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Assessment of radioactivity levels in some oil samples from the western desert, Egypt

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Abstract Distributions of natural gamma-emitting radionuclides were determined in 93 oil samples collected from some petroleum fields in the western desert of Egypt. The radioisotope activities in the area under investigation lay in the range of (21 ± 4) to (38 ± 13) Bq•L^{-1} for ^{226}Ra, (9 ± 2) to (15 ± 5) Bq•L^{-1} for ^{232}Th, and (154 ± 28) to (303 ± 54) Bq•L^{-1} for ^{40}K. The mean values were 27, 12, and 201 Bq•L^{-1}, respectively. Among oils, variations in radionuclide activities could be on account of differences in TDS, HCO_3, and Ba, with high or low pH. In this environment, oil properties differently affected the mobilization of natural radionuclides. The range of ^{226}Ra variation had been compared with available data from other countries. The calculated absorbed dose rate ranged from 22.33 to 32.66 nGy•h^{-1} in location (B) and (E) respectively, which was less than the accepted value.

Keywords Radionuclides, Oil, Activities, Absorbed dose rate
CLC numbers TE622.1^{+}4, TB98, O615.3

1 Introduction

Petroleum is a naturally occurring liquid with widely different compositions of very great complexity. Although there are a few surface seepages, the vast majority of petroleum is found well below the surface of the earth and can be reached only by drilling. Oil wells tap into pools of oil, or into porous rock containing the oil, called reservoirs or fields. The oil is sometimes found, under sufficiently high pressure, to flow to the surface without pumping, but for most wells pumping is required. The amount of oil recoverable from a field by pumping may be only 5%, more frequently 25-30 %, of the oil believed to be present. In fields where the oil is very heavy, steam injection may be used. Complete removal of the oil from a field is not possible even with enhanced recovery methods.

Radioactive substances occur naturally in the environment, but can also result from human activity. Until about a century ago the only sources of exposure to radioactive substances were those that existed in their undisturbed natural state. There were a few exceptions such as the use of uranium oxide as a coloring agent for ceramic glazes[1], and thorium in gas mantles[2], but these were exposed to relatively few people and to only a moderate degree.

Naturally occurring radioactive material (NORM) has been recognized since the early 1930s in petroleum reservoirs, in oil and gas production, and in processing facilities. NORM is typically observed in the barite scale that accumulates on the interior of oil production tubing and in storage tanks and heater-treated separation sludge. Recently, concern has been expressed about the impact on health, from the uncontrolled release of NORM[3]. Trace quantities of the radioactive elements ^{238}U and ^{232}Th as well as ^{40}K have been present in the earth's crust since its formation. Both ^{238}U and ^{232}Th are parents of a complex series of successive decays, producing many radioactive daughters. Some of these daughters may be coproduced with oil/gas well fluids and may concentrate in ordinary deposits (e.g. scale, sludge) and/or be leached in production water. There are several potential exposure pathways to humans from oil field

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NORM, such as, inhalation of radon gas. From the radiological point of view, the only important isotope of radium is ^{226}Ra, which has an average concentration in the earth's crust of about 10^{-12} g/g (40 Bq•kg^{-1}).

The source of most oil and gas NORM is dissolved radium that is transported to the surface in the produced water waste stream. Radium dissolution and precipitation depend on the formation of water salinity, pH, temperature, and pressure. Dissolved radium either remains in solution in the produced water, or under proper conditions, coprecipitates with barium, strontium, or calcium to form either hard sulfate scales or more granular silicate and carbonate sludge[4-6]. The presence of a technologically enhanced naturally occurring radioactive material (TENORM) in the petroleum industry equipment and wastes can be extensively found in literatures[4,5].

The present paper reports a study carried out to determine the concentrations of natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K, in oil samples from oilfields in the western desert of Egypt.

2 Sample preparation

Ninety-three oil samples were collected from different fields in the western desert of Egypt and analyzed by means of scintillation spectrometry. These fields were Wadi El Rayan, Qarun, Harun, Karama, and Beni Suef [Location (A), (B), (D), (E), and (F)] respectively. The oil samples that were collected from a depth that ranged from 9000 to 10000 feet were pure oil, without any water in it. The oil samples were packed in 100 mL cylindrical containers and closed tightly. To reach the radioactive equilibrium between ^{226}Ra and ^{222}Rn and their progenies, the containers were kept sealed for four weeks.

3 Measuring system

Gamma-ray spectrometry is a commonly used technique for direct determination of radionuclides in crystal and extra-terrestrial materials[7]. It is a relative method of analysis and has the advantage of being simple and essentially nondestructive.

