and peak energy of 226Ra alone. Thus the radioactivity of 226Ra alone calculated by subtracting the specific radioactivity of 235U which is calculated from the peak energy of 143.76 keV from the total specific radioactivity calculated for 226Ra. can be measured from the weighted mean of the activity concentration of Pb-214 and Bi-214. 232Th activities were determined from the average concentrations of 238.6 keV peak of its 212Pb progeny and 911.1keV peak of its 228Ac progeny or by 208Tl gamma-ray emission probability corrected for 212Bi α decay branching ratio of 35.94 %. Activities of 40K were calculated from the 1,460.7 keV peak.

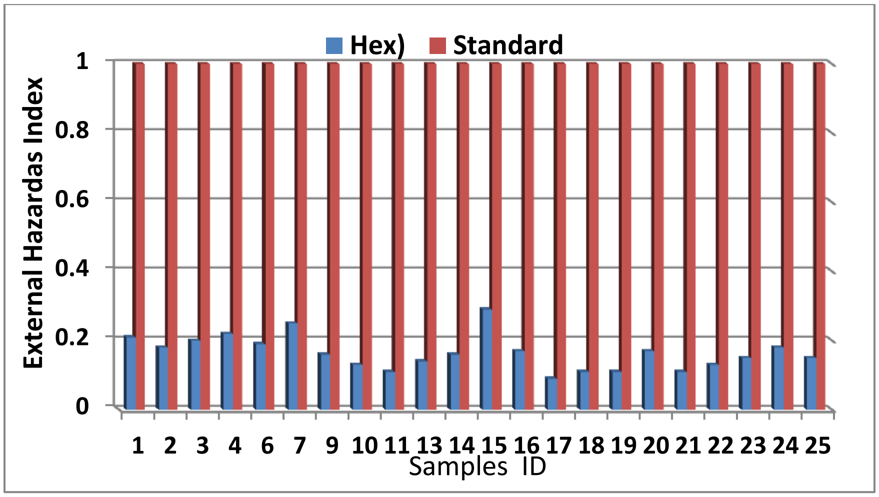

Fig (3). External hazard index H_{ex} of soil samples not contaminated with NORM at Dukhan oil field in Qatar

Table 1. Radioactivity concentrations of ^{226}Ra , ^{232}Th, and ^{40}K in Bq/kg, and risk indices in soil samples at Dukhan oil field in Qatar.

Sam Id	^{40}K	^{232}Th	^{226}Ra	H_{ex}	D	Ra_{eq}
1	284.20	24.80	21. 3	0.21	25.82	48.04
2	190.80	46.60	15.70	0.18	44.17	95.69
3	167.05	17.69	35.90	0.20	34.49	72.48
4	261.59	24.49	25.49	0.22	37.83	78.82
6	234.86	22.65	20.85	0.19	33.40	69.68
7	252.95	18.53	45.64	0.25	43.05	89.84
9	206.02	10.67	26.40	0.16	27.34	55.68
10	198.79	6.78	23.69	0.13	23.37	47.30
11	174.39	10.69	12.32	0.11	19.56	39.81
13	249.63	10.82	16.44	0.14	24.65	49.39
14	218.21	17.14	16.59	0.16	27.36	56.37
15	211.89	20.61	30.94	0.29	35.87	75.24
16	229.58	12.54	26.12	0.17	29.36	60.12
17	135.56	7.77	12.79	0.09	19.48	33.39
18	127.74	13.35	12.34	0.11	19.29	40.37
19	192.39	9.83	12.09	0.11	19.66	39.61
20	255.19	19.14	17.26	0.17	30.44	62.49
21	179.52	11.09	10.87	0.11	19.35	39.29
22	222.16	12.48	14.68	0.13	23.74	48.08
23	254.19	10.69	21.05	0.15	26.89	54.13
24	272.70	19.35	19.25	0.18	32.22	66.00
25	247.90	13.80	16.29	0.15	26.37	53.37
Average	216.69	16.43	20.05	0.16	26.04	53.29
5	**112.50**	**8.80**	**226.70**	**0.67**	**114.65**	**247.16**
8	**109.90**	**6.70**	**289.50**	**0.83**	**142.18**	**306.77**
12	**120.80**	**9.80**	**198.30**	**0.599**	**102.52**	**220.70**

