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Radiological Impacts of NORM and Poly Aromatic Hydrocarbon in Petroleum Industry Process on Marine Ecosystem at the Red Sea, Egypt



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Abstract

This study was designed to realize the hazardous effects on marine ecosystem, and assess the impacts of (NORM) ²³⁸U, ²³²Th, and ⁴⁰K radioactivity and polycyclic aromatic hydrocarbons in petroleum industry at red sea coastline in Abu Zenima and Abu Rudeis sites. Polycyclic aromatic hydrocarbons(PAHs) individuals Naphthalene, Acenaphthene, Acenaphthylene, Fluorene, phenanthrene and Fluoranthene were analysed with HPLC with fluorescence detection for sea water, sediments and marine organisms Bivalves, coral reef and starfish. HPGe gamma ray spectrometric technique used for measured. The ²³⁸U, ²³²Th, and ⁴⁰K radioactivity concentration in sea water, sediments and marine organisms Bivalves, coral reef and starfish in both Abu Zenima and Abu Rudeis sites. The data revealed an increase in the ²³⁸U, ²³²Th, and ⁴⁰K and (PAHs) individuals for sea water, sediments and marine organisms Bivalves, coral reef and starfish in Abu Zenima and Abu Rudeis sites respectively as compared to international maximum allowable concentrations. On the other hand, ²³⁸U, ²³²Th, and ⁴⁰K and (PAHs) individuals for sea water, sediments and marine organisms Bivalves, coral reef and starfish were recorded higher concentration levels in Abu Zenima sites than Abu Rudeis sites. The data revealed an increase in concentration of (NORM) ²³⁸U, ²³²Th and ⁴⁰K and PAHs individuals for starfish and coral reef than bivalves marine organisms for both Abu Zenima and Abu Rudeis sites respectively as compared to international maximum allowable concentrations.

Keywords: NORM; Poly Aromatic Hydrocarbon; Petroleum Industry; Radiological Impacts; Marine Ecosystem

Introduction

The oil and hence polycyclic aromatic hydrocarbons (PAHs) are naturally present in the marine environment, although levels have increased significantly following human extraction and use of oil and gas [1]. Naturally occurring radioactive materials (NORM) generally contain radio nuclides found in nature, i.e., thorium, uranium, and their progeny [2]. The TENORM waste is produced from several industrial sectors such as uranium and metal mining, phosphate ores. Processing and petroleum industry [3]. The petroleum waste (scale or sludge) have been produced by two mechanisms: either incorporation or precipitation onto the production equipment such as: pipelines, storage tank, pumps [4]. The precipitated TENORM wastes around walls of the petroleum pipes reduce their efficiency and then disposed and replaced periodically by new ones [5]. In the last decade, attention was focused on the environmental and health impact from the release of TENORM wastes.

Studies of naturally occurring radioisotopes of uranium and thorium decay series and primordial potassium in aquatic environment provide information on the environmental pollutants in water bodies, whereas marine invertebrates include sea slugs, sea anemones, starfish, octopuses, clams, sponges, sea worms, crabs, lobsters, coral reef and Bivalves, most of these animals are found close to the shore [6]. The possible radionuclide transfer provide an easy assimilation of radiation exposure into human body following their consumption [7,8]. Moreover, marine organisms such as filter feeders and piscivorous have the capacity of bioaccumulation more radionuclide's and toxic elements from water due to the physical and chemical nature of their body surfaces and feeding habit in their natural habitat, and thus, the determination of radioactivity in the marine organisms and estuaries marine animals assumes greater importance [8]. The level of natural radionuclides in marine organisms (²²⁶Ra, ²²⁸Ra and ⁴⁰K) in mostly consumed marine fishes and shell fishes due to their importance as sources of high-quality protein [9].

Releases of radio-nuclides TE-NORM from marine petroleum industry (226 Ra, 228 Ra and 40 K) effects on aquatic animals and marine food chains [9]. Among the many radio nuclides discharged into marine environments, (226 Ra, 228 Ra and 40 K) the fate of these radio nuclides in the aquatic food chain is essential

for a realistic assessment of the risk of their potential impact on human health [10]. The contribution of naturally occurring radio nuclides reflected in the sediments and marine coastline water. The radiation dose received and accumulated in the body by marine fauna comes from the naturally occurring uranium series [11]. On the other hand petroleum industry in marine have the Polyaromatic hydrocarbons (PAHs) beside ²³⁸U, ²²⁶Ra, ²³²Th and ⁴⁰K are formed during the manufactured oils in marine and sea water [11]. Oil and its derivatives PAHs have been found to be toxic to a wide array of marine organisms from marine invertebrates to large seabirds and marine mammals [12]. The effects of PAH son single organisms may add up to affect populations and communities, as well as whole ecosystems [12]. Phytoplankton, the major primary producer in the marine environment, is also sensitive to environmental stressors such as pollution. Phytoplankton plays a crucial role in the inflow of primary energy to coastal food webs; thereby changes in phytoplankton may cause significant damages to the functioning of marine ecosystems [13]. Abu Zenima and Abu Rudeis located on Suez gulf, this area have many oil offshore platform, several of these offshore platforms have been in operation to extract and process oil, processing and petroleum products distribution activities and have resulted in tremendous impacts on the safety and environmental conditions in the Suez Gulf. PAHs leaks and spills have resulted in serious contamination to the Suez gulf. PAHs contain many aromatic generation compounds such as naphthalene, Acenaphthalene, Acenaphthene, fluorene, phenanthrene; Fluoranthene Each PAHs can also be released into the environment, mainly due to offshore oil production or petroleum transportation [14]. Each PAHs have bad impacts contamination on bivalves and particularly mussels coral reef and star fish and marine invertebrates the status of the marine environment for large number of pollutants [15]. Where, PAHs are lipophilic and coplanar; they can accumulate in adipose tissues or secretions. Coral reefs are threatened by small chronic PAHs spills in particular, but larger acute oil spills may also affect coral reefs [15]. Observed biological impacts of oil spills in reef areas range from mass mortality of fish and invertebrates apparently marine ecosystem devastation [16]. PAHs components can dissolve in water to some extent which exposes the corals to potentially toxic compounds. However, toxic concentrations are only encountered in the uppermost part of the water-column [16]. In the Suez Gulf PAHs easily gets trapped in the mangroves and usually persists for a very long time. The PAHs is subject to microbial degradation which may be a rather rapid process in aerobic environments. However, if the PAHs are buried within the anaerobic sediments, bio-degradation proceeds very slowly [17].