The methodology for natural gamma-emitting radionuclide analysis is well described in the literature, for example, Qunidos[8]. The applied low level gamma-ray spectrometer basically consists of a sodium iodide detector (3" × 3", ORTEC model 266) and electronic circuits. The applied detector has the following characteristics: resolution 7.5% for 662 keV, peak efficiency at 1.33 MeV ^{60}Co γ-ray is 4.8×10^{-5}**,** and operation bias voltage is 700 V dc. The system automatically computes and displays the elapsed real-time and life-time, the total counts including the background, net total counts, and the counting rate in each peak.

The background was measured frequently and subtracted from the net count for all measured samples. To get reasonable counting statistics, the measuring time was at least 12 hours.

4 Analytical methods

The primary assessment of radiation exposure of individuals should be carried out in terms of the absorbed dose. In respect of exposure to radiation it is important to assess the gamma- ray hazards because of the specified radionuclides, Ra, Th, and K. This is done by calculating the absorbed dose rate.

The average absorbed dose rate (nGy•h^{-1}), in air one meter above the ground level, in each location, is calculated using the following equation[9]:

$$D = 0.427\,A_{\mathrm{Ra}} + 0.662\,A_{\mathrm{Th}} + 0.043\,A_{\mathrm{K}}$$

To estimate the annual effective dose, account must be taken of the conversion coefficient from the absorbed dose in air to the effective dose. The average numerical values of those parameters vary with the age of the population and the climate at the location considered. In the UNSCEAR[10], the committee used 0.7 Sv/Gy as the conversion coefficient from the absorbed dose in air to the effective dose received by adults[11] and 0.8 as the indoor occupancy factor, that is, the fraction of time spent indoors and outdoors is 0.8 and 0.2, respectively.

5 Results and discussion

The amount of radioactivity that accumulates in oil depends on a variety of factors including the amount of uranium and thorium present in the subsurface formation, the formation fluid chemistry, extraction and treatment processes, and the age of the production well. In all regions under investigation the maximum values for ^{226}Ra, ^{232}Th, and ^{40}K are 62 ± 8,

26 ± 9, and 501 ± 90, ,whereas the minimum values are 5 ± 2, 5 ± 2, and 43 ± 8 (Bq•L^{-1}) respectively, and (^{226}Ra/^{232}Th) ratio is almost constant, ranging from 1.65 to 2.85. The results indicate that ^{40}K exhibits the highest values of activity concentration compared to ^{226}Ra and ^{232}Th. The lowest value of activity is found in the case of ^{232}Th. The average values of ^{226}Ra, ^{232}Th, and ^{40}K are calculated and listed in Table 1.

Table 1 Mean activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K (Bq•L^{-1}) and absorbed dose rate (nGy•h^{-1}) in oil samples for all areas under investigation in Egypt

Location name	No. of samples	Ra-226 (Bq•L^{-1})	Th-232 (Bq•L^{-1})	K-40 (Bq•L^{-1})	Dose rate (nGy•h^{-1})
A	7	24 ± 6	9 ± 4	200 ± 44	25.18
B	45	21 ± 4	9 ± 2	178 ± 33	22.33
D	3	22 ± 5	14 ± 5	303 ± 54	31.46
E	34	38 ± 13	14 ± 5	170 ± 33	32.66
F	4	31 ± 10	15 ± 5	154 ± 28	29.72

As the distribution of the isotopes in different regions is exempted from regulatory control, the values of their activity concentration spread in a wide range for all regions under investigation.

A more detailed analysis of the results for oil samples show the presence of a positive correlation between ^{226}Ra and ^{232}Th for all samples (r = 0.66). The mean values of activity concentration are plotted as a histogram (See Fig.1).

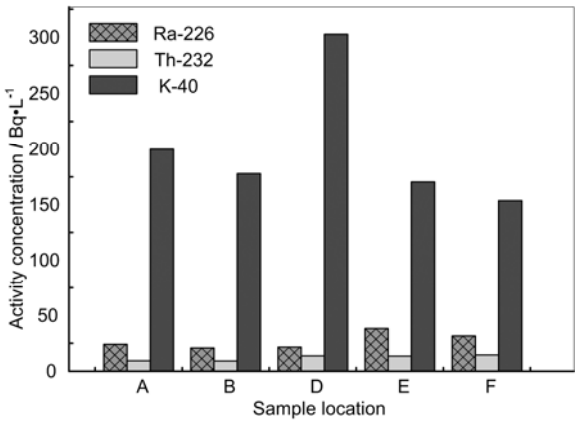

Fig.1 Mean values of activity concentration in oil samples under study.

The ranges of ^{226}Ra level in oil samples at Algeria, USA, and the present investigation are listed in Table 2. The comparison shows that the samples from the western desert of Egypt are at the same level as the ones from USA. Compared to the literature, these results are very close to those reported by Diyashev[12], Snavely[13], and Hamlat[14], though lower values have been reported by others. This is close to the reported range measured by Diyashev and Snavely in USA, but higher than those mentioned by other authors. The highest single measurement of ^{226}Ra in this study is very close to the maximum value reported by Diyashev and Snavely and approximately an order of magnitude higher than other reported values. The minimum of 0.019 Bq/L here, is 200 times higher than the minimum obtained in USA.

