



Natural Radionuclide Analysis on Crude Petroleum from Some Oil Fields in Ghana

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

The petroleum industries benefit immensely from developing scientific processes and procedures that determine characteristics of crude oils and distinct them. This is because crude oils are acquired from various geological sources and each sedimentary rock has its own distinct chemical properties that affect extraction, refinery and environmental safety. Extraction of crude oils may result in exposure of some radionuclide elements from sedimentary rocks into the environments. This nuclear exposure may affect lives and can cause ecological imbalance. Applications of gamma emission spectroscopy has grown rapidly around the world because of its accurate measurement and precision. Such technique has not been applied for the study of naturally occurring radionuclide materials (NORMs) in crude oils from the active oil fields in Ghana. The study employed the use of gamma spectroscopic technique for characterization of crude oil samples from Jubilee Oil Field, Tweneboa Enyenra Ntomme (TEN) Oil Field and Saltpond Oil Field. The technique was applied on four crude oil samples. The results of this paper actually report on radionuclide concentrations of naturally occurring radioactive materials in four crude samples using a Gamma-Ray spectrometer. Three radionuclides, radium-226 (²²⁶Ra), thorium-232 (²³²Th) and potassium-40 (⁴⁰K), were identified from each crude sample (JF, TF SF-1 and SF-2). It was observed that samples from Saltpond Oil Field estimated the highest activity concentration and the

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least was crude oil from Jubilee Oil Field. The activity concentration values were far less than the approved values by International Atomic Energy Agency for basic safety standard. Therefore, the radionuclides activity concentration values of crude oil samples from Jubilee, TEN, and Saltpond oil fields were within the basic safety standards exemption levels of the International Atomic Energy Agency (IAEA). The crude oils from the oil fields can therefore be considered safe to handle. Also, the results of the activity concentrations from the sample used for this study indicate that crude petroleum from the oil fields may relatively have little nuclear activity impact to ecology and human health. The result of this study may be adopted by the Environmental Protection Agency and the Petroleum Stakeholders of Ghana for development of petroleum geo-radionuclides radiation safety measures.

Keywords: Gamma-Ray spectroscopy; crude oil; geo-radionuclide and activity concentration.

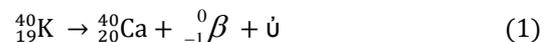
1. INTRODUCTION

Studies on radioactive materials have shown that coming into contact with extreme levels of radiation can lead to damages to live tissues and cells (Agency for Toxic Substances and Disease Registry [1]. An epidemiological study concerning people exposed to radiation during the 1945 atomic bombing of Hiroshima and Nagasaki has proven that exposure to nuclear radiation usually causes potential health threat. Thus, it is significant to subject every human activity that involve nuclear radiation exposures to strict safety standards to avoid nuclear radiation hazards on human, animals, and vegetation (International Atomic Energy Agency [2,1]. Crude oil exploration and exploitation are human activities that usually involve exposure of radionuclide materials found in sedimentary rocks to our environment. Crude petroleum is mostly found in sedimentary rocks; and each sedimentary rock has its own geo-radionuclide properties. Therefore, samples of materials from sedimentary rock under exploration or exploitation need thorough investigations to avoid dangerous geo-radionuclide exposure.

Uranium, radium, thorium and radon are naturally occurring radionuclides in the earth crust [3]. They are dissolved in very minute quantity during chemical reactions between water and rocks [4]. The coexistence of underground-groundwater and petroleum oil reveals that there exists significantly high concentration of dissolved mineral constituents that accumulate during prolonged periods of water-rock contact. Majority of waters located in oil fields are rich in chloride, and this enables the solubility of other elements including radioactive elements such as radium and thorium, which is one of the most dangerous decay products of uranium [5,6]. Radium is heavy metal, radioactive and a potential alpha emitter. As it decays, it produces radon gas as a

by-product. Radium is chemically similar to calcium, so when ingested, it migrates to the bones, teeth and milk of humans and animals [6-7]. Although some radium may be still used for medical purposes, only small quantities are needed. Most of the world's radium are discarded with the crushed rock left over from uranium mining, even though it is known to be hazardous [6]. The radioactive decay of uranium and thorium found in rocks produces radium-226 and radium-228 (United States Geological Survey [1].