Materials and Methods

Study Area

The investigated area expanded along the coast of Suez gulf, Abu Zenima $29^{\circ}3'20"N$ $33^{\circ}6'11"E$ to Abu Rudeis $28^{\circ}53'N$ $33^{\circ}11'E$.

Sampling and Sample Preparation

Six representative superficial shore sediment, sea water and marine organisms samples (Bivalves, Coral reef and Star fish) were collected from six different stations located along the offshore oil platforms in Abu Zenima (Abu Zenima 1, Abu Zenima 2, Abu Zenima 3), and Abu Rudeis 1, Abu Rudeis 2 and Abu Rudeis 3). An area of about 25×25 cm2 up to a depth of 5cm was cut out using the stainless steel template for guidance [18]. The collected shore sediment and sea water samples were transferred to labelled polyethylene bags, closed and transported to the laboratory for preparation and chemical analysis. The shore sediment samples were air-dried at room temperature for a week. And also were dried in an oven at 80°C (for 48h) till constant dry weight was obtained, crushed and homogenized. Then milled and sieved through 0.4mm mesh sieves and stored for further analysis. Water samples, 5 liter of each, were collected using the water sampler.

They were collected in polyethylene containers. Then, the samples were acidified with Nitric acid to pH lower than 2 to avoid micro-organisms growth. The samples were stored for radioactivity and Polycyclic aromatic hydrocarbons measurements. The marine organisms samples of Coral reef, Bivalves and Star fish were collected from Abu Zenima and Abu Rudeis and transported to the laboratory in ice boxes and stored at (-10°C) until subjected for further analysis, about 20 individuals of each species were collected from the study area; at the same locations of the water samples. The samples of marine organisms were then cut into smaller pieces to ensure effective grinding. The cleaned samples were dried in an oven at 70°C for five days (until there was no detectable change in the mass of the samples) to ensure that the sample were completely moisture free, a constant dry weight being obtained. Dried samples were ground to fine grain sizes by using a stainless steel cutter blender, and sieved in order to obtain homogeneity. All homogenized samples were divided into two parts. The first part was transferred into 250ml sizes Marinelli beaker, sealed hermetically, and left for about 4 weeks at room temperature in order to attain secular equilibrium among the ²³⁸U-series and ²³²Th-series precursors with their short-lived progenies [19]. The second part was kept in laboratory room temperature for using it in analysis the polycyclic aromatic hydrocarbons Individuals.

Radioactivity Measurements

The activity concentration of the natural radioactivity $^{238}\text{U},$ ^{232}Th and ^{40}K in the investigated samples were determined using a high-resolution HPGe $\gamma\text{-spectrometry}$ system with 30% counting efficiency. This was performed by taking $250~\text{cm}^3$ counting vials filled up to a height of 7 cm, which correspond to $170~\text{cm}^3$. The measurement duration was up to 80,000sec and was carried out in the Laboratory of Egyptian Nuclear and Radiological Regulatory Authority. The obtained spectra were analyzed. The determination of the presence of radio nuclides

and calculation of their activities were based on the following gamma-ray transitions (in keV): the ^{226}Ra activities (or ^{238}U activities for samples assumed to be in radioactive equilibrium) were estimated from ^{234}Th (92.38keV, 5.6%), while $\gamma\text{-energies}$ of ^{214}Pb (351.9keV, 35.8%) and ^{214}Bi (609.3,45%), 1764.5keV, 17%) and ^{226}Ra (185.99KeV, 3.5%) were used to estimate the concentration of ^{226}Ra .

The Gamma-ray energies of ²¹²Pb (238.6 keV, 45%), and ²²⁸Ac (338.4keV, 12.3%), (911.07keV, 29%), (968.90 keV, 17%) were used to estimate the concentration of ²³²Th. The activity concentrations of ⁴⁰K were measured directly by its own gamma rays (1460.8keV, 10.7%). In order to determine the background distribution due to naturally occurring radio nuclides in the environment around the detector, an empty polystyrene container was counted in the same manner as the samples. The activity concentrations were calculated after measurement and subtraction of the background. The activities were determined from measuring their respective decay daughters [20,21].

PAHs (Polycyclic Aromatic Hydrocarbons) Measurements

Water samples (2.5-L) were collected in glass bottles at the water surface and 50 cm below water level from the six different sites on Abu Zenima and Abu Rudeis. The bottles were covered with screw caps and the samples were immediately transported to the laboratory for analysis. Water samples were filtered to remove sand and debris. Sediment samples (about 2kg for each sample) were taken from the same locations and time for water sampling at a depth 5cm of sediment surface. The water was removed from the sediments by decantation and then transferred to the laboratory. Samples were air dried in dark for 48 hours before analysis. Marine organisms (Bivalves, coral reef and star fish) were caught by fishermen three representative samples from the different sites at Abu Zenima and Abu Rudeis at the same times as water and sediment sampling.

They were transport to the laboratory. Standard solutions of reference materials were prepared in hexane. A stock solution containing the following PAHs was used for quantization: naphthalene, Acenaphthalene, Acenaphthylene, fluorene, and phenanthrene, Fluoranthene by dilution to create a series of calibration standards of PAHs at 0.1, 0.25, 0.5, 1.0, 5.0 and 10μg/ml. All solvents used in this study were of HPLC grade and were purchased from Alliance Bio, USA. Anhydrous Sodium sulfate and potassium hydroxide were of analytical grade PAHs were analyzed by using HPLC with fluorescence detection at at central lab , National Research Center, Egypt. The mobile phase was a mixture of acetonitrile and water with a gradient concentration mode of acetonitrile. The flow rate was 1mL/min. The time program of the fluorescence detector was set to detect at optimum excitation and emission wavelength for each PAHs according to laboratory detection method [22,23].

