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Norm in soil and sludge samples in Dukhan oil Field, Qatar state

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ABSTRACT

The main objective of this work is to measure the activity concentrations of Naturally Occurring radioactive Materials (NORM) produced as a buy products in oil production. The analyses of NORM give available information for guidelines concerning radiation protection. Recently NORM subjected to restricted regulation issued by high legal authority at Qatar state. Twenty five samples of soil from Dukhan onshore oil field and 10 sludge samples collected from 2 offshore fields at Qatar state. High resolution low-level gamma-ray spectrometry 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 (H_{ex} , D , and Ra_{eq}), for the 22 samples were below the word permissible values .This means that the human exposure to such material not impose any radiation risk. The average hazards indices (H_{ex} , D , and Ra_{eq}),for 3 soil samples and sludge samples are higer than the published maximal permissible. Thus human exposure to such material impose radiation risk.

Keywords:

1. INTRODUCTION

Series of naturally occurring radionuclides are found at 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, some state called this enhancement as TENORM. (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 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 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 greater solubility (2).

2. MATERIAL AND EXPERIMENTAL ARRANGEMENTS

2.1. SAMPLES LOCATIONS

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

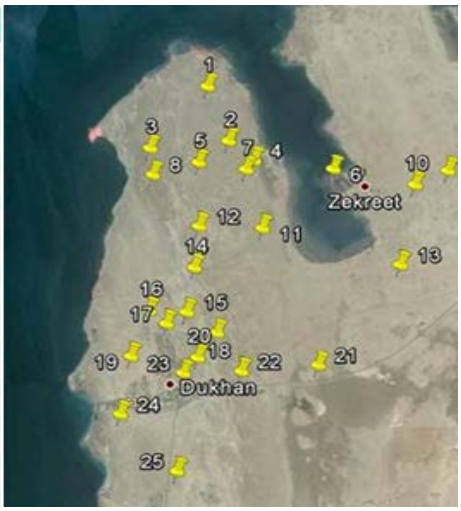
The most important production of oil in the state of Qatar are the offshore oil fields which are operated by QP, sharing with other international oil companies according to exploration and development sharing agreements (DPSA).

Ten samples of sludge collected from separation tanks from two offshore the first one the field PS and the second location for sludge collection was from Al Shaheen field. **samples preparation**

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

Figure 1: Samples location on Qatar map

Samp ID	X- coordinate	y- coordinate
1	25,513939	50,794430
2	25496432	50801298
3	25494108	50775719
4	25490234	50809536
5	25489459	50791512
6	25487445	50835457
7	25487290	50806961
8	25485585	50776615
9	25486825	50872879
10	25482331	50862064
11	25468384	50812283
12	25469004	50791683
13	25456605	50857258
14	25455675	50790138
15	25441414	50787649
16	25441879	50775375
17	25437694	50781040
18	25434749	50797434
19	25427230	50769453
20	25426765	50791254
21	25424362	50830479
22	25422346	50805416
23	25421804	50786576
24	25408547	50766018
25	25390249	50784473



The samples were dried to get rid from 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 was stored for 30 days before its counting radioactivity to achieve the secular equilibrium between the daughter products of radon (^{222}Ra), (^{220}Ra) and their short lived decay products.

2.2. GAMMA RAY SYSTEM CALIBRATION

Measurements were conducts 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 ^{60}Co. The output of the detector is connected to PC. The spectral data analyzed using the "Genie 2000 Gamma Analysis Software package". The detector was has a graded shielding made of lead of 10 cm thick shield to reduce the background radiation level of the system, and lined inside with 1 mm 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 ^{152}Eu in 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 ^{226}Ra under the peak energy of 186.21 keV is the sum of ^{235}U under the peak energy of 185.7 keV and peak energy of ^{226}Ra alone. Thus the radioactivity of ^{226}Ra alone calculated by subtracting the specific radioactivity of ^{235}U which is calculated from the peak energy of 143.76 keV from the total specific radioactivity calculated for ^{226}Ra. can be measured from the weighted mean of the activity concentration of Pb-214 and Bi-214. ^{232}Th activities were determined from the average concentrations of 238.6 keV peak of its ^{212}Pb progeny and 911.1keV peak of its ^{228}Ac progeny or by ^{208}TI gamma-ray emission probability corrected for ^{212}Bi α decay branching ratio of 35.94 %. Activities of ^{40}K were calculated from the 1,4607 keV peak.