Potassium-40 (^{40}K) is a radioactive isotope of potassium, which has a half-life of 1.251×10^9 years. It makes up 0.012 % (120.0 ppm) of the total amount of potassium found in nature. Potassium-40 decays to calcium-40 (^{40}Ca) with the emission of a beta particle and antineutrino [8] with maximum energy of 1.31MeV as shown in equation (1).



where $\bar{\nu}$ is antineutrino and $^0_{-1}\beta$ is beta particle.

Although potassium-40 isotope makes up 0.012 % of the total amount of potassium found in nature there is no proven health danger of its radiation from nuclear decay to lives [9]. Conversely, thorium-232 is a weakly radioactive metallic chemical element that decays very slowly through alpha decay [8]. Thorium-232 (^{232}Th) usually cause health risk in the environment because of the rapid build-up of radium-228 (^{228}Ra) and its associated gamma radiation [10,11]. Radon-220 (^{220}Rn) and its decay species cause lung infections. Thorium is present in very low quantities in the environment. Human exposure to thorium is mainly by air through dust inhalation; and oral through food and water [10].

Also, Thorium, Bismuth and Uranium on earth are the only three radioactive elements that still exist naturally as primordial elements in significant amounts [10-11].

This paper aims to employ Gamma-Ray spectrometer to determine the radionuclide activity concentration (specifically, the naturally occurring radioactive elements) in crude oil samples to enhance better environmental nuclear radiation pollution assessment for the petroleum industries of Ghana. Information about radionuclide isotopes is very important because it provides key for the assessment of radiological risk and hazard. Since crude oil exploration may lead to dangerous radionuclides exposure and contaminations, all ecological life species need to be protected [11]. Crude oil exploration and exploitation activities have started intensively at different geological sites along the coast of Ghana in the Accra-Keta Basin, Saltpond Basin and Tano Basin over the last three decades. These crude petroleum exploitation activities produce sludge and scale formation in oil production pipes that ought to be removed [11]. Removal of such petroleum wastes may result in dangerous radionuclide exposure and contamination in the ecological system, hence an interesting area that calls for thorough scientific investigations. Improper evaluation and monitoring on the handling of such petroleum industrial exploitation waste could bring potential health risk to marine aquatic lives, staff of petroleum industries and general public [11]. Series of investigative research work conducted for accurate information on NORMs in crude oil samples from Jubille Oil Field, TEN Oil Field and Saltpond Oil Field will help to preserve our natural environment. It is therefore important to conduct spectral analysis on crude oils from Ghana to identify and quantify the geo-radionuclides particles present in them for safety measures. Investigation into the crude oil samples using Gamma-Ray spectroscopic technique will provide accurate data on radioactivity content to all petroleum stakeholders for proper handling of the crude oils and their industrial waste.

2. MATERIALS AND METHODS

2.1 Crude Oil Samples

Four different composite run-down petroleum crude oil samples were obtained from Jubilee, TEN and Saltpond oil fields through the Research and Development Unit of Ghana

National Petroleum Corporation (GNPC). The packaged samples were transported from GNPC to the Laser and Fiber Optics Centre (LAFOC), University of Cape Coast for inspection. The crude samples were kept at average temperature of 28.0 °C at National Nuclear Research Institute, Ghana Atomic Energy Commission till gamma spectra measurements were taken.

Table 1 shows the summary of the crude oil samples, oil field locations and sample codes used in this study. 2.0 litres each of composite run-down crude samples were acquired from Jubilee oil Field and Tweneboah Enyenra Ntomme. However, 2.0 litres each run-down but non-composite crude samples were acquired from the oil well-2 and well-4 Saltpond Oil Field.