Results and Discussion

Radioactivity and Radionuclide in Water and Sediment in Coastline in Abu Zenima and Abu Rudeis

(Table 1) revealed the activity concentrations of, ²³⁸U, ²³²Th and 40K for sea water coastline in three sites at Abu Zenima1, Abu Zenima2 and Abu Zenima3. The activity concentrations of 238U, 232Th and 40K in seawater at site of Abu Zenima1 19.6mBq/L, 16.8mBq/L and $13^{40}.7mBq/L$ respectively. On the other hand ²³⁸U, ²³²Th and ⁴⁰K recorded in Abu Zenima2 highly activity 24.6mBq/L, 20.8mBq/L and 1448.7 mBq/L respectively. Also in site of Abu Zenima3 238U, 232Th and 40K were recorded 25.2mBq/L, 22.6 mBq/L and 1270.4mBq/L respectively. On the other hand (Table 2) reveled 238U, 232Th and 40K in sea water at Abu Rudeis1 recorded 10.2mBq/L, 7.33mBq/L and 632.5mBq/L respectively. Also 238U, 232Th and 40K recorded in sea water at Abu Rudeis2 9.47 mBq/L 13.9 mBq/L and 735.3 mBq/L respectively. Also (Table 3) revealed levels of ²³⁸U, ²³²Th and 40K in sea water at Abu Rudeis (Tables 4 & 5) 14.25mBq/L , 19.3mBq/L and 946.4mBq/L. Regarding to the present data and in compared to International permissible limits for NORMS radionuclide's in (Table 3), where data in the present study for ²³⁸U, and ²³²Th lay under the International permissible limits for NORMS radionuclide's in red sea 25mBg/L and 36mBg/L for ²³⁸U, and ²³²Th respectively. On the other hand El Afifi et al. [24] was recorded high levels activity concentration for ²³⁸U, ²³²Th and ⁴⁰K in the TE-NORM waste at four different sites for petroleum and gas production in Egypt, in the South Sinai Governorate, in the Suez Gulf area, in the Matrouh Governorate, Abu Rudeis onshore oil and gas field and Gabal El Zeit offshore oil and gas field at the Suez Gulf. The results showed that the average activity concentrations of ²²⁶Ra changed between 5.9 and 68.9kBq/kg (dry weight) in the waste samples from Gabal El Zeit and Abu Rudeis fields, respectively. In Gabal El Zeit field, the lower activity concentrations (28.6kBq/kg) of ²²⁶R were found in granular sample, while higher values (56.6kBq/kg) were found in the massive samples [25,26]. Regarding the (Table 1) reveled sediment concentration of ²³⁸U, ²³²Th and ⁴⁰K for sea water coastline at Abu Zenima1, Abu Zenima2 and Abu Zenima3, where ²³⁸U, ²³²Th and ⁴⁰K recorded in sediment coastline at Abu Zenima1 22.5Bq/kg-1, 24.53Bq/kg-1 and 1658.6Bq/kg-1 respectively? On the other hand Abu Zenima 2 recorded highly concentration in where ²³⁸U, ²³²Th and ⁴⁰K in costal sediment than Abu Zenima1, where concentrations levels 26.3Bq/kg-1, 28.22Bq/kg-1 and 1680.5Bq/kg-1 respectively. Also Abu Zenima 3 recorded moderate levels for ²³⁸U, ²³²Th and ⁴⁰K in costal sediment. Also (Table 5) revealed sediment concentration ²³⁸U, ²³²Th and ⁴⁰K for sea water coastline at Abu Rudeis (1), (2) and (3) sites. The Abu Rudies (3) site recorded highly concentration 238 U, 232 Th and 40 K than Abu Rudeis (1), (2), at levels 18.68Bq/ kg-1, 23.3Bq/kg-1 and 1240.3Bq/kg-1 respectively. These results were confirmed with Uosif et al. [27], who noticed highly activity

in ^{232}Th and ^{40}K in beach sediment at Suez gulf. On the other hand (Table 4) highly activity in ^{232}Th and ^{40}K in beach sediment at Suez gulf as compared to North east coast of Tamilnadu, India and other studies in different beaches of the world. The Egyptian beach sediments recorded $^{238}\text{U},\,^{232}\text{Th}$ and ^{40}K 26.53Bq/

kg-1, 177Bq/kg-1 and 815Bq/kg-1 respectively. Many causes for increased the level of concentration ^{238}U , 232Th and ^{40}K in Red sea coastline. The petroleum industries in red sea coastline are considered the highly important factors for increased activity concentration in Red sea coastline, the scales and petroleum

sludge contains many radioactive nuclides and highly concentration NORMS and TE-NORMS.

Table 1: Concentration of ²³⁸U, ²³²Th and ⁴⁰K in seawater and coastal sediments collected from different locations along the Abu Zenima coastline.

			Sit	tes		
Radionuclide's	Abu Ze	nima 1	Abu Ze	enima 2	Abu Zenima 3	
	Water (mBq/L)	Sediment (Bq/kg ⁻¹)	Water (mBq/L)	Sediment (Bq/kg ⁻¹)	Water (mBq/L)	Sediment (Bq/kg ⁻¹)
²³⁸ U	19.6±0.42	22.5±0.61	24.6±0.54	26.3±0.41	25.2 ±0.31	29.6±0.48
²³² Th	16.8 ±0.22	24.53±0.82	20.8±0.62	28.22±0.58	22.6±0.24	27.5±0.44
⁴⁰ K	1340.7±3.22	1658.6±4.91	1448.7±3.8	1680.5±5.2	1270.4±2.3	1545.8±3.9

Table 2: Concentration of ²³⁸U, ²³²Th and ⁴⁰K in seawater and coastal sediments collected from different locations along the Abu Rudeis coastline.