Exposure to radiation sources and risk assessment

Natural radionuclides232Th, 226Ra and40K in terrestrial soils or sludge, decay to its progeny, which produce external radiation exposure to human beings due to gamma-beta radiation.. The exposure rate to individual depend on the concentrations of radionuclides in the sample. The main objective of external hazard index Hex is to limit the radiation dose to the admissible annual dose equivalent limit of 1 mSv/y (3). Beretka and Mathew in 1985 (4) drive the equation:

$$H_{ex} = \{ (C_{Ra}/370) + (C_{Th}/259) + (C_K/4810) \} \leq 1 \quad ..... 1$$

Where:

C_{Ra}, C_{Th} and C_K are the activity concentrations in Bq/kg of ^{226}Ra, ^{232}Th and ^{40}K in Bq/ kg, respectively. The values of this index must be less than unity in order to keep the radiation hazard without posing any significant radiological threat to public. The Radium equivalents (Ra_{eq}) in Bq/ kg are used to assess the radiological hazards associated with materials that contain natural radioactivity in Bq/kg (5). Yu et al. 1992 (6) using the following equation to calculate Ra_{eq}:

$$Ra_{eq} = C_{Ra} + 1.43\, C_{Th} + 0.077\, C_K \quad ----- 2$$

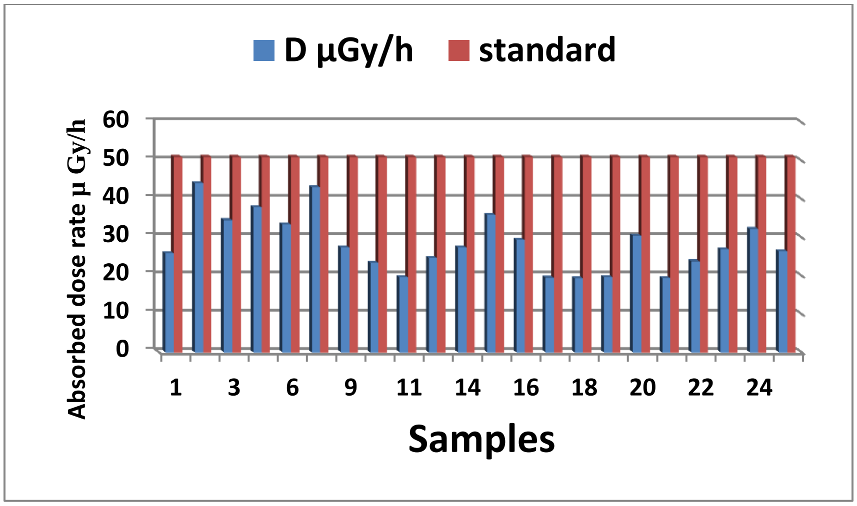

Fig (4).