2.2 Experimental Setup

The experiment was conducted using AMETEK ORTEC high purity germanium (HPGe) GEM series coaxial detector system (P and N-type) at the National Nuclear Research Institute, GAEC. Its resolution (Full Width at Half Maximum) at 1.33 MeV is 1.8 keV with a Peak-to-Compton ratio of 62:1. The Gamma-Ray spectrometer device has a nominal efficiency of 1.33 MeV. It has an in-built cryogenic cooling system, which uses both liquid nitrogen and an electro-mechanical cooler. The low-frequency digital signal processor of the gamma spectrometer (DSPEC LF) analyzed the stream of electric pulses and produce a spectrum of the nuclear activity versus pulse-height, which often relates to the energy or time of arrival. The analogue-to-digital converter of DSPEC LF has a signal gain of 64 K channels for expanded energy range applications.

2.3 Gamma-Ray Measurement

For Gamma-Ray determinations, the samples were each transferred in 500.0 ml Marinelli beakers and sealed with masking tape to avoid spillage as shown in Fig. 1. The samples were analyzed by a high purity germanium (HPGe) GEM series coaxial detector system of both P-type and N-type. The resolution (full wave at half maximum) of the detector system was kept at 1.8 keV for Gamma-Ray energy of 1.33 MeV of ⁶⁰Co and a nominal efficiency of 38.0 %. The data acquisition of the gamma spectrum was done for 24 hours (86400.0 s) using an ORTEC Gamma-Ray spectroscopy system. Fig. 2 presents a Marinelli beaker installed on the detector of the Gamma-Ray spectrometer.

To account for background contribution to the sample spectrum, an empty sample container was counted for 86400.0 seconds (24 hours) as well. The embedded multichannel analyzer

controls the advanced spectrum analysis functions, automation for routine operations, quality control and security, and data evaluated using the Gamma Vision software package.

Table 1. Summary of the Crude Oil Samples, Oil Field Locations and Sample Codes used

Oil Field	Crude Oil Sample	Sample Code
Jubilee	FPSO MV21 Kwame Nkrumah	JF
Tweneboa Enyenra Ntomme	FPSO MV25 Atta Mills	TF
Saltpond	Saltpond Well-2	SF-1
Saltpond	Saltpond Well-4	SF-2

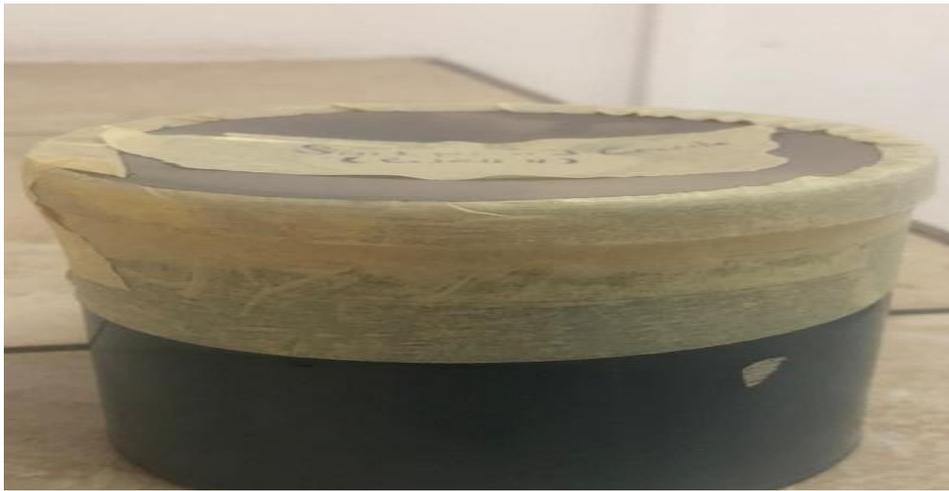


Fig. 1. Marinelli beaker filled with crude oil and sealed



Fig. 2. Detector-beaker installation

The Fig. 3 shows the efficiency calibration curve obtained using a standard calibration source with radionuclides (⁶⁰Co) of known activity. From the equation in the fig. 3, r-square value is 0.9997 which implies very strong correlation between the two parameters.