	Sites									
Radionuclide's	Abu Ze	nima 1	Abu Ze	nima 2	Abu Zenima 3					
nauronacinae s	Water (mBq/L)	Sediment (Bq/kg ⁻¹)	Water (mBq/L)	Sediment (Bq/kg ⁻¹)	Water (mBq/L)	Sediment (Bq/kg ⁻¹)				
²³⁸ U	10.2±0.22	12.3±0.24	9.47±0.18	13.46±0.16	14.25 ±0.17	18.68±0.21				
²³² Th	7.33±0.14	14.63±0.17	13.9±0.24	16.44±0.28	19.32±0.19	23.33±0.32				
⁴⁰ K	632.5±2.4	789.4±2.2	735.3±3.1	822.94±3.5	946.4±3.7	1240.3±4.1				

Table 3: International permissible limits for NORM's radionuclides 238U, 232Th and 40K in sea watermBq/L (EPA).

Logation	N	Mean activity concentration (mBq/L)						
Location	²³⁸ U (mBq/L)	²³² Th(mBq/L)	⁴⁰ K(mBq/L)					
The Mediterranean Sea	22	30	350					
The Caribbean Sea	25	35	320					
The Black Sea	27	37	275					
The Red Sea	25	36	325					
The Baltic Sea	30	33	400					

Table 4: Comparison of activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in beach sediment samples of North east coast of Tamilnadu, India and other studies in different beaches of the world.

	Maa			
Location	Mea	n activity concentration(Bq	/ Rgj ⊤	Reference
2000000	²³⁸ U	²³² Th	⁴⁰ K	
World	370	25	25	UNSC EAR
India	327.6	63.83	28.67	UNSC EAR
Beach sand Egypt	815	177	-	Uosif et al.
Beach sand Read sea coast Egypt	338	7.2	23.1	Harb
Hungary	302.4	27.96	28.67	UNSC EAR
Kuwait	22.7	6	36	Saad, Al-Azmi
Nigeria	35	24	16	Arogunjo et al.
Kalpak kamin Tamilnadu India	351	1455.8	112	Kannan et al.
Ullal in Karna taka, India	158	158	374	Radhakrishna et al.
North east coast of Tamilnadu , India	274.87	24.52	7.82	Ramasamy et al.

Table 5: Concentration of ²³⁸U, ²³²Th and ⁴⁰K in marine organisms collected from different locations along the Abu Zenima coastline.

Radio	Sites									
nucli des		Abu Zenima	1		Abu Zenima 2			Abu Zenima 3		
238U	10.55 ±0.21	15.86 ±0.32	18.52±0.24	9.21±0.13	21.37±0.25	19.63±0.28	8.22±0.11	24.48 ±0.29	20.43±0.21	
232 Th	9.2 ± 0.22	19.86 ±0.36	22.36±0.29	8.22±0.14	24.85±0.34	25.73±0.65	8.9±0.12	22.59±026	31.21±0.92	
40K	355.7 ±2.7	575.9 ±3.8	649.74±4.1	325.5±0.98	650.8±3.9	678.6±4.2	285.9±2.3	722.5±4.4	583.79± 1.13	

By the way many companies in past time follow the regulations and criteria for radiation safety regarding waste of NORMS and TE- NORMS. These regulation which settled by UNSCEAR, and ICRP and IEAE. After year of 2012 most of petroleum companies have many problems in cost for preservation of radioactive waste [28]. The decreased price of petroleum oil effect on regulation issues especially environmental safety and ecomarine safety. So most of the petroleum companies in Red sea coastline discharge the sludge and oil waste in sea water. Also many petroleum accident take place in these industries, which resulting of discharged the huge of petroleum oils in sea water. On the other hand, cracks in petroleum pipelines under sea water led to discharged oils and formed oils spots in coastline of sea water.

Gazineu, Hazin [29], were recorded average activity concentration of ²²⁶Ra, 232Th and ⁴⁰K in the scale, sludge and sand samples collected from disposal wasted petroleum pipes concentration of soil (33 and 45 Bqkg_1 for ²³⁸U and 232Th series respectively) which indicates its contamination with radionuclide from TENORM in the wasted petroleum pipes. ²²⁸Ra activity concentration is ranged from 32 to 50 kBq kg_1 for Scale and from 1 to 1.9kBq kg_1 for sludge. The sand samples have average ²²⁸Ra activity concentration equal to 0.042kBqkg_1 which within the worldwide average [30]. Many oil-field brines are particularly rich in chloride, enhancing the solubility of other elements including the radioactive element radium. Radium concentrations tend to be higher in more saline water [31]. Some of this saline, radium bearing water is also extracted with the

oil and gas. Some radium and radium daughter compounds are slightly soluble in water and may become mobilized when this production water is brought to the surface [32].

Radioactivity and Radionuclide for Marine Organisms in Coastline at Abu Zenima and Abu Rudeis Sites

(Table 5) revealed the ²³⁸U, 232Th and ⁴⁰K concentration in marine organisms Bivalves, coral reef and star fish at red sea coastline in Abu Zenima sites. The concentration of ²³⁸U, 232Th and ⁴⁰K in Bivalves in Abu Zenima1 10.55Bq/kg, 9.2Bq/ kg and 355.7Bq/kg respectively. Also the coral reef recorded in Abu Zenima 1 15.86Bq/kg, 19.86 and 575Bq/kg respectively. The star fish was recorded 18.52Bq/kg, 22.36 and 649.7 in Abu Zenima 1. On the other hand (Table 5) reveled the highly concentration for ²³⁸U, 232Th and ⁴⁰K in coral reef and star fish at Abu Zenima 2 and Abu Zenima 3, where coral reef recorded in Abu Zenima2 21.3Bq/kg, 24.85Bq/kg and 650.8 respectively. Also coral reef in Abu zenima3 recorded 24.48Bq/kg, 22.59Bq/ kg and 722.5Bq/kg respectively. On the other hand (Table 6) revealed marine organisms Bivalves, coral reef and star fish Abu Rudeis sites recorded low concentration activity in 238U, 232Th and 40K than Abu Zenima sites. The coral reef and star fish was recorded highly activity concentration than Bivalves in ²³⁸U, 232Th and ⁴⁰K where coral reef in Abu Rudeis1 11.34Bq/kg , 12.61Bq/kg and 346.81Bq/kg respectively, and in Abu Rudeis2 11.37Bq/kg, 13.85Bq/kg and 396.83 Bq/kg respectively, while more activity concentration for ²³⁸U, 232Th and ⁴⁰K in coral reef was recorded in Abu Rudeis3 12.22Bq/kg, 15.49Bq/kg and 466.6Bq/kg respectively.