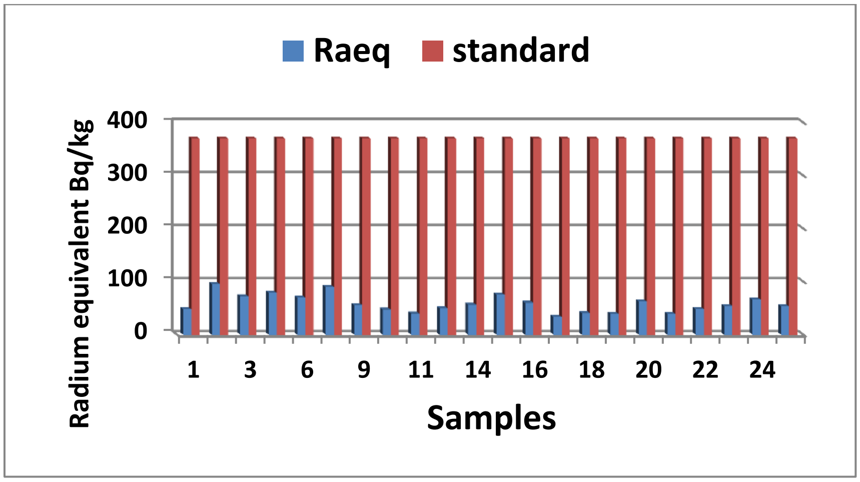

Fig (5). Radium Equivalent of soil samples not contaminated with NORM

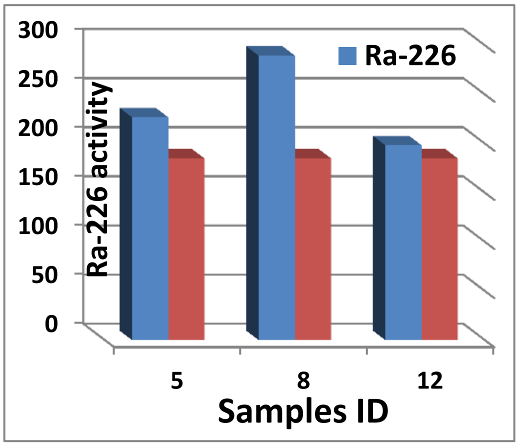

Fig (6). ^{226}Ra concentration of soil samples

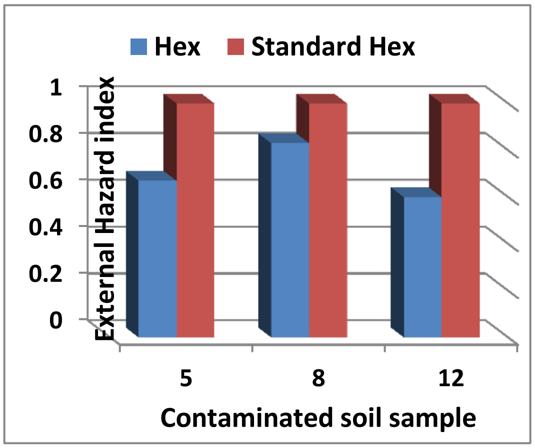

Fig (7). H_{ex} of contaminated soil

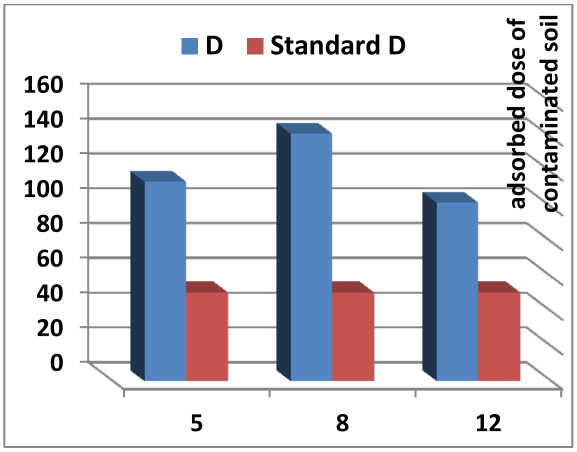

Fig (8). Absorbed dose rate of contaminated

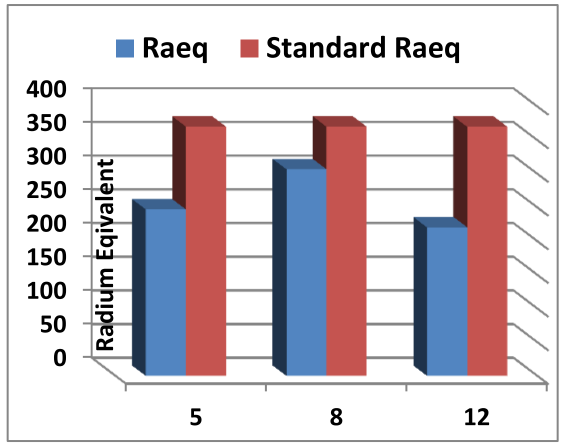

Fig (9). Radium equivalent of soil

Table 2. Radionuclide concentration of NORM (Bq/kg) in sludge samples

Sample ID	226Ra	232Th	40K	H_{ex}	D	Ra_{eq}
1	394.49	74.57	62.8	1.37	230.92	505.52
2	1189.3	30.6	110.6	3.36	571.91	1240.8
3	1253.98	46.75	107.34	5.21	611.65	1328.34
4	27884.9	94.07	98.6	75.74	12917.63	28026.32
5	13983.4	53.99	102.7	38.02	6484.23	14067.79
6	16080.3	79.8	106.8	47.8	7467.16	16201.89
7	20827.5	64.43	97.9	56.55	9645.67	20926.49
8	25805.84	85.48	86.5	70.1	11953.32	25934.13
9	18289.9	54.6	99.8	49.66	8469.79	18374.96
10	20065.7	67.2	100.5	54.51	9296.31	20168.83

