

REVIEW



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Concentrations of TENORMs in the petroleum industry and their environmental and health effects

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Crude oil and its products and wastes are among the significant sources of naturally occurring radioactive materials (NORMs). These materials may be enhanced to high levels due to technological and human activities, which are called technologically enhanced naturally occurring radioactive materials (TENORMs). Thus, the average radioactivity of these radionuclides sometimes exceeds the exemption level of $10\ 000\ \text{Bq kg}^{-1}$, which is recommended by the IAEA's safety standards. TENORMs in the oil and gas industry may generate greater radioactivity levels, which eventually represents potential environmental and health risks. This will require continuous attention by monitoring and surveillance during routine processes in the petroleum industry. In this paper, a comprehensive review of the published literature is conducted to evaluate the TENORM concentrations in the oil and gas industry. Moreover, their environmental and health hazards in different regions of the world are discussed.

1 Introduction

At present, various environments suffer from the excessive accumulation of radioactive pollutants and their hazardous results, where radionuclides naturally decay. Examples of such radionuclides are: ^{226}Ra , ^{228}Ra , ^{222}Rn , ^{210}Pb , ^{40}K etc., which decay along with many other man-made radionuclides.¹⁻⁴ Approximately 80% of human exposures to radiation comes from the radioactive sources that naturally occur, which may lead to several harmful effects on humans, animals, or the environment.⁵⁻¹⁰ Materials that contain large amounts of natural radionuclides are referred to as naturally occurring radioactive materials (NORMs). They are in the crust of the earth and we are exposed to them every day. NORMs are an integral part of the planet, our bodies, the food we eat, the air we breathe, the places we live and work, and the products we use. Treatment of some natural resources enhances naturally occurring radionuclides to the extent that they may pose risks to humans and the environment. These by-products are named as follows: technically enhanced naturally occurring radioactive materials (TENORMs). The majority of radionuclides are found in TENORMs, uranium and thorium chains.¹¹⁻¹⁴ Therefore, many non-nuclear industries are forced to take radiation protection measures. Examples of raw materials, products and waste that can cause problems for the non-nuclear industry are zircon sand used in ceramic industry;

phosphate rocks, fertilizers, slag, and phosphorus from phosphate industry; fly ash from electricity production; and metrology and sludge from oil and gas production. In these substances, the activities specified are often higher than $1\ \text{Bq g}^{-1}$, and for more fly ash and some oil/gas scales, the radioactivity can reach $1000\ \text{Bq g}^{-1}$.¹¹⁻¹⁷ In this study, the focus is on the TENORM produced in the oil and gas industry. High concentrations of background radiation in crude oil were reported for the first time by Hempstead and Burton between 1920 and 1930; also the term has been reported by Russian and German researchers.¹⁸⁻²² Preliminary assessments of occupational radiation exposure were recorded in the oil and gas industries a few decades ago, when Kolb and Wojcik discovered TENORMs in petroleum in 1985.^{3,22,23} They measured the dose rate at many oil field sites in northern Albania and determined the values of concentrations of these radionuclides: ^{228}Ra , ^{226}Ra and ^{222}Rn .^{3,20-23} The first official survey from the point of view of radiation protection was conducted in 1970s and early 1980s, when the TENORMs were detected in the North Sea Oil Platforms.²⁴⁻²⁶ Several types of "scattered TENORM wastes," such as scales, sludge, and produced water, were found in the petroleum industries, where the radium bearing scale was discovered in North Sea oil.²⁴⁻²⁷ Thus, the accumulation of radium in US oil field equipment became apparent for the first time in the 1980s, when scrap metal traders began routinely detecting unacceptable levels of radioactivity in pipeline shipments.²²⁻²⁶ Since then, many oil and gas industries have sought to effectively determine the TENORM problems in the oil fields and develop techniques for forecasting, protection, correcting, and eliminating TENORMs from the fields and facilities.^{24-26,28} The TENORMs concentrations in the wastes of the petroleum industries were studied

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and reported by many researchers from different regions of the world.

The release of radioactive particles from the radionuclides might be one of the potential bases of energy related with the conversion of the organic materials to oil in the long term, where naturally occurring radioactive elements dissolve at highly depressed concentrations through the natural interactions among soil, rock, and water.^{9,29–31} Therefore, oil fields may contain many natural radionuclides such as ²²⁸Ra, ²²⁶Ra, ²²⁴Ra, ²¹⁰Pb, and ⁴⁰K which are transported to the surface of the earth and may include high levels of radioactivity.^{23,32–34} Many oil fields are rich in chloride, thus enhancing their relative solubility. When oil is pumped from the depths of the wells to the surface, it also produces water. This produced water is extracted from oil containing volatile metal materials, some of which are free of active substances due to the presence of radium isotopes and their decay products.^{27,35–38} The initial products of the oil and gas are usually dry. When the produced water is brought to the surface, the decrease in temperature and pressure allows the use of melted soluble radioactive substances in this water.²⁷ Elements containing natural radionuclides appear in the form of complex compounds, such as sulfates, silicates, and carbonates, which are present in scale and sludge.³⁸ These sediments are formed at sufficient temperatures with oil/water pumping to the surface.^{35–37} Thus, the quantity of precipitation depends on the chemical-physical properties of this water.