Table 6: Concentration of ²³⁸U, ²³²Th and ⁴⁰K in marine organisms collected from different locations along the Abu Rudeis coastline..

Radio	Sites									
nucli des		Abu Zenima	1		Abu Zenima 2			Abu Zenima 3		
238U	10.55 ±0.21	15.86 ±0.32	18.52±0.24	9.21±0.13	21.37±0.25	19.63±0.28	8.22±0.11	24.48 ±0.29	20.43±0.21	
232 Th	9.2 ± 0.22	19.86 ±0.36	22.36±0.29	8.22±0.14	24.85±0.34	25.73±0.65	8.9±0.12	22.59±026	31.21±0.92	
40K	355.7 ±2.7	575.9 ±3.8	649.74±4.1	325.5±0.98	650.8±3.9	678.6±4.2	285.9±2.3	722.5±4.4	583.79± 1.13	

Table 7: International permissible limits for ²³⁸U, ²³²Th and ⁴⁰K (Bq/kg dry weight) in some marine biota in sea water, IEAE (1990), US,EPA.

	Activity concentrat	cions of ²³⁸ U, ²³² Th and ⁴⁰ K (mBq/kg dry weight)	
Locations	Species	²³⁸ U	²³² Th	⁴⁰ K
		Marine	organisms	
	Fish (Engraulis encrasicolus)	22	43	101
	Sepia Officianalis	35	38	125
	Crab	30	45	112
	Diatom Amphora	18	47	120
The Mediterranean Sea	(Om-Elkhlol "Donax trunculus	9	55	135
	Gandofelly "Tapes decussates	25	60	140
	Bivalves	18	32	196
	Coral reef	23	33	188
	Star fish	15	22	179
	Fish (Engraulis encrasicolus)	26	35	210
	Sepia Officianalis	15	22	255
	Crab	18	33	240
	Diatom Amphora	15	21	222
The red sea	(Om-Elkhlol "Donax trunculus	13	22	287
	Gandofelly "Tapes decussates	22	18	270
	Bivalves	18	37	214
	Coral reef	24	33	176
	Star fish	16	25	188
	Fish (Engraulis encrasicolus)	29	35	288
	Sepia Officianalis	23	37	320
	Crab	25	23	310
	Diatom Amphora	22	25	355
Baltic sea	(Om-Elkhlol "Donax trunculus	18	33	345
	Gandofelly "Tapes decussates	16	26	322
	Bivalves	19	27	366
	Coral reef	22	34	295
	Star fish	18	28	325

On the other star fish recorded highly activity concentrations in ²³⁸U, 232Th and 40K at Abu Rudeis3 than Abu Rudeis1 and Abu Rudeis2, 19.82Bq/kg, 22.21Bq/kg and 520.4Bq/kg respectively. Regarding to our results and compared to International permissible limits for ²³⁸U, 232Th and 40K (Bq/kg dry weight) in some marine biota in sea water. Our results confirmed with IEAE [33] and US EPA [34] in (Table 7) where revealed the activity concentration in red sea of ²³⁸U, 232Th and 40K for many biota and marine organisms like Fish (Engraulis encrasicolus), Sepia Officianalis, Crabs, Diatom Amphora, (Om-Elkhlol "Donax trunculus, Gandofelly "Tapes decussates, Bivalves, Coral reef and

Star fish. Bivalves were recorded activity concentration for $^{238}\text{U}, 232\text{Th}$ and 40K 18Bq/kg, 37Bq/kg and 214Bq/kg respectively. Also coral reef and star fish in red sea were recorded 24Bq/kg, 33Bq/kg, 176Bq/kg. 16Bq/kg, 25Bq/kg and 188Bq/kg respectively. When compared the activity concentration of 40K in marine biota Bivalves, Coral reef and Star fish at red sea coastline in Abu Zenima and Abu Rudeis sites, the highly activity and increased concentration of 40K may be due to the petroleum industry in marine and petroleum platforms. This platform discharged huge amount of petroleum waste in red sea coastline.

On the other hand our results in (Tables 4 & 6) revealed the marine organisms have different concentration in 238U, 232Th and 40K. The Bivalves recorded low concentration for 238U, 232Th and 40K, but coral reef and star fish recorded highly concentration for 238U, 232Th and 40K in Abu Zenima and Abu Rudeis sites. The morphological structures and also chemical structures of Bivalves, where Bivalves have smooth surfaces from two sides and have no pores and have no pits on exoskeleton of their body. The morphological structures gained it more resistance against accumulation of oils waste, also the smooth exoskeleton prevents discharged the waste into internal organs for organism. On the other hand coral reef and star fish have roughly surface and contain more small pores on exoskeleton. Also the coral reef and star fish contain many pits and spiny exoskeleton. Morphological structures for coral reef and star fish led to make preservation of these organisms with huge amount form petroleum oil waste. So the elimination process from oil waste in coral reef and star fish become more difficult than any other types of marine organisms. By the way elimination and biodegradation process of 238U, 232Th and 40K and other radioactive waste become very slowly and take more time. This data conformed to Khan [35], who studies the distribution of natural radio nuclides like 226Ra, 228Ra, 40K, 238U and manmade radio nuclides in the dietary sources such as fish, crab, prawn, salt and drinking water from the aquatic environment. The natural radio nuclides activity concentrations of in 238U,

232Th and 40K 14.9,22, 641Bq kg-1 in marine samples fishes, Crab and Prawn. Among different type of marine fishes analysed, the maximum ²²⁶Ra activity of 22Bqkg-1 was observed in the fish Sardinella sp. and Liognathus sp. was found to accumulate the highest ²²⁸Ra activity level [36]. Among the shells of animals tested, Crustacean exoskeleton registered relatively higher level of 238U, 232Th and 40K as compared to the shell of molluscan species. This is attributed to its chemical nature [37].