processed and stored in the channels allocated by the DSPEC LF Multi-Channel Analyzer. The required energy and efficiency calibrations performed are presented in Fig. 3. The figure basically shows the logarithmic efficiency as against energy.

2.4 Radionuclides Analysis of the Crude Oils

The gamma energies of the radionuclides of interest in each crude sample were identified,

Fig. 4 shows a graph of energy calibration as against channel. From the graph, there is strong positive coefficient between the Gamma-Ray intensity and the result of Multi-Channel Analyzer (MCA).

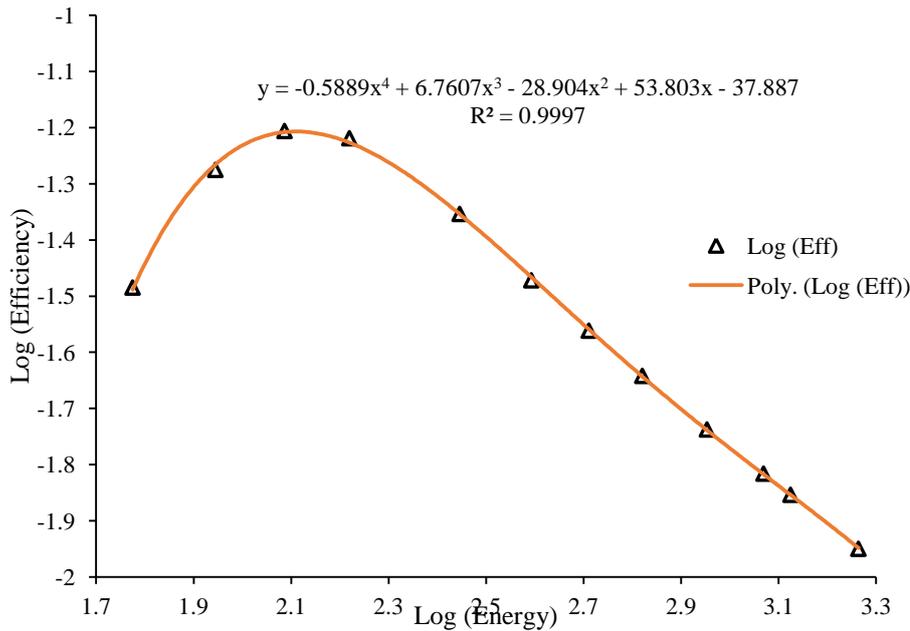


Fig. 3. A Logarithmic Graph of efficiency as against energy

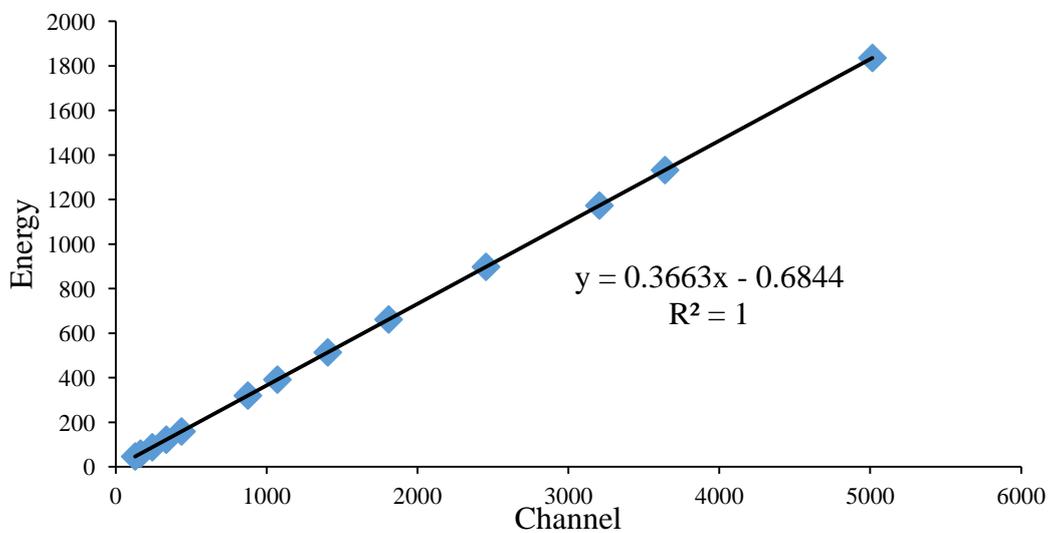


Fig. 4. Energy calibration graph showing the relationship between the channels and their gamma intensities

The r-squared value obtained from the bivariate regression analysis is 1 showing how strong the number of channels could predict the energy or intensity of the gamma-ray. In this case, 100.0 % of the variance in energy is expected from the channels since there is strong relationship [12]. Thus, for each unit increase of energy, the number of channels receiving the signals is increased correspondingly.

3. RESULT AND DISCUSSION

After series of computational run, the results of the radioactivity concentrations of naturally occurring radionuclides that were identified in the four crude samples are shown in Table 2. The radionuclides were radium-226 (^{226}Ra), thorium-232 (^{232}Th) and potassium-40 (^{40}K). The activity concentrations are recorded in Becquerel per gram. From the table, radionuclide activity concentration of crude sample JF and TF represent activity from Jubilee Oil Field and TEN Oil Field respectively. However, the average radionuclide activity of crude sample SF-1 and SF-2 represent Saltpond Oil Field.

3.1 Discussion