2.3. EXPOSURE TO RADIATION SOURCES AND RISK ASSESSMENT

Naturally radionuclides ^{232}Th, ^{226}Ra and^{40}K in terrestrial soils or sludge decay to its progeny, which produce external radiation exposure 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 H_{ex} is to limit the radiation dose to the admissible annual dose equivalent limit of 1 mSv/y (3). Beretka and Mathew at 1985 (4) drive the equation:

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

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 equivalent (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}:

$$\mathbf{Ra_{eq} = C_{Ra} + 1.43\ C_{Th} + 0.077\ C_K} \qquad \textbf{eq 2}$$

Maximum value of Ra_{eq} must be less than 370 Bq/kg that equivalent to the annual dose equivalent of 1.5 mSv/y. Radium equivalent activity defined on the assumption that 1 Bq/kg of ^{226}Ra, 0.7 Bq/kg of ^{232}Th, and 13 Bq/kg of ^{40}K 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:

$$\mathbf{D(\ nGy/h) = 0.461\ CRa + 0.623CTh + 0.0414CK} \qquad \mathbf{eq\ 3}$$

3. RESULTS AND DISCUSSIONS

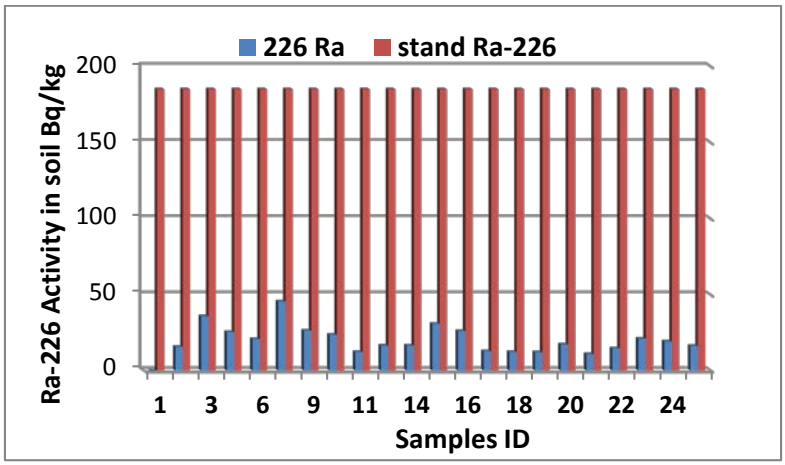
The activity concentrations of natural radionuclide Ra-226,Th-232,and K-40, (Hex), (Raeq), and (D) of the 25 samples from Dukhan oil field were calculated for each samples are summarized in Table 1 and Figure 2. 22 of investigated soil samples which have normal activity of Ra-226, and shows that the 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).

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**

Figure 2: ^{226}Ra concentration of soil samples not contaminated with NORM at Dukhan oil field in Qatar



The average Hex for the 22 samples were less than unity Figure 3 .The average value the of (D) for 22 investigated soil samples was found to be 26.04 nGy/h which is less than the published maximal permissible, Figure 4. The average value (Raeq) was 53.29 which is less than the published maximal permissible value, Figure 5, 370 Bq/kg (5).

Figure 3: External hazard index H_{ex} of soil samples not contaminated with NORM at Dukhan oil field in Qatar