Poly Aromatic Hydrocarbons PAHs for Seawater and Coastal Sediments at Abu Zenima and Abu Rudeis

Polycyclic aromatic hydrocarbons (PAHs) are known as prevalent contaminants in marine environment. (Tables 8 & 9) showed the distribution and concentration levels of Poly aromatic hydrocarbons PAHs found in seawater and coastal sediments in Abu Zenima and Abu Rudeis coastline. Our results revealed that increased concentration levels of PAHs in Abu Zenima sites than Abu Rudeis sites. The data recorded that Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene in sea water at Abu Zenima1 34.6, 22.8, 18.4, 12.13, 9.23 and 15.3ng/L respectively. On the other hand Abu Zenima2 and Abu Zenima3 recorded highly levels for PAHs, Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene 95.3, 37.58, 66.54, 22.16, 21.8 and 34.78ng/L in Abu Zenima2 respectively; 135.68,45.37, 95.56, 35.27, 40.58, 55.38ng/L in in Abu Zenima3 respectively [38].

Table 8: Distribution and concentration levels of Poly Aromatic Hydrocarbons PAHs found in seawater and coastal sediments in Abu Zenima coastline

		Sites									
PAHs	Abu Ze	enima 1	Abu Ze	enima 2	Abu Ze	enima 3					
	Sea water ng/L	Costal sediments ng/g dw	Sea water ng/L	Costal sediments ng/g dw	Sea water ng/L	Costal sediments ng/g dw					
Naphthalene C10H8	34.6±1.2	44.3±2.42	95.3±4.62	122.5±5.33	135.68±6.4	155.25±6.8					
Acenaphthylene C12H8	22.8±0.92	28.5±0.83	37.58±1.2	64.22±2.93	45.37±2.2	86.49±3.1					
Acenaphthene C12H10	18.4±0.73	55.2±2.35	66.54±3.7	73.25±2.84	95.56±2.73	112.35±3.86					
Fluorene C13H10	12.13 ±0.8	18.37 ±1.13	22.16±0.92	28.36±1.34	35.27±1.45	43.62±1.67					
Phenathrene C14H10	9.23±0.45	16.74±0.73	21.8±1.9	33.59±1.44	40.58±2.26	55.93±2.41					
Fluoranthene C16H10	15.3±0.93	25.64±1.35	34.78±1.6	40.22±1.63	55.38±1.55	79.24±2.11					

<u>Table 9:</u> Distribution and concentration levels of Poly Aromatic Hydrocarbons PAHs found in seawater and coastal sediments in Abu Rudeis.

	Sites									
PAHs	Abu Ze	enima 1	Abu Ze	nima 2	Abu Ze	enima 3				
171113	Sea water ng/L	Costal sediments ng/g dw	Sea water ng/L	Costal sediments ng/g dw	Sea water ng/L	Costal sediments ng/g dw				
Naphthalene C10H8	22.2±0.9	34.5±1.33	45.6±1.62	62.34±1.83	76.21±2.3	92.57±2.7				
Acenaphthylene C12H8	11.8±0.53	14.3±0.44	22.33±0.82	33.52±1.11	25.68±1.21	37±1.25				
Acenaphthene C12H10	8.42±0.31	22.73±0.84	28.35±0.96	42.29±1.25	53.78±1.42	65.43±1.85				
Fluorene C13H10	6.75 ±0.21	9.44 ±0.32	12.63±0.42	15.79±0.53	16.86±0.58	22.43±0.77				
Phenathrene C14H10	4.69±0.26	9.55±0.28	12.39±0.29	15.64±0.31	22.47±0.37	29.77±0.41				
Fluoranthene C16H10	8.33±0.32	13.62±0.36	18.73±0.42	26.73±0.43	32.48±0.48	44.82±.0.51				

On the other hand and regarding to results in (Table 9) showed concentration levels of Poly Aromatic Hydrocarbons PAHs found in seawater at Abu Rudeis. The Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene in Abu Rude is 1 were recorded levels 22.2, 11.8, 8.42, 6.75, 4.69, 8.33ng/L respectively. on the other hand (Table 9) showed highly concentration levels of PAHs in seawater at Abu Rudeis2 and Abu Rudeis3 were Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene recorded in Abu Rudeis2 45.6, 22.33, 28.35, 12.63, 12.39 and 18.73 ng/L respectively; also Abu Rudeis3 were recorded 76.21, 25.68, 53.78,16.86, 22.47 and 32.48 respectively. Spilled petroleum products are the largest single source of PAHs. Crude oils contain up to 10 percent PAHs, while the PAH content of shale oils and coal-derived synthetics can be as high as 15 percent. Incomplete combustion of wood and fossil fuels are important sources, as are incineration of garbage, steel and coke production, coal liquefaction, and coal gasification.

Although most emissions stem from human activities, there are some natural sources such as microorganisms that are known to produce small amounts of PAHs. Produced water from both oil and gas platforms contains PAHs. Taking into account the large volumes of produced water discharged from oil production, the yearly input of PAHs into the environment, even from a single offshore oil field, may be significant. These results was found to be in agreement with that observed by Omayma [39], who observed that the petroleum company's takes water from Suez bay and mixed it with fresh water to utilize in washing the crude

oil and different cooling purposes, some oily smuggling occurs for water. The oily water is characterized by that they contain a high proportion of the oils which is caused by refinery production units (Coking- distillation- oils), liquidation warehouses crude and petroleum products, water waste companies neighboring filter wards pumps, Trenchant grids crude and petroleum products. The increased of PAHs was due to found the more petroleum operations in these sites. On the other hand the treatment plant cannot process in the company anymore, which is directly (untreated) discharged into Abu Zenima sites. Also the conjugated crude and petroleum products, which have TENORM and PAHs may be increased for marine environmental pollutants stress.