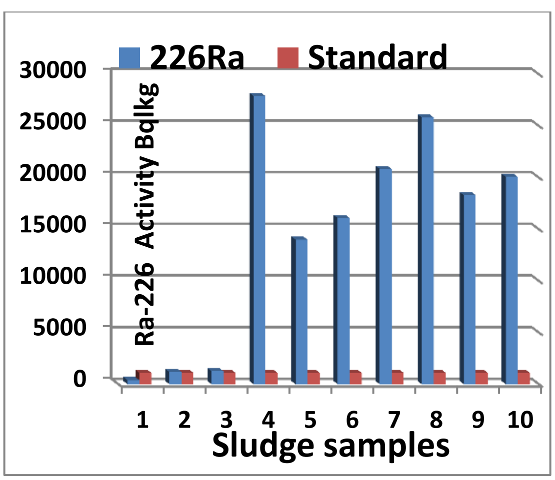

Fig (10). ^{226}Ra concentration of sludge

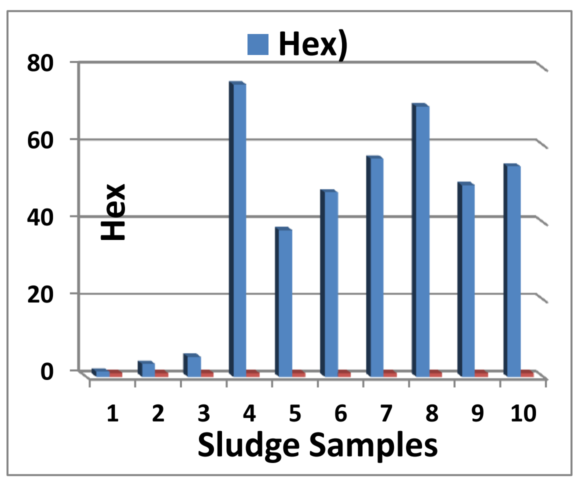

Fig (11). External Hazards of sludge

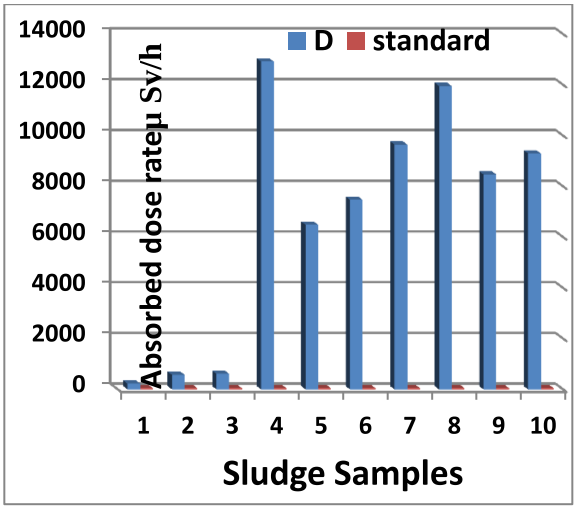

Fig (12). Absorbed dose of sludge

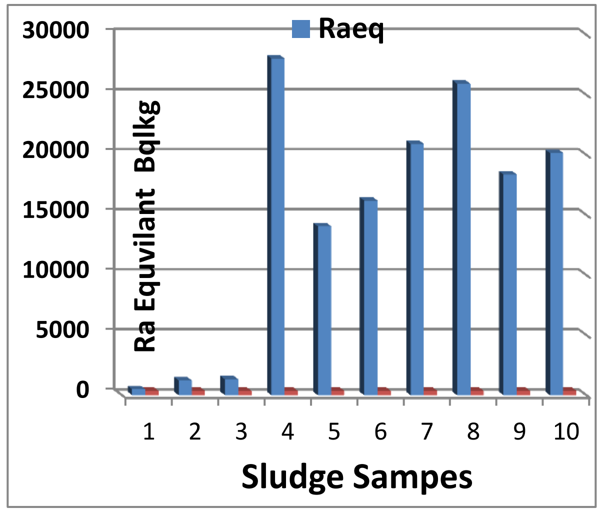

Fig (13). Radium equivalent of sludge

Maximum value of Ra_{eq} must be less than 370 Bq/kg that is equivalent to the annual dose equivalent of 1.5 mSv/y.
Radium equivalent activity defined with the assumption that 1Bq/kg of 226Ra, 0.7Bq/kg of 232Th, and 13Bq/kg of 40K produce the same radiation dose rates. The total external terrestrial Gamma radiation absorbed dose rate (D) in air is the quantity that cocidering radiation risk due to gamma rays emitted by the ^{238}U , ^{232}Th decay chain and ^{40}K at 1m above the ground level. The published maximal permissible dose rate is 51 nGy/h. Rohit Mehra et al (7) use the following equation to calculate the absorbed dose rate (D) in air:

D(nGy/h) =0.461 CRa + 0.623CTh + 0.0414CK 3

RESULT AND DISCUSSION

The activity concentrations of natural radionuclide Ra-226, Th-232, and K-40, (H_{ex}), (Ra_{eq}), and (D) of the 25 samples from Dukhan oil field were calculated for each sample and summarized in Table 1 and Fig (2). 22 of the investigated soil samples which have normal activity of Ra-226, and shows that the average activity concentration of Ra-226, Th-232, and K-40, were 20.05, 16.43, and 216.69 Bq/kg, respectively, which is below the world wide averaged value of Ra-226, Th-232, and K-40 in normal soil which are 33, 45, and 420 Bq/kg respectively cited by UNSCEAR (5)

The average H_{ex} for the 22 samples were less than unity Fig (3). The average value of (D) for 22 investigated soil samples was found to be 26.04 nGy/h which is less than the published maximal permissible Fig (4). The average value (Ra_{eq}) was 53.29 which is less than the published maximal permissible value Fig (5) 370 Bq/kg (5). All data for the 22 samples indicates that the soil sample are normal and represent the a normal level of natural background radiation of soil at that area.