The uncontrolled release of radioactivity related to TENORM levels may pollute the environment and endanger human health. Hazardous radiation may enter the human body through various ways, of exposure that are classified either internal or external, such as absorption, wounds, inhalation and ingestion.^{22,27,39,40} Controls have been adopted for TENORMs to avoid the risk of these wastes, efforts to shield radiation should be made to protect workers and the public, and any regulations set in place for radiological protection must consider the additional risks that exceed the exposure to native natural radioactivity.^{22,27,39,40} The IAEA has published the overall radiological safety criteria that depend on the recommendations of the International Commission on Radiation Protection. These criteria were also recommended for adoption by the European Commission as the basic safety standards (BSS) by all European society countries.^{20,22,25,39–41} Several countries have implemented their own regulations on TENORMs based on these recommendations. The concentrations of TENORMs in oil and gas industries,^{25,39–41} measured globally and adopted by IAEA are shown in Table 1. It is noted from the table that ²²⁶Ra is the most concentrated isotope, especially in the scales. Therefore, scales are considered one of the most dangerous waste of the petroleum industry.

Studies on the radioactivity of the oil and gas industries with a focus on ²²⁶Ra and ²²⁸Ra, in petroleum wastes reported in,^{24,25,42–47} were presented in previous literature reviews. However, this study is very detailed. On the one hand, the study focused on most natural and artificial radionuclides. On the other hand, most previous studies on the presence of TENORMs in petroleum products and wastes and samples from the environment surrounding oil and gas industry facilities have been studied. This work aims to review studies conducted on the

TENORMs radioactivity resulting from the oil and gas industries in different regions of the world in addition to its environmental and health effects. The issues mentioned in previous studies confirm that an update is necessary, and new methods of managing, evaluating, and eliminating the potential risks of TENORMs must be developed, which is part of an integrated system for the administration of professional safety.

2 Evolution of the term TENORMs

To understand the naturally occurring radioactive materials in the solids, liquids and gases generated by natural processes, many researchers have used many of the terms that symbolize these materials. The industrial enhancement of TENORMs in the petroleum industries are created in many different ways as a result of enhanced oil recovery and other industrial practices used during petroleum industry activities. For this reason, many terms that symbolize these activities and processes have evolved over time. K. Al Nabhani *et al.*¹⁹ have defined some terms used by many researchers, which include: NOR^{48–50} is the abbreviation for the description of Naturally Occurring Radionuclides, which is explained by its concentration on radioactive elements and not on the materials in which radionuclides are stored. HINAR^{51,52} is the abbreviation for the definition of High Natural Radioactivity, which is interpreted by a focus on areas affected by high natural radioactivity. NARM^{53–55} is the abbreviation for the definition of Naturally Accelerator-Produced Radioactive Materials, which is interpreted by naturally radioactive materials being artificially produced during the operation of atomic particle accelerators. ENOR^{19,56} is the abbreviation for a definition of Enhanced Naturally Occurring Radioactivity, which is interpreted by naturally occurring radioactivity technologically enhanced. TENR^{56–61} is the abbreviation for a definition of Technologically Enhanced Natural Radioactivity, which is interpreted by natural radioactivity technologically enhanced. NORM^{9,33,62,63} is the abbreviation for the definition of Naturally Occurring Radioactive Material, which is interpreted by all solid radioactive materials being created by the natural process. This definition is the most common term used by researchers and specialists. TENORM^{19,57–64} is the abbreviation for the definition of Technologically Enhanced Naturally Occurring Radioactive Materials, which is interpreted by radionuclide content of naturally radioactive materials is enhanced by manmade procedures (common in industries and highly used). This term is considered the most important radiological point of view because it distinguishes between these naturally occurring radioactive substances and those that are promoted through human activity in many industries and fields. In this study, this naming was adopted because it is more comprehensive and capable of describing radionuclides in petroleum industries.

3 TENORMs sources in the oil and gas industries

Radioactive materials have been an integral part of the earth's crust ever since the earth was formed. Where the natural



Table 1 Concentrations of natural radioactive materials in petroleum industry^{85,90,228,278}

Radioactive isotope	Hard scales (Bq kg ⁻¹)	Sludge (Bq kg ⁻¹)	Produced water (Bq L ⁻¹)	Crude oil (Bq kg ⁻¹)	Natural gas (Bq m ⁻³)
²³⁸ U	1–500	5–10	0.0003–0.1	0.0001–10	—
²³² Th	1–2	2–10	0.0003–0.001	0.3–2	—
²²⁶ Ra	100–15 000 000	5–800 000	0.002–1200	0.1–40	—
²²⁸ Ra	50–2 800 000	500–50 000	0.3–180	3–17	—
²²⁴ Ra	—	—	0.5–40	—	—
²²² Rn	—	—	—	3–17	5–200 000
²¹⁰ Pb	20–75 000	100–1 300 000	0.05–190	—	0.005–0.02
²¹⁰ Po	20–1500	4–160 000	—	0–10	0.002–0.08

radiation chains are found in nature: ^{65–68} ²³⁸U series form 99.27% of natural uranium, ²³⁵U series accounts for 0.725% of natural uranium, ²³⁴U is a very small percentage (0.005%) of natural uranium and ²³²Th series and ⁴⁰K. ^{6,69,70} These chains are present in the oil and gas fields in the ground. These radioactive materials are transported with liquids from the oil fields to the surface of the earth. ^{71–74} It's known that the atoms of these chains are unstable, which means that they undergo spontaneous self-transformation so that their atom becomes more stable. This process of transformation is called radioactive decay. Natural radioisotopes are present in oil and gas industries with varying concentrations. ^{75–77} In certain places of production and processing facilities, these isotopes accumulate and lead to enhanced levels of radioactivity. These materials are treated as a closed radiant source as long as they are within the parts of the facility, and when opening one of the parts of the facility, they are treated as an opened radiant source. ^{64,78–81} Natural radioactive materials are found in oil and gas basins where other mineral elements are in different concentrations and these materials are released with the production fluids during the extraction processes as shown in Fig. 1:

It is noted from Fig. 1 that the origin of ²²⁶Ra, ²²⁸Ra and ²²²Rn are progeny resulting from the dissolution of the parent isotopes such as ²³⁸U or ²³²Th, which is found in the geological strata below the surface of the earth, especially in the clay mud rocks, ^{81,83–85} which researchers found in previous studies. ^{50,85–87} As both uranium and thorium salts are part of these layers, they do not dissolve significantly in pelvic fluids (fresh water, salt water, oil and gas). While salts of radium dissolve in water and move from these layers to the surface of the earth with oil-related water. ^{82,87,88} The quantities of radium present in the produced water and associated with the oil depend on the nature and quantity of these rocks and their content of uranium and thorium, in addition to the physical and chemical conditions such as pressure, temperature and pH. Fig. 2 shows the recombination and distribution of natural radioactive substances in different oil liquids by extraction processes.

4 Concentration of TENORM in petroleum products

Petroleum is a broad concept and includes crude oil and its products, natural gas, and others. The term "petroleum" refers

only to crude oil but can be sometimes used to describe any solid, liquid, or gaseous hydrocarbons. ^{23,91} Crude oil is a kind of naturally flammable liquid and can be found in natural reservoirs in geological formations below the surface of the earth. Such oil consists of a complex mixture with different molecular weights of hydrocarbons and other organic liquid compounds. ^{41,91,92} Most petroleum components are fossil fuels formed when considerable dead organisms, usually zooplankton and algae, are buried under sedimentary rocks and subject to intense temperature and pressure. ^{41,92,93} Then, petroleum is brought to the surface and its components are easily separated by the boiling point property into a substantial number of consumer petroleum products. ^{23,41,91–94} These products include diesel, benzene, gasoline, jet fuel, natural gas, kerosene, naphtha, plat format, flushing oil, asphalt, green coke, sulfur, oil slope, vacuum, unconverted oil, and chemical reagents used in plastics, lotions, treatments, and others. ^{23,41,91–94} Petroleum is used in many industries, and 88 billion barrels per day are estimated to be consumed around the world.

The results of a study⁹⁷ in the United States showing low concentrations of uranium in crude oils in the range of 0.0015–0.015 ppm for uranium in crude oil is typical. In study,¹ the concentration of uranium in petroleum products in Saudi Arabia was reported to be <8.5 Bq kg⁻¹ (see Table 2 for the rest of the studies). Evidently, the concentration of uranium is particularly low in raw oil and petroleum products. The reported radium isotopes in crude oil and products are also low. For example, the concentration of ²²⁶Ra in Algerian crude oil in 95 is in the range of 6–20 Bq kg⁻¹. The maximum concentration of radium isotopes in Egyptian petroleum products was found in,¹¹¹ wherein ²²⁶Ra was in the average of 11 512.8 Bq kg⁻¹. Thus, ²³²Th, ⁴⁰K, and other radioisotopes reported in previous studies in Table 2 are at low concentrations below the minimum value set by the IAEA, WHO, EPO, and others.

5 Concentrations of TENORMs in petroleum wastes

5.1 Produced water

Petroleum reservoirs include oil, gas and water in the pores of the earth rocks, where water quantities tend to vary. ^{35,42,113} The extraction of petroleum is often accompanied by large