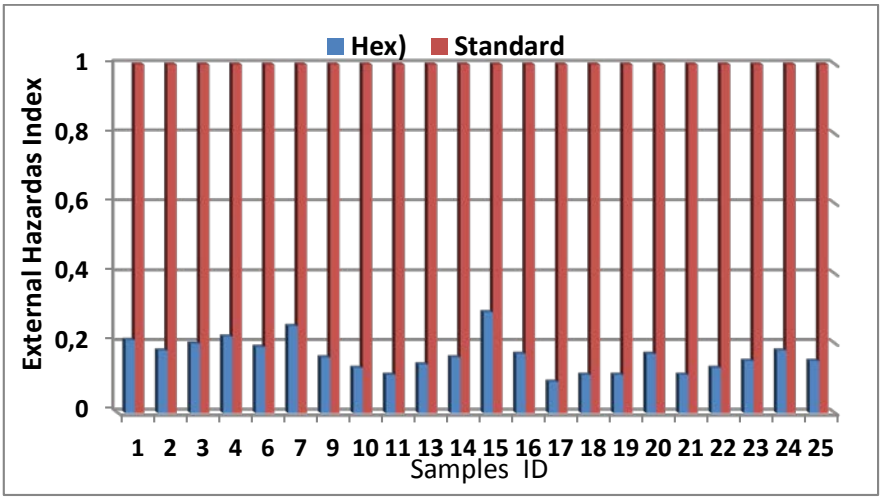


Figure 4: The average absorbed dose rate of the soil sample

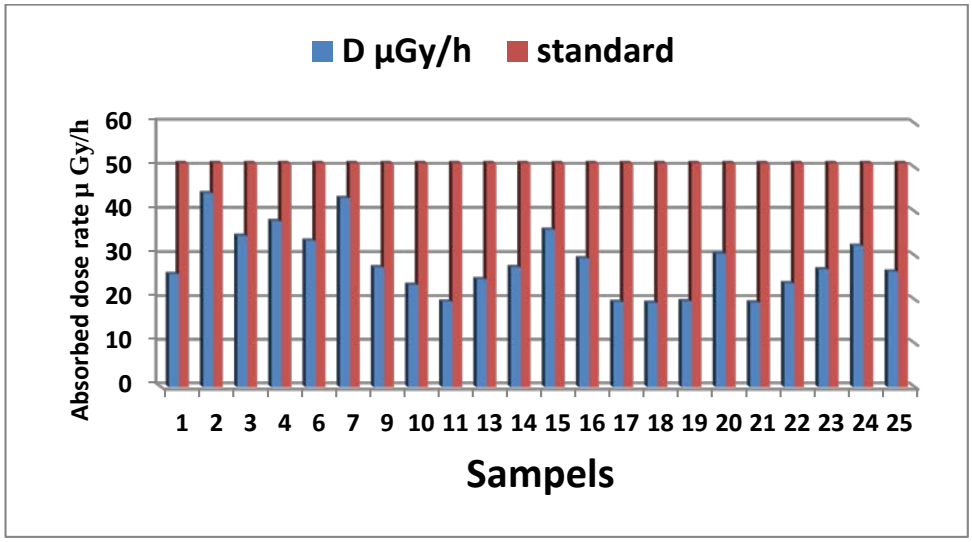
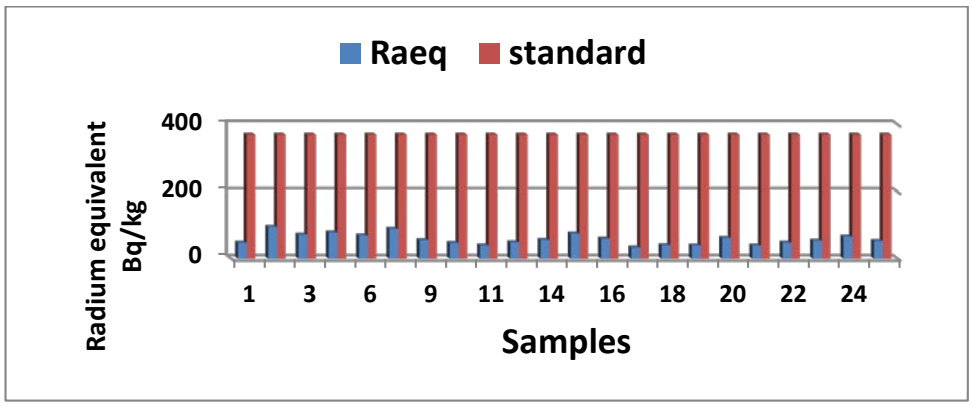