From Table 2, it is observed that the radionuclide activity of potassium-40 in the four samples were relatively higher than radium-226 and the least activity element is thorium-232. Therefore, radionuclide activity concentrations of the three elements in Becquerel per grams among the four crude samples decreased orderly from potassium-40, radium-226 and thorium-232. Also, the presence of radium-226 and thorium-232 in minute quantity in the crude samples may suggest the presence of uranium in the sedimentary rocks from which the samples were acquired [6]. This is because scientific studies have shown that both radium-226 and thorium-232 may be one of the usual decay products of uranium [6,13]. Comparing the activity concentrations of the two harmful radionuclides radium-226 and thorium-232, the earlier is observed to be relatively higher than the latter.

From Table 2, the values of radionuclide activity concentrations of the same element in crude samples from Saltpond Oil Field are not the same, even though SF-1 and SF-2 are oil wells located in the same sedimentary rock. For example, the activity of radium-226 of sample SF-1 is 1.646 times that of SF-2. Concerning the activity concentration of thorium-232 in crude samples from Saltpond Oil Field, the average

activity of thorium-232 in sample SF-2 is 1.4054 times that of SF-1. Similarly, radionuclide activity of radium-226 in sample TF is 1.181 times that of sample JF even though the samples were acquired from different oil fields; however, they are located in the same sedimentary rock called Tano Basin [14]. Also, nuclear activity of thorium-232 in crude sample TF is 1.7262 times that of sample JF. These suggest that crude oil composition varies significantly from each oil field as well as the oil-wells due to the differences in their geological origins [14-16]. Other physical conditions such as temperature and pressure affect the chemical composition of crude oils during formation of the sedimentary rock. These effects may lead to differences in geochemical elemental distribution, hence the variation in radionuclide activity concentrations among the crude samples from oil fields located in the same sedimentary rock. These distinct variations in activity concentrations were observed to confirm the observations by other researchers that the geochemical elemental distribution of each of the two rocks, Saltpond Basin and Tano Basin, is not uniform [14-15].