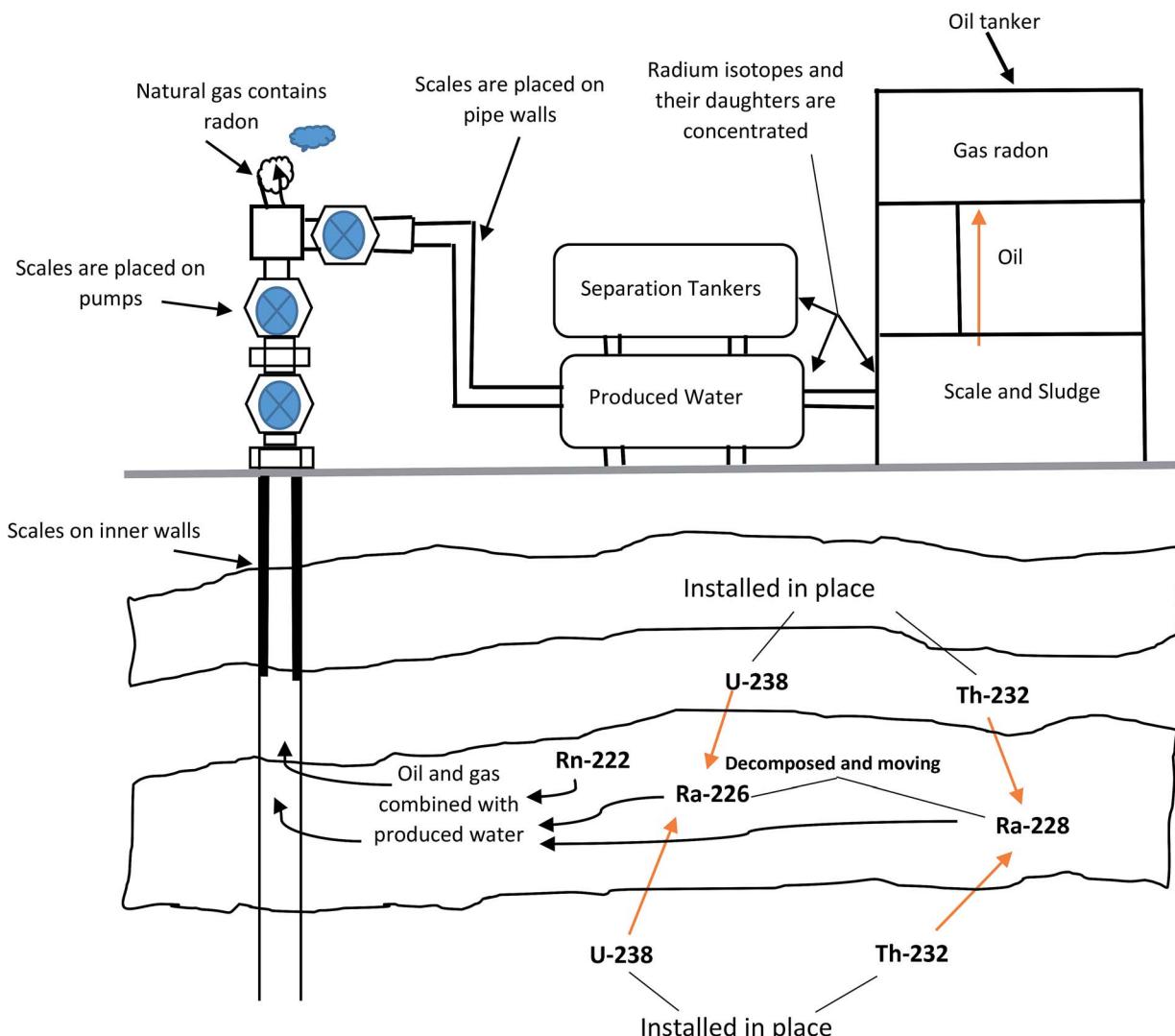


Fig. 1 The presence of natural radioactive materials in the oil industry.^{78,82,83}

quantities of such water. This water is generally referred to as “produced water,” “composition water”, or “formation water”, in oil fields.^{35,42,113,114} In many studies, gas production fields produce small amounts of produced water, while oil production fields produce large quantities of water. Thus, petroleum reservoirs contain formation water, which later becomes produced water when brought to the surface during the extraction of hydrocarbons.¹¹³⁻¹¹⁵ TENORMs are brought into the surface by the water formation that is produced when the pressure of the reservoir decreases with time while petroleum is extracted. The amount of TENORMs produced in the petroleum fields accompanying the petroleum extraction process are directly proportional to the size of produced water through the oil pumping process.³⁴ That is, the water contaminated with TENORMs is considered a major waste of petroleum, and this proportion of radioactivity concentration from produced water usually ranges between 1–10 Bq kg⁻¹.^{25,116} The American Petroleum Institute²⁹ reported that over 18 billion barrels of petroleum liquids is produced per year in the United States

compared with the gross crude oil size of 2.5 billion barrels, which is equal to 400 million cubic meters. The gross of produced water accounted for 91% of what considered to be a waste.¹¹⁷⁻¹¹⁹ Alternatives to seawater injection are the re-injection of produced water or the injection of water from nearby formations.^{20,35,44,119,120} The vast majority of produced water from the underground is usually disposed by injection into the ground either in the extraction or in the disposal wells. In marine oil boreholes seawater usually injected to preserve the pressure at a certain level to ease the production process. In addition to that the produced water is already in the hole beside the oil. In the latter case, the produced water is usually salty and consist high degree of chloride-ions which form with Radium chemical compounds, thereby Radium nuclides then transferred from geological rocks adjacent to the waters into the water itself.^{42,121-123} It is also very common that produced water might be dissolved with a mixture of organic compounds (e.g., scale inhibitors and corrosion added to hydrocarbons production, namely, residual chemical additives, organic acids, and