Table 2 Natural radioactivity levels of ^{226}Ra in oil samples in different countries

Country	^{226}Ra (Bq•L^{-1})	References
Egypt	0.019 - 0.044	Present study
USA	0.0001 - 0.0400	[12], [14]
Algeria	0.006 - 0.020	[15]

Total radium concentrations depend on the amount of radium present in the subsurface formation, formation water chemistry, extraction processes, treatment processes, and age of production. In general, radium solubility increases in water that has a high saline content and either low or high pH values. Radium precipitation rates increase with decreasing temperature and pressure conditions, such as those encountered when subsurface fluids are brought to the surface[12].

The wide variation observed in both the amount of oil extracted and in the increase of ^{226}Ra and ^{232}Th concentration activities cannot be easily explained, as no direct correlation is found between the two sets of data. Further studies will be carried out to try and determine the factors affecting the oil distribution be-

tween the solid and liquid phases.

6 Radiation hazard indices

The average absorbed dose rates (nGy•h^{-1}) for oil samples under investigation are listed in Table 1. The variation of associated element concentration and radiometric content in some selected oil samples has been studied. Table 3 shows the concentrations of Ba, Sr, HCO_3, SO_4, and total dissolved salts (TDS) (in μg•g^{-1}) determined by XRF, and the corresponding values of ^{226}Ra and ^{232}Th.

Table 3 Chemical and radionuclide content of oil samples

Sample code	Chemical content of oil samples/μg•g^{-1}					Activity concentration /Bq•L^{-1}	
	Ba	Sr	HCO_3	SO_4	TDS × 10^3	Ra-226	Th-232
BO.2	20	936	100	180	250	29.85	11.15
BO.4	17	930	116	300	230	17.62	11.14
BO.8	16	865	133	300	226	9.94	8.85
BO.9	24	949	117	200	256	25.76	9.24
BO.13	20	948	147	275	229	16.34	8.28
BO.16	21	1164	140	300	233	25.03	11.58
BO.17	18	925	150	325	238	12.50	7.95
BO.19	17	730	134	225	210	24.47	8.29
BO.24	17	940	140	275	220	22.46	8.64
BO.28	23	768	146	225	230	25.84	10.27
BO.29	17	890	128	280	230	18.26	11.15
BO.35	17	750	136	290	210	15.02	6.66
BO.38	18	780	110	330	230	29.27	8.29
BO.41	10	525	120	300	200	33.06	10.46
DO.2	9	340	450	450	139	26.51	19.35
EO.4	10	294	400	250	112	46.43	11.38

From the result, it can be seen that ^{226}Ra and ^{232}Th concentrations increase with increasing the TDS and increasing barium (Ba) with high or low pH. This means that radium solubility increases in oil, which has a high saline content, and high barium concentrations also increase with an increase in Sr, SO_4, and HCO_3 concentrations.

There is probably a positive relation between ^{226}Ra and ^{232}Th with Ba and TDS, which means that radium solubility increases in oil with high saline content.

Future research should include the systematic studies on ^{226}Ra and ^{232}Th concentrations in sapropel and other organic materials known to be associated with petroleum genesis; and the correlation between the age of the petroleum and the concentrations of ^{226}Ra and ^{232}Th in sapropel and NORM.

7 Conclusions

From the results one can summarize the following:

(1) In the areas under investigation, high concentration values for ^{226}Ra and ^{232}Th were observed in locations E and F, and the lower value was observed in location B. The maximum value of ^{40}K was observed in location D and the minimum value in location F.

(2) The results indicated that ^{40}K exhibited the highest value of activity concentration compared to ^{226}Ra and ^{232}Th, and the lowest value of activity was found in the case of ^{232}Th.

(3) ^{226}Ra behaved in parallel with ^{232}Th in all areas under investigation, where the two isotopes reached their maximum and minimum values mostly in the same samples, that is, the ^{226}Ra/^{232}Th ratio was almost constant ranging from 1.65 to 2.85.

(4) As the distribution of the isotopes in different

regions was exempted from regulatory control, the values of their activity concentration spread in a wide range in all regions under investigation.

(5) ^{226}Ra and ^{232}Th concentrations increased with increasing the total dissolved salts (TDS), barium (Ba), strontium (Sr), sulphate (SO_4), and bicarbonates (HCO_3).

It must be taken into consideration that the wastes with granular characteristics associated with oil and gas fields are more hazardous to the staff operators as compared to the wastes with small grain size.

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