Fig. 5 shows three bar graphs; each showing the activity concentrations of a geo-radionuclide element measured from the four crude oil samples. Fig. 5 shows the activity concentrations of potassium-40 in each sample whilst Figs. 6 and 7 show that of thorium-232 and radium-226 respectively.

From the Fig. 5, it is obviously seen that the highest activity concentration of potassium-40 was recorded from crude oil sample JF and the least from sample TF. However, on average, the activity concentration of potassium-40 of crude samples from Saltpond Basin (SF-1 and SF-2) was 1.0672 times the concentration of samples from Tano basin (JF and TF). Conversely, from Fig. 6, samples from Tano Basin (JF and TF) on average recorded the highest activity concentration of thorium-232 as compared to the average of SF-1 and SF-2. With regard to activity concentration of radium-226, in Fig. 7, the opposite trend is observed.

Concerning the oil fields, the highest activity concentration was acquired from Saltpond Oil Field; followed by TEN Oil Field and the least is Jubilee Oil Field. The highest radionuclides activity contents of crude samples from the Saltpond Oil Field may be due to the fact that relatively, Saltpond Basin has higher metallic elemental concentration as compared to the

Tano Basin [14]. Also, it may suggest that larger number of heavy metallic ions were involved during the period of formation of the sedimentary rock of Saltpond Oil Field many years ago. These accumulated heavy metallic ions formed various chemical bonds with the hydrocarbons in the Saltpond Basin and continuously undergoes series of nuclear decay to achieve stability,

hence recording higher radionuclide activity concentration than Jubilee Oil Field and TEN Oil Field. Since TEN Oil Field and Jubilee Oil Field are located in the Tano sedimentary rock, it implies that Tano Basin produces petroleum crude of lower radionuclide activity content than Saltpond Basin.

Table 2. Activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K

Oil Fields	Crude Oil Samples	Radionuclide Activity Bq/g		⁴⁰ K
		²²⁶ Ra	²³² Th	
Jubilee Oil field	JF	0.00337	0.00084	0.04815
TEN Oil Field	TF	0.00398	0.00145	0.03088
Saltpond Oil Field	SF-1	0.00556	0.00074	0.04485
	SF-2	0.00322	0.00104	0.03949