Radioactive Decay in Thorium and Uranium Series

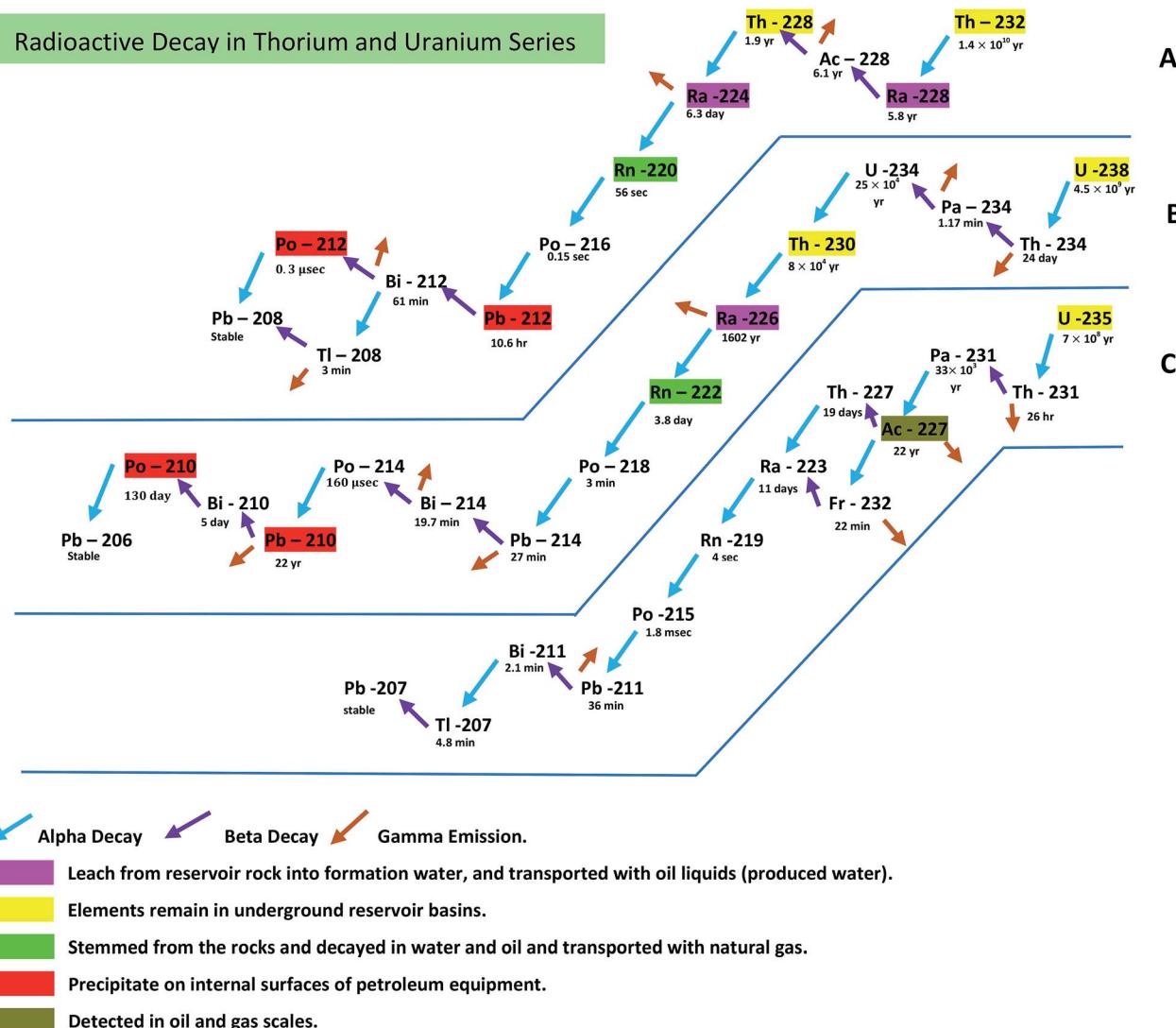


Fig. 2 Primordial radioactive decay series (A) ^{232}Th , (B) ^{238}U and (C) ^{235}U

dissolved and dispersed hydrocarbons) and inorganic compounds (e.g., suspended particles, trace minerals, and dissolved salts).^{42,115,120-124} The existing organic compounds are composed of several groups, such as polycyclic aromatic hydrocarbons and alkylate phenols, which produce different effects in various organisms. The chemical decomposition of the produced water in the petroleum industries varies from one field to another. This difference relies on several factors such as operating conditions, type of produced hydrocarbon, age of the field towards the end of its productive life and geological characteristics of the surrounding rocks.^{42,117,122,124-137} The physical characteristics of the produced water is similar to seawater, especially if seawater is used for injection. Furthermore, the produced water contains partially dissolved mineral salts, along with radon or radium. Although uranium and thorium do not normally enter the sol, the stream of the produced water could be the major waste in terms of size resulting from the petroleum industries. Thus, the main source of TENORMs in the petroleum industries is the dissolved radionuclide in fluxes

of produced water or suspended microscopic radionuclide as a result of the solubility & mobility status of U and Th, which is then transported to the surface with petroleum (Tables 3-5).

The activity concentrations of radium isotopes in most studies range from detection limits (DL) to less than 100 Bq L^{-1} . Most results appear in the lowest range even in 10 Bq L^{-1} . For example, concentrations of isotopic activity in the samples studied by^{119,130,136,139} were less than 11.1 Bq L^{-1} . Previous studies have shown that the main radionuclides involved are ^{226}Ra and ^{228}Ra . Numerous researchers, such as, ¹⁵³ reported the presence of ^{224}Ra isotope in addition to ^{226}Ra and ^{228}Ra isotopes in the produced water in the In the Syrian, Turkish, Ukrainian and Ghanaian petroleum industries. Previous studies have shown that the U and Th do not extensively migrate during oil and gas extraction. Meanwhile, ^{40}K is one of the abundant elements in produced water and reported in many studies. For example, ^{40}K was expressed in produced water in the Egyptian,⁹³ Tunisian,²³ and Nigerian^{141,142} petroleum industries. ^{137}Cs is also present in the produced water of the US and Iraqi

Table 2 Shows the range and mean values of the TENORMs concentrations in crude oil in diverse areas of the world, including recent data^a