Three soil samples (NO 5, 8, and 12) has high activity concentration of Ra-226 which is more than 185 Bq/kg, the exempted level for NORM in the Quatrain regulation (Fig 6) recently issued by the legal authorization. It is likely that the high radioactivity of Ra-226 for these samples is as a result of contamination of this soil by NORM these may be contaminated by produced water or scale from oil well.

The Ra-226 concentration was more than 185 Bq/kg, this is agrees with Al-Sulaiti et al finding of the same area (8).

The external hazard index, (Hex) was less than the world permissible value of unity (Fig 7). This indicates that the values will not lead to respiratory cancer and external other radiation diseases such as erythema, and cataracts.

The measured value of Radium equivalent ($R_{aeq)}$ of the three contaminated samples were 247.16, 306.77, and 220.7 Bq/kg, which are less than the internationally safe limit 370 Bq/kg. and (D) also less than published maximum permissible value 51 nGy/h.

Table 2 shows the activity concentrations Bq/kg of natural radionuclide ^{226}Ra, ^{232}Th, and ^{40}K of the 10 sludge samples from offshore oil fields. The calculated H_{ex}, Ra_{eq} (Bq/kg) and the absorbed dose D (nGy/h) in each of these samples are also shown.

The Ra-226 activity concentrations of 9 of these samples were greater than 1100Bq/kg, the exempted values set by Quatrain regulation Fig 10. The (Hex) values were more than unity Fig 11, (Raeq) and D was more than the published maximum permissible value by many factors, Fig 12, 13 which means that the human exposure to such material impose high risk, thus the sledges must be stored in safe storage for final disposal

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