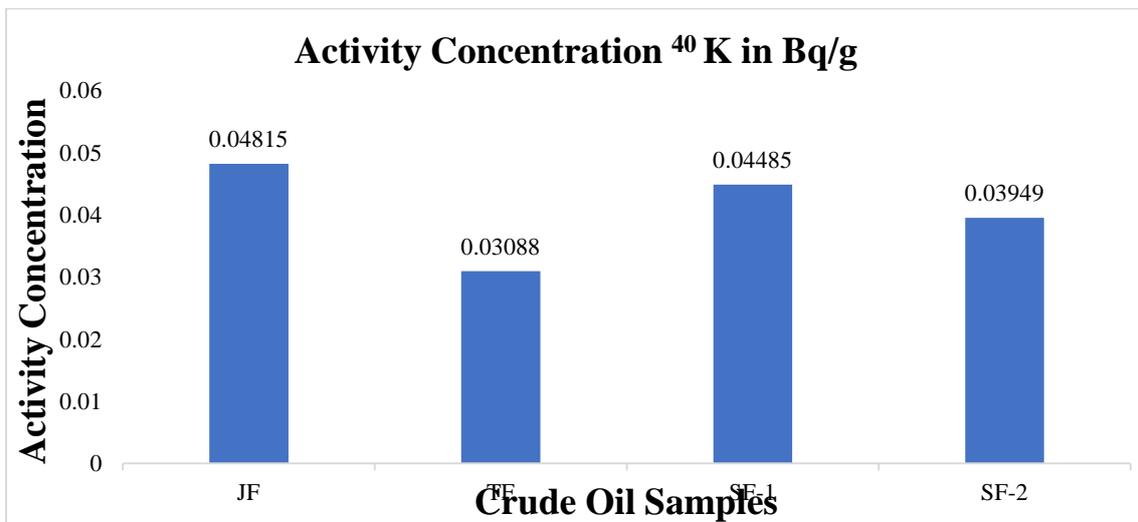


Fig. 5. Radionuclide activities concentrations of potassium-40 in each sample

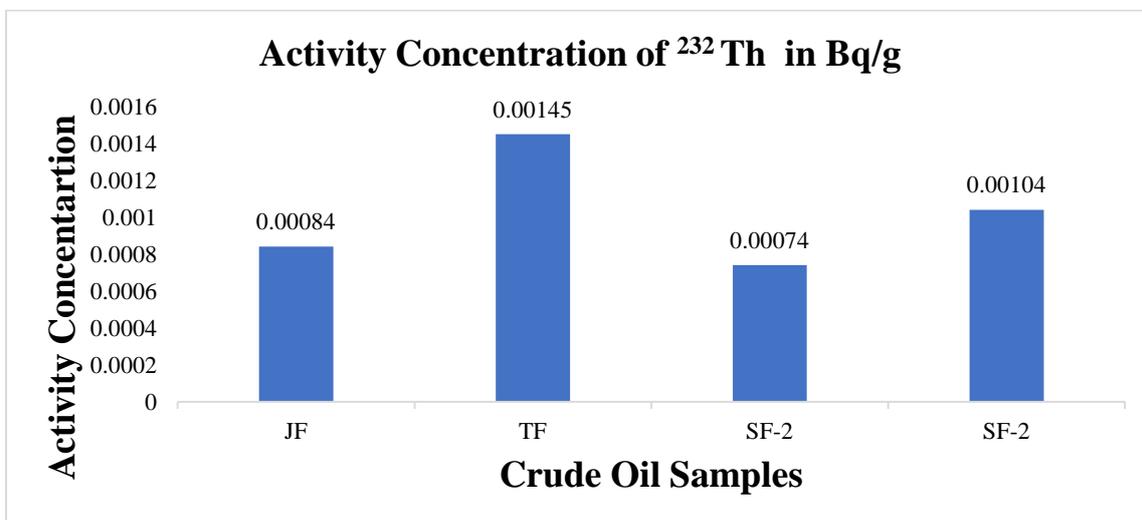


Fig. 6. The radionuclide activity concentrations of thorium-232 in each sample

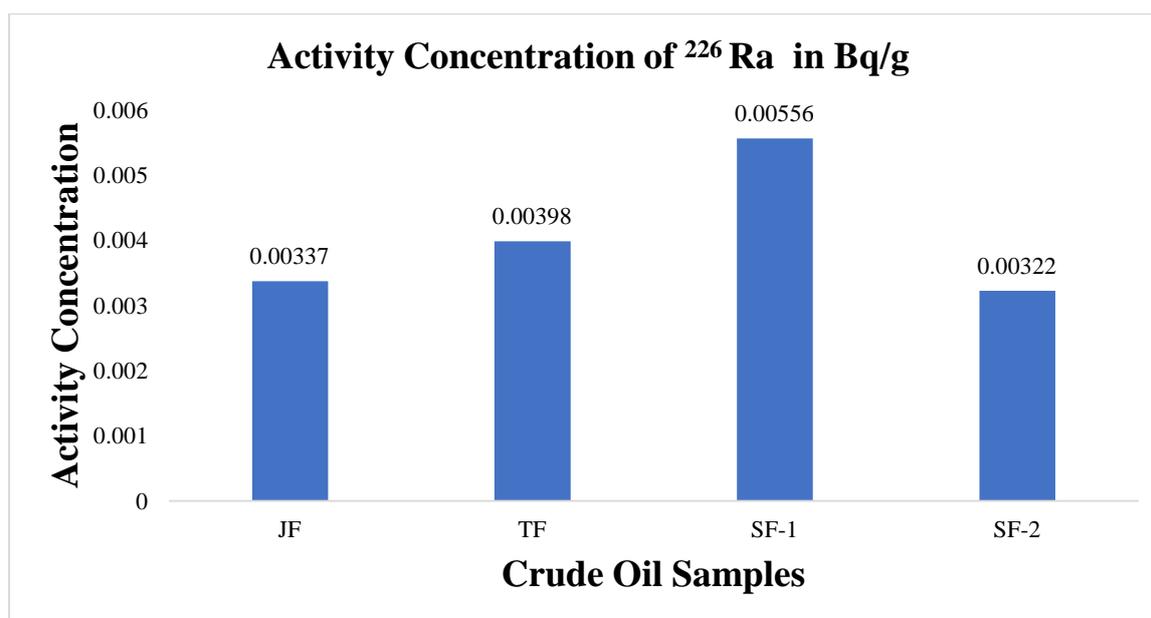


Fig. 7. The radionuclide activity concentrations of radium-226 in each sample

From the basic safety standard of IAEA, the accepted radionuclide activity concentration level of potassium-40, radium-226 and thorium-232 of natural origin is 10.0 Bq/g [17]. Comparing the concentrations of the three geo-radionuclide elements that were identified from table 2 to the accepted safety level by IAEA, it may be observed that the activity concentration level of each sample is relatively low [17]. Therefore, based on the result of the study, the oil fields located in the two sedimentary rocks, Saltpond Basin and Tano Basin, of Ghana produce crude oil of very low level of geo-radionuclide activity concentration.

4. CONCLUSION

From the study, three naturally occurring radioactive nuclides, potassium-40, radium-226 and thorium-232, were identified from each crude sample, with activity concentration quantified. Among the three identified radionuclides, the activity concentration of potassium-40 was the highest and thorium-232, the least in each sample. The results of the Gamma-Ray spectral experiment conducted showed that the crude samples have non-uniform radionuclide activity concentrations. That is, the sedimentary rocks of the oil fields from which the samples were acquired have non-uniform geo-radionuclide distributions, hence the two rocks are naturally geo-radioactively dissimilar.

However, it is worth mentioning that although the activity concentrations of radium-226 (^{226}Ra) and