Country	Concentrations of the TENORM in crude oil with their range, averages and units	Ref
Algeria	^{226}Ra (6–20) Bq g^{-1}	95
USA	^{238}U (0.0015–0.015) ppm, ^{226}Ra (1–40) Bq g^{-1} ^{238}U (0.0001–10), ^{226}Ra (0.1–40), ^{210}Po (0–10), ^{232}Th (0.03–2) mBq g^{-1}	96 and 97
Saudi Arabia	^{222}Rn (5–200 000), ^{210}Pb (0.005–0.02) ^{210}Po (0.002–0.08) Bq m^{-3}	105
Egypt	^{238}U LD ^a , ^{232}Th 0.30 ^a , ^{226}Ra 0.65 ^a , ^{224}Ra < LD ^a , ^{40}K 4.3 ^a Bq kg^{-1} ^{226}Ra 3.02 ^a , ^{228}Ra < 0.02 ^a , ^{40}K < 0.2 ^a Bq kg^{-1}	1
Tunisia	^{226}Ra (31–2669), ^{232}Th (<DL–913.4), ^{40}K (<DL–98) Bq L^{-1}	93
Iraq	^{226}Ra (ND–21.30), ^{232}Th (ND–10.50), ^{40}K (56.6–126) Bq kg^{-1} ^{226}Ra (<0.106–0.38), ^{232}Th (0.02–0.12), ^{40}K (0.057–1.308) Bq kg^{-1}	98
Turkey	^{238}U (15.23–33.16), ^{232}Th (5.82–19), ^{40}K (14.9–422.28), ^{137}Cs (1.17–10.64) Bq L^{-1}	99
Nigeria	^{226}Ra 33.6 ^a , ^{232}Th 13.0 ^a , ^{40}K 197.0 ^a Bq kg^{-1} ^{226}Ra (2.30–5.80), ^{232}Th (2.90–5.50), ^{40}K (5–36) Bq kg^{-1}	23
Ghana	^{224}Ra (3 to <11) 7.75 ^a , ^{226}Ra (5–16) 9.25 ^a , ^{228}Ra (2–10) 7 ^a Bq kg^{-1}	100
Sudan	^{40}K (2.24–19.73) 10.52 ^a , ^{238}U (ND–1.29) 0.80 ^a , ^{232}Th (ND–0.25) 0.17 ^a Bq kg^{-1} ^{238}U (0.000001–0.01), ^{226}Ra (0.0001–0.04), ^{210}Po (0–0.01) ^{232}Th (0.00003–0.002) Bq L^{-1}	102
	^{226}Ra (2.42–10.14), ^{40}K (21.08–34.39), ^{232}Th (11.5–12.65), Bq kg^{-1}	103
	^{238}U 64.11 ^a , ^{232}Th 63.69 ^a , ^{40}K 22.13 ^a Bq kg^{-1}	104
		106

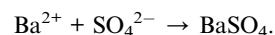
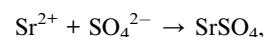
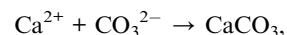
^a () the data between the brackets means the range/^a is the averages of concentration/LD = lower than the detection limits.

petroleum industries. The available information on the concentrations of nuclides ^{210}Po , ^{210}Pb , and ^{228}Th in produced water is scarce compared with Ra isotopes.¹⁴⁴ In 1997, the presence of radionuclides ^{210}Po , ^{210}Pb , ^{228}Th , ^{226}Ra , and ^{228}Ra were reported in produced water in minerals and hydrocarbons in oil and gas, discharge processes, and in environmental models associated with offshore production facilities in Texas/Louisiana in the United States. In conclusion, the amounts of TENORM in the produced water in the oil and gas industry contribute to varying levels of pollution to the environment and slightly enhanced radioactivity in the terrestrial and marine environments. This condition may affect workers, the general public, plants, the surrounding environment, and marine organisms living near oil and gas platforms.

5.2 Scales

The scale in the oil and gas industry is one of the additional wastes composed of complex materials containing geological formations.^{37,155} Such materials are often composed of alkaline earth metals, such as silicate, carbonate, and sulfate, particularly BaSO_4 , SrSO_4 , and CaCO_3 .^{37,68,129,155} These metals are produced by many physical and chemical processes, such as injection of water in reservoirs, pressure changes, temperature, evaporation in gas extraction tubes, pH balance, fluid expansion, changes in water acidity, inconsistent water mixing, different flow rates, and additives or impurities, during extraction.^{37,45,116} Radium carbonate, radium sulfate and, in some cases, radium silicate, are formed due to radium deposition with sulfur, strontium, and calcium because of the similar chemical composition of these elements.^{37,42,45,68,116,129,155} The sudden changes in pressure and temperature and the acidity of the formation water brought to the surface are factors contributing to the accumulation of scales; moreover, at the process of injecting petroleum wells, mixing the sea water rich in sulfate with the composition water rich in salt solutions also increases

the scales.^{34,129,156,157} Several studies have shown that the solubility variability of sulfates and carbonates, which can lead to the formation of the scale, is related to the physical and chemical processes in varying proportions as follows: (5%), pressure changes (10%), evaporation in extraction pipes, especially gas extraction (10%), non-compatible water injection (70%), and other factors (5%). The scale composition is influenced by the re-entry of water into the wells to maintain production compression during field exploitation.^{44,45,83} Numerous factors, such as the amount of radium found below the surface of the earth, the components that make up the water, and the applied processes during extraction, can increase radium concentrations and radon in pipelines, equipment, and wastes in the gas and oil industry. Alkaline earth metal compounds, such as sulfates and carbonates, are deposited in oil and gas production equipment and produce the scale according to the following chemical equations:



The American Petroleum Institute Dallas^{34,83,155} reported that scales in the oil and gas industry are often found inside pipes and tubes where concentrations are as high as tens of thousands of Bq g^{-1} .^{34,83,155} Therefore, the maximum concentrations of scale radioactivity are in the wellhead pipes and the production pipes near the good nozzle. However, the maximum size of the scale occurs in three areas, namely, the water lines associated with the separators (gas separated from oil and water), heater treaters (split the oil and water stages), and dehydrated gases, where sediments may accumulate in sizes up to 4 inch in thick.^{20,35,83} The mean concentration of



Table 3 Shows the range and mean values of the TENORMs concentrations in petroleum products in diverse areas of the world, including recent data^a