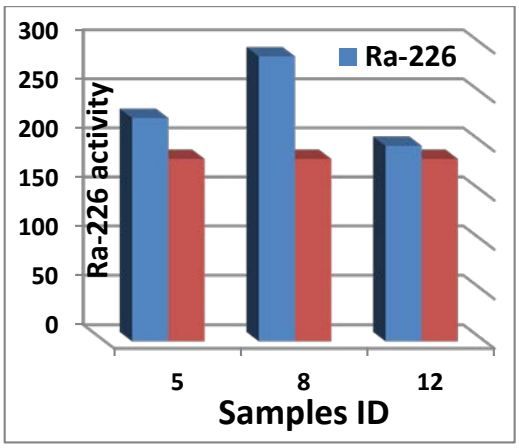
Figure 5: Radium Equivalent of soil samples not contaminated with NORM



All data for the 22 samples indicate that the soil sample are normal and represent the a normal level of natural background radiation of soil at that area.

Three soil sample (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, Figure 6, recently issue 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 many be contaminated by produced water or scale from oil well.

Figure 6: ^{226}Ra concentration of soil samples



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

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

Figure 7: Hex of contaminated soil

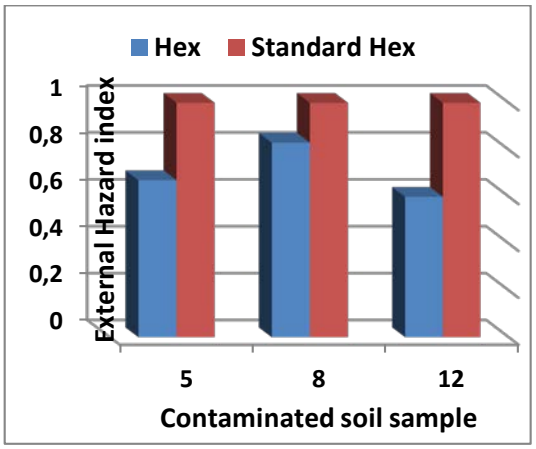
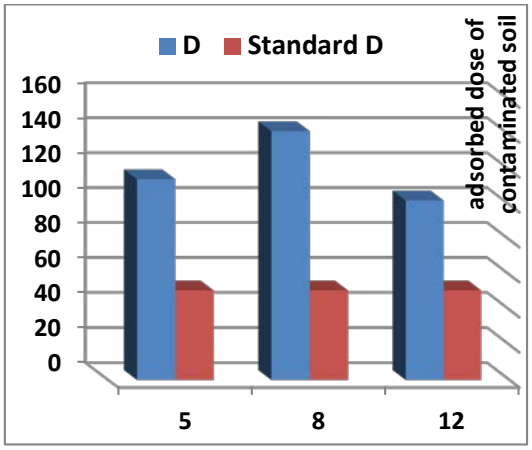


Figure 8: Absorbed dose rate of contaminated soil



The measured value of Radium equivalent (Raeq) 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 maximal 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 Hex, Raeq (Bq/kg) and the absorbed dose D (nGy/h) in each of these samples are also shown.

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

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 was more than unity Fig 11, (Raeq) and D were more than the published maximal 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 store in safe storage for final disposal.