thorium-232 (^{232}Th) which naturally produce harmful decay product fell below the accepted value of the IAEA basic safety standards. The results obtained using gamma spectrometry in analyzing the activity concentrations showed that the activity concentrations of the radionuclides (radium-226, thorium-232 and potassium-40) present in the four samples are very low. This implies that the crude petroleum and its products from the Saltpond Basin and Tano Basin of Ghana are safe to handle. Handling samples of these crude oils during the refinery processes, transportation, and research would not pose any major health threat. Therefore, based on the radionuclide activity concentration level of the study, crude oils produced from the three oil fields have relatively very low nuclear radiation health hazards. Hence the offshore petroleum activity may pose little ecological imbalance to both aquatic and terrestrial lives.

Since exposure to these radionuclides have proven health consequences, care must be taken to prevent any mode of exposure (inhalation, oral and contact) to avoid radionuclide health risk. Lastly, natural decay products of uranium were observed in all the four crude samples from the three oil fields. Therefore, the sedimentary rocks along the coast of Ghana in the Gulf of Guinea may be a potential source of uranium since radium-226 and thorium-232 are natural decay products of uranium. The results of the study may be employed by Environmental Protection Agency, Fishing Industries and Petroleum

Stakeholders for decision making on environmental radionuclide radiation impact assessment from crude oil exploration and exploitation.

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COMPETING INTEREST

The authors of this paper do hereby declare that no competing interest exist in the study conducted.

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