Country	Concentrations of the TENORM in petroleum productions with their range, averages and units	Ref
Saudi Arabia	^{238}U (<LD-8.5), ^{232}Th (<LD-0.62), ^{226}Ra (<LD-0.45), ^{224}Ra (<LD-0.598), ^{40}K (0.13–57.05) Bq kg^{-1}	1
Syria	^{224}Ra (9–14) 11.43 ^a , ^{226}Ra (4–11) 7 ^a , ^{228}Ra (3–13) 7.3 ^a , ^{210}Po (224–2371) 971.67 ^a , ^{210}Pb (12–160) 115.33 ^a Bq g^{-1} ^{210}Pb (498–4023), ^{210}Po (380–3100), ^{224}Ra (14–36), ^{226}Ra (77–135), ^{228}Ra (24–31) Bq kg^{-1}	30
Malaysia	^{238}U (13–40) 29.44 ^a , ^{232}Th (37–48) 42.56 ^a , ^{226}Ra (104–167) 141.76 ^a , ^{228}Ra (117–158) 135.84 ^a Bq kg^{-1}	108
Egypt	^{226}Ra (<0.2 to 46.78), ^{228}Ra (<0.2 to 10.50), ^{40}K (<0.2 to 17.80) Bq kg^{-1} ^{226}Ra (1.13–15.70) 8.04 ^a , ^{232}Th (3.31–23.32) 8.62 ^a , ^{110}Pb (17.16–48.55) 35.86 ^a , ^{40}K (19.24–211.25) 72.84 ^a Bq kg^{-1} ^{226}Ra (21–38), ^{232}Th (9–15), ^{40}K (303–154) Bq kg^{-1} ^{238}U 2.7 ^a , ^{232}Th 9284.1 ^a , ^{226}Ra 11 512.8 ^a , ^{210}Pb 266.4 ^a , ^{40}K 801 ^a Bq kg^{-1} ^{226}Ra (10.2–21.3), ^{232}Th (3.4–6.2), ^{40}K (66.5–121) Bq kg^{-1}	93, 107, 109, 110, 111
Kuwait	^{226}Ra (ND–17.0), ^{232}Th (ND–10.5), ^{40}K (13.6–126) Bq kg^{-1}	99
Iraq	^{238}U (1.59–6.63), ^{232}Th (DL–35.47), ^{40}K (14.16–208.50), ^{137}Cs (1.30–11.16) Bq L^{-1}	27 and 41
Tunisia	^{226}Ra 31.6 ^a , ^{232}Th 14.4 ^a , ^{40}K 387.6 ^a Bq kg^{-1} ^{226}Ra (0.07–0.64), ^{232}Th (<LD–0.16), ^{40}K (0.142–3.7) Bq kg^{-1}	100, 23
Ghana	^{226}Ra (<0.12 to 17.81), ^{40}K (<0.15 to 43.46), ^{232}Th (<0.11 to 18.19) Bq kg^{-1}	104
Venezuela	^{228}Th 112 ^a , ^{226}Ra 16 ^a , ^{212}Pb 63 ^a , ^{214}Pb 17 ^a , ^{208}Tl 27 ^a , ^{214}Bi 16.7 ^a , ^{212}Bi 70 ^a , ^{228}Ac 14 ^a , ^{232}Th 0.48 ^a , ^{232}Th 38 ^a , ^{235}U 4×10^{-4} ^a , ^{238}U 0.65 ^a , ^{40}K 20 ^a Bq kg^{-1}	112

^a () the data between the brackets means the range/^ais the averages of concentration/LD = lower than the detection limits.

radionuclides in the scale is 17.76 Bq g^{-1} (*i.e.*, approximately 18 000 Bq kg^{-1}), according to the latest estimate of the Environmental Protection Agency (EPA, 2017). This value can be much higher (up to 148 000 Bq g^{-1}) or less depending on regional geology. The scale of wells and gaseous equipment can also contain radon offspring, such as ^{210}Pb and ^{210}Po *etc.*

The survey included several studies on the scales generated in the oil and gas industry in different parts of the world. However, the scales were obtained in facilities, mostly in the section separators, oil and gas storage tanks, and pipelines. Some examples include the scales formed in the oil and gas fields in Italy, Egypt, Syria, Kazakhstan,^{20,30,128,181} and in other studies. The reported studies showed that the scale phenomenon is a result of the slow fluid flow rates, which facilitate the scale configuration process, and the large changes in pressure, temperature, and other physical and chemical factors on the liquids when passing through the machinery and equipment in the oil and gas industry. Some reported scales contain

carbonate and are often offered in strange crystal forms, such as those in the oil–gas industry in Italy and Malaysia. Few scales contain carbonate, sulfate, and silicate, such as those of the oil plants reported in Brazil, North Africa (Tunisia), and Egyptian oil industry. However, the scale composition may contain some sulfur compounds, such as those reported in the Tunisian, Congo, and Saudi Arabia oil industries. Previous studies reported that many scales are formed in the oil and gas industry. The main types of scales are BaSO_4 , SrSO_4 , and CaSO_3 . The most common scale is BaSO_4 , also known as barite, which has the form of crystalline powder and exhibits various features (*e.g.*, white color) that can vary depending on the crystalline impurities during configuration, high densities, and chemically inert and insoluble in water.^{20,23,41,128,137,159,170} It is observed that a significant radioactivity in some samples taken from different areas in Indonesia, having higher concentrations may be due to the chemical behavior of radium and thorium elements in the deep aquifer filling process, as well as the geological nature of



Table 4 Shows the typical ranges or mean values of concentrations of TENORMs in produced water in many oil fields worldwide, including recent data^a