The sediments and biota, Bivalves, coral reef and starfish were affected by different concentration of PAHs. (Tables 10 & 11) showed an increase in PAHs Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene at Abu Zenima sites than Abu Rudeis sites. The sediments in Abu Zenima1 were recorded for PAHs Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene 44.3, 28.5, 55.2, 18.3, 16.74 and 25.64ng/g dw respectively as compared to Maximum allowable concentrations (Table 12). On the other hand PAHs Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene in Abu Zenima2 and Abu Zenima3 sites were recorded 122.5, 64.22, 73.25, 28.36, 33.59 and 40.22ng/g dw respectively;155.25, 86.49, 112.35, 43.62, 55.93 and 79.24ng/g dw respectively as compared to Maximum allowable Concentrations (Table 12).

(Table 9) showed also the concentration in the costal sediments Fluorene, Phenanthrene and Fluoranthene at Abu Rudeis1 34.5, for PAHs Naphthalene, Acenaphthylene, Acenaphthene, 14.3, 22.73, 9.44, 9.55 and 13.62ng/gdw respectively.

Table 10: Distribution and concentration levels of PAHs found in marine organisms of different sampling sites in Abu Zenima coastline.

PAHs					Abu Zenima 2		Abu Zenima 3		
μg g-1 dry weight	Bivalves	coral reef	star fish	Bivalves	coral reef	star fish	Bivalves	coral reef	star fish
Naphth alene C10H8	6.4±0.33	8.68±0.54	12.3±0.74	9.65±0.63	9.22±0.61	13.7±0.81	12.9±0.72	15.3±1.2	18.7±1.1
Acenaph thylene C12H8	1.2±0.02	3.2±0.13	6.25±1.1	3.8±0.8	7.24±0.9	11.4±1.3	4.8±0.62	9.2±0.73	12.3±0.92
Acenaph thene C12H10	3.2±0.2	2.55±0.11	5.11±0.7	7.8±0.4	8.9±0.6	7.9±0.4	5.3±0.22	8.8±0.6	13.6±0.8
Fluorene C13H10	7.4±4.6	15.3±1.2	18.7±0.9	9.5±0.33	11.2±0.8	19.3±1.2	14.7±0.7	22.6±0.8	28.4±1.3
Phenath rene C14H10	4.4±0.07	7.9±0.3	10.2±0.6	11.7±0.5	13.8±0.3	17.2±0.8	16.8±0.7	19.4±0.4	22.6±1.2
Fluoran thene C16H10	1.9±0.03	2,6±0.02	4.7±0.2	3,5±0.1	4.6±0.3	6.8±0.5	9.3±0.4	12.3±0.7	16.8±0.8

Table 11: Distribution and concentration levels of PAHs found in marine organisms of different sampling sites in Abu Rudeis coastline.

PAHs					Abu Zenima 2			Abu Zenima 3		
μg g-1 dry weight	Bivalves	coral reef	star fish	Bivalves	coral reef	star fish	Bivalves	coral reef	star fish	
Naphth alene C10H8	1.28±0.02	1.63±0.03	2.14±0.01	2.43±0.05	5.42±0.08	7.21±0.26	3.79±0.06	9.35±0.37	11.22±0.28	
Acenaph thylene C12H8	0.79±0.02	0.98±0.01	1.13±0.04	1.79±0.05	1.96±0.04	3.15±0.07	5.45±0.03	7.26±0.06	7.94±0.04	
Acenaph thene C12H10	0.21±0.01	0.52±0.02	0.73±0.04	0.94±0.03	0.85±0.02	1.33±0.08	1.62±0.05	1.96±0.08	2.37±0.07	
Fluorene C13H10	1.31±0.07	3.68±0.22	5.92±0.43	8.33±0.62	10.17±0.41	12.15±0.72	9.48 0.25	11.72±0.57	14.63±0.78	
Phenath rene C14H10	1.73±0.04	3.12±0.05	6.47±0.48	3.56±0.44	6.97±0.37	8.84±0.39	5.28±0.46	9.34±0.68	12.39±0.52	
Fluoran thene C16H10	0.28±0.024	0.59±0.028	0.85±0.053	0.46±0.027	0.89±0.046	1.26±0.22	0.89±0.42	1.94±0.47	2.64±0.79	

By the way Abu Rudeis2and Abu Rudeis3 sites were recorded 62.34, 33.52, 42.29, 15.79, 15.64 and 26.73ng/g dw respectively; 92.57, 37, 65.43, 22.43, 29.77 and 44.82ng/g dw respectively. These results was found to be in agreement with that observed by Hussain et al. [39], who observed the relatively high concentrations of Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene at certain locations Suez Gulf Abu Zenima sites. In the present study PAHs individuals for sediment at Abu Zenima sites (Naphthalene, Acenaphthene,

Acenaphthylene and Fluoranthene were recorded highly concentration than other PAHs individuals. In this study and when the results compared to the national environmental standard, it was noticed that the low concentration in total and individuals PAHs. This result was found to be in agreement with that observed by El Nemr [40] who recorded that PAHs in the sediments of the Suez Gulf may produce adverse effects on certain organisms living in and coming into contact with the sediments. However, the individual PAH concentrations in this

study were lower than the national sediment quality criteria proposed by the USEPA [41] for Fluoranthene (3000ng g-1), Acenaphthylene (2400ng g-1) and phenanthrene (2400ng g-1).