Figure 9: Radium equivalent of soil

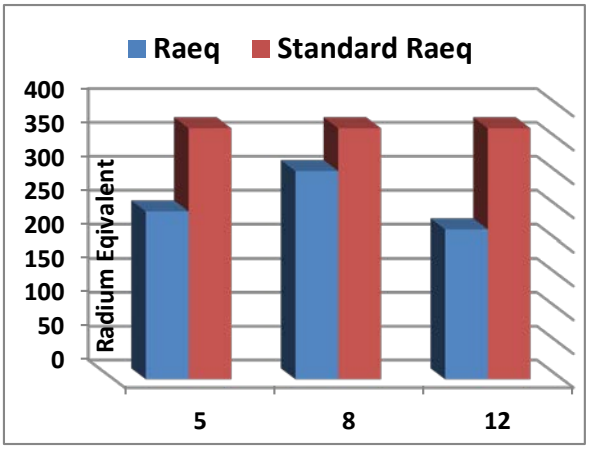


Figure 10: ^{226}Ra concentration of sludge

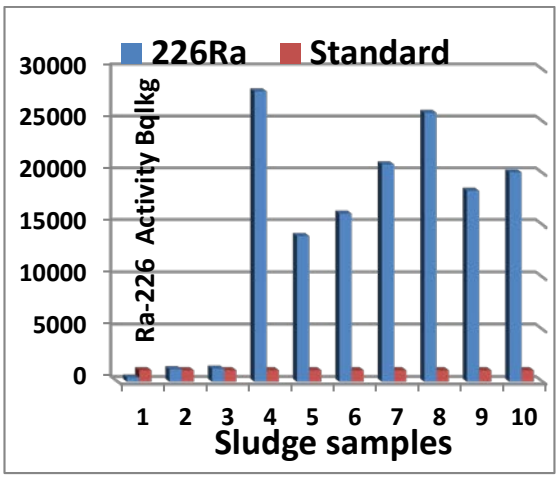


Figure 11: External Hazards of sludge

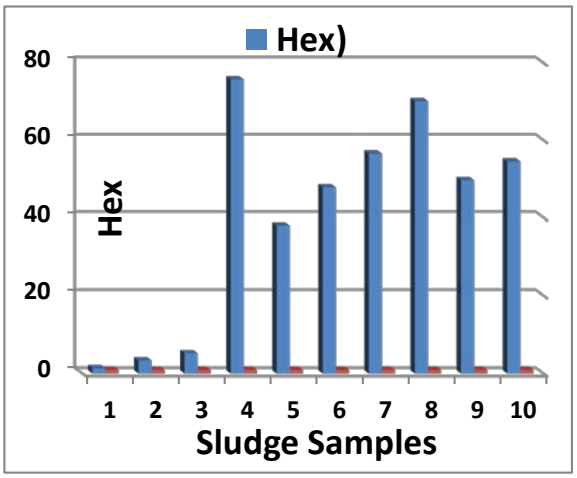


Figure 12: Absorbed dose of sludge

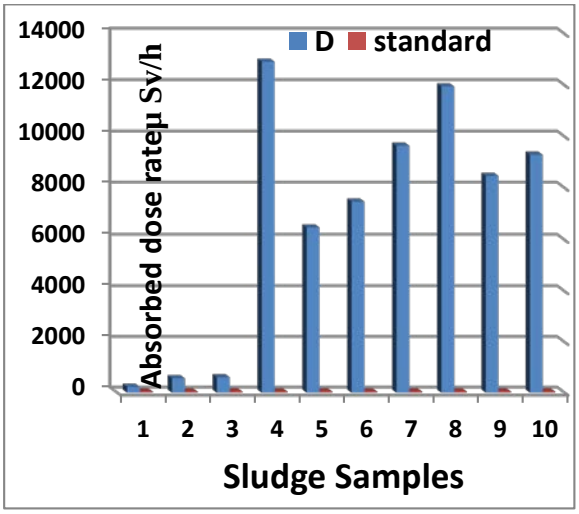
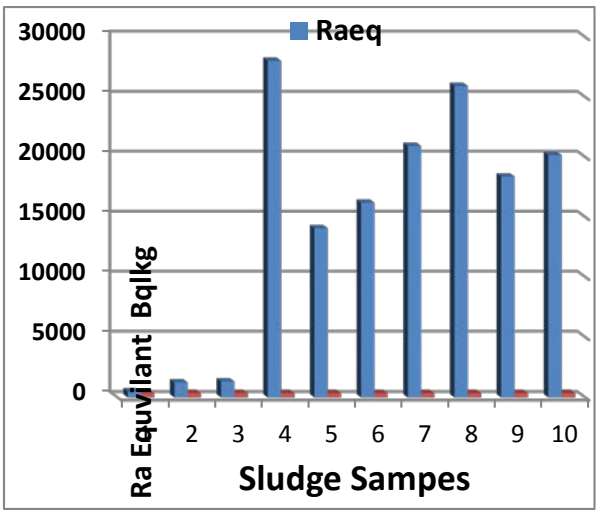


Figure 13: Radium equivalent of sludge



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