Country	Concentrations of the TENORM in produced water with their range, averages and units	Ref
Algeria	^{226}Ra (5.1–14.8) Bq g^{-1}	95
Brazil	Ba (0.36–25.7) 8.80^{a} mg L^{-1} , ^{226}Ra (0.012–6.0) 1.95^{a} , ^{228}Ra (<0.05 to 12.0) 2.91^{a} Bq L^{-1}	128
Congo	^{238}U < 4.5×10^{-3} , ^{232}Th < 4.5×10^{-3} , ^{226}Ra 5.1 ^a , Bq dm^{-3}	129
Italy	^{238}U (7.3×10^{-3} to 1.5×10^{-2}), ^{232}Th < 4.5×10^{-3} , ^{226}Ra (0.06–20) Bq dm^{-3}	129
Norway	^{226}Ra 3.3 ^a , ^{228}Ra 2.8 ^a Bq L^{-1} , ^{210}Pb < 10^{a} , ^{210}Po < DL^{a} mBq L^{-1}	119
Syria	^{226}Ra (0.5–16), ^{228}Ra (0.5–21), ^{210}Pb – DL Bq L^{-1}	130
Egypt	^{226}Ra (13.8–111.2) 51.9 ^a , ^{228}Ra (12.4–67.4) 37.5 ^a , ^{224}Ra (0.2–3.7) 1.1 ^a Bq L^{-1}	24 and 26
	^{226}Ra 186.2 ^a , ^{232}Th 19.2 ^a , ^{40}K 1460.8 ^a Bq kg^{-1}	131
	^{226}Ra (9.90–111.2), ^{224}Ra (0.20–3.70), ^{228}Ra (8.80–67.40), Bq L^{-1}	132
	^{226}Ra (1.07–34.15) 16 ^a , ^{228}Ra (<0.02 to 13.26) 4.35 ^a , ^{40}K (3.6–15.37) 7.37 ^a Bq kg^{-1}	93
	^{226}Ra (5–40) 15.92 ^a , ^{214}Pb 0.8–27) 15.93 ^a , ^{214}Bi (1.3–27) 15.43 ^a , ^{228}Ac (1.1–59) 38.37 ^a , ^{212}Bi (0.7–12) 6.9 ^a , ^{208}Tl (1.1–4) 2.7 ^a , ^{40}K (19–43) 30.33 ^a Bq kg^{-1}	37
	^{226}Ra (7.98–17.82) 11.98 ^a , ^{232}Th (4.55–9.60) 7.62 ^a , ^{210}Pb (4.03–41.49) 21.98 ^a , ^{40}K (31.55–166.88) 98.14 ^a Bq kg^{-1}	109
	^{238}U (9.47–25.2), ^{232}Th (7.33–22.6), ^{40}K (632.5–1448.7) Bq L^{-1}	133
	^{226}Ra 8.04 ^a , ^{232}Th 8.62 ^a , ^{210}Pb 35.86 ^a , ^{40}K 72.84 ^a , Bq kg^{-1}	109
Tunisia	^{226}Ra (29.8–46.3), ^{228}Ra (8.5–10.2) Bq L^{-1}	134
Iraq	^{226}Ra 19 ^a , ^{232}Th 39.9 ^a , ^{40}K 66 ^a Bq kg^{-1}	23
	^{238}U 4.12 ^a , ^{232}Th DL^{a} , ^{40}K 14.16 ^a , ^{137}Cs 11.16 ^a Bq L^{-1}	41
	^{226}Ra 20.3 ^a , ^{232}Th 9.4 ^a , ^{40}K 66.4 ^a Bq kg^{-1}	100
	^{226}Ra 1.20 ^a , ^{232}Th 9.4 ^a , ^{40}K 66.4 ^a Bq kg^{-1}	101
Iran	^{226}Ra (<DL–28) 12.35 ^a , ^{228}Ac (<DL–10) 4.88 ^a , ^{235}U (<DL–1.6) 0.71 ^a , ^{214}Pb (<DL–334.1) 142.97 ^a , ^{214}Bi (<DL–471) 178.78 ^a , ^{40}K (4.4–43.7) 24.29 ^a , ^{238}U (30–211) 126.88 ^a , ^{208}Ti (4–44.3) 21.36 ^a , Bq L^{-1}	135
Poland	^{40}K (5–499) 75 ^a , ^{238}U < 30 ^a , ^{226}Ra < 2 ^a , ^{210}Pb < 5 ^a , ^{228}Ra < 2 ^a , ^{228}Th < 2 ^a Bq L^{-1}	136
Turkey	^{224}Ra (<1 to 4) 2.83 ^a , ^{226}Ra (<3 to 10) 6 ^a , ^{228}Ra (<1 to 4) 3.17 ^a Bq L^{-1}	94
Romania	^{238}U (0.043–1.1), ^{226}Ra (23–45), ^{232}Th (0.2–8), ^{40}K (221–899) Bq L^{-1}	137
Omán	^{228}Ac (1019–1040) 1030 ^a , ^{226}Ra (514–529), ^{40}K (1522–1535) 1528 ^a , Bq L^{-1}	138
Ghana	^{238}U (0.11–1.03) 0.54 ^a , ^{232}Th (0.21–0.56) 0.41 ^a , ^{40}K (1.65–11.99) 7.76 ^a Bq kg^{-1}	139
	^{234}U (<DL–6.10), ^{238}U (<DL–5.50), ^{210}Po (22–145), ^{230}Th (2.9–15), ^{232}Th (1.6–5.6) Bq L^{-1}	140
	^{226}Ra (6.20–22.30), ^{228}Ra (6.40–35.50), ^{228}