The present work also monitoring the marine organisms (Bivalves, coral reef and starfish) in coastline at Suez Gulf in Abu Zenima and Abu Rudeis sites. (Tables 10 & 11) showed the Distribution and concentration levels of PAHs individuals in Abu Zenima and Abu Rudeis sites. (Table 10) showed the increased PAHs individuals, Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene in starfish and coral reef than Bivalves. On other hand PAHs individuals recorded highly concentration in Abu Zenima3 and Abu Zenima2, than Abu Zenima1. Where levels was recorded in Abu Zenima1 for concentrations of Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene in Bivalves 6.4, 1.2, 3.2, 7.4, 4.4 and 1.9µg g-1 dry weight respectively. On the other hand coral reef and starfish recorded highly concentration than Bivalves, where levels reaching to 8.68, 3.2, 2.55, 15.3, 7.9 and 2,6µg g-1 dry weight respectively; 12.3, 6.25, 5.11, 18.7, 18.7 and 4.7µg g-1 dry weight respectively. The Bivalves, coral reef and starfish in Abu Zenima3 and Abu Zenima2 recorded highly levels in concentrations for Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene, where Bivalves recorded in Abu Zenima2 9.65, 3.8, 7.8, 9.5, 11.7 and 3.5 µg g-1 dry weights respectively. Also coral reef and starfish were recorded in Abu Zenima2 9.22, 7.24, 8.9, 11.2, 13.8 and 4.6µg g-1 dry weight respectively; 13.7, 11.4, 7.9, 19.3, 17.2 and 6.8µg g-1 dry weight respectively. On the other hand Bivalves were recorded in Abu Zenima3 for Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene 12.9, 4.8, 5.3, 14.7, 16.8 and 9.3 µg g-1 dry weights respectively.

The coral reef and starfish also recorded15.3, 9.2, 8.8, 22.6, 19.4 and 12.3 μ g g-1 dry weight respectively; 18.7, 12.3, 13.6, 28.4, 22.6 and 16.8 μ g g-1 dry weight respectively. When compared our results with the data of Maximum allowable Concentrations (MACs) USEPA: 1993 in (Table 12), where recorded for Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene 0.007, 0.008, 0.007, 0.001, 0.009 and 0.009 μ g/kg dry weight respectively. The concentration levels of PAHs individuals in Abu Zenima sites were recorded higher than that in Maximum allowable Concentrations (MACs) USEPA for PAHs in marine organisms

[41]. These results was found to be in agreement with Al Deep et al. [42], who was observed the accumulation of total PAHs were more pronounced in the tissues of higher lipids contents, which increase in the large and old individuals, and they found that the coral reef and starfish comes second regarding to their PAHs accumulations ratios after the dwelling fish species.

Size of the marine organisms may be not only contributed to change the accumulation factors responsible for the quantities of pollutants such as time exposure and lipids contents but also in the factors responsible for the pollutants selectivity. For example, Maioli et al. [43] demonstrated that coral reef and starfish of greater size accumulated PAHs of low molecular mass, whereas the smaller mussels had accumulated greater concentrations of high molecular mass PAHs Metabolism may explain this pattern, because it is suspected that PAH of High Molecular Mass (HMW) are more rapidly metabolized than Low Molecular Mass (LMW) due to differences in enzyme affinity [44]. The increased of concentration levels of PAHs individuals in Bivalves , coral reef and starfish at Abu Zenima were be attributed to the presence of many petroleum industrial marine plates in this region at south canal. Also this petroleum plates discharge some oils into the water plus to their incomplete fuel combustion emissions [45].

On the other hand (Table 11) showed the low level concentration of PAHs individuals in Abu Rudeis sites as compared to Abu Zenima sites in (Table 10) and Maximum allowable Concentrations (MACs) USEPA for PAHs in marine organisms in (Table 12). The Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene and Fluoranthene were recorded in Bivalves at Abu Rudeis1 1.28, 0.79, 0.21, 1.31, 1.73 and 0.28 µg g-1 dry weights respectively. The impact of PAHs individuals and TE- NORM 238U, 232Th and 40K in marine organisms make dangerous effects on marine ecosystem. These effects were to be attributed for spreading the petroleum marine plates in these sites and areas. On the other hand many of these petroleum industries do not have following environmental regulations and marine protection standards during process for removal petroleum waste. Also during the process of oil transportations and drilling for sludge and oil tanks. The accumulation of double effects of and also many effects of hazardous materials like TE- NORM 238U, 232Th, 40K and PAHs individuals may be devastation the marine ecosystems in red sea coastline and Gulf of Suez.

Table 12: Maximum allowable Concentrations (MACs) of PAHs in water, soil and marine organisms USEPA.

РАН	MAC (water), ppm	MAC (soil), ppm	MAC marine organisms), μg/kgdry weight
Naphthalene	3.0	1.0	up to 0.007 μg/ kg
Phenanthrene	3.0	3.0	up to 0.009 μg/ kg
Fluoranthene	3.0	3.0	up to 0.009 μg/ kg
Acenaphthene	3.0	3.0	up to 0.007 μg/ kg
Acenaphthylene	3.0	3.0	up to 0.008 μg/ kg
Fluorene	3.0	3.0	up to 0.001 μg/ kg

Conclusion

The concentrations of natural radioactive series nuclides varied widely within the oil fields and from one oil field to another through spectrum of this study area. This study revealed the correlation between the effects of naturally occurring radioactive materials (NORM) 238U, 232Th and 40K on marine ecosystem. In this study, the increased of 238U, 232Th and 40K in sea water and sediments were related to increased in PAHs individuals. The 238U, 232Th and 40K in Abu Zenima 3 and Abu Zenima 2 sites were recorded higher concentration than Abu Zenima1. Also Abu Rudeis 3 and Abu Rudeis were recorded highly activity than site of Abu Rudeis1. The levels of 238U, 232Th and 40K in sea water and sediments at Abu Zenima and Abu Rudeis were parrled and synchronized with levels of 238U, 232Th and 40K in sea water and sediments at Abu Zenima PAHs individuals in both sites. The study showed the increased level concentrations of 238U, 232Th and 40K and PAHs individuals in sea water and sediments Abu Zenima sites than Abu Rudeis sites as compared to international Maximum allowable concentrations. On the other hand this study revealed the effect of (NORM) 238U, 232Th and 40K and PAHs individuals on marine organisms Bivalves, coral reef and starfish on both sites. The data revealed increased concentration of (NORM) 238U, 232Th and 40K and PAHs individuals for starfish and coral reef than bivalve's marine organisms for both Abu Zenima and Abu Rudeis sites respectively as compared to international maximum allowable concentrations. Regarding to these data, all petroleum industries in this sites must be take into consideration the roles and standers' of protection for marine ecosystem. On the other hand, the Egyptian government must be applied the regulation for marine environmental protection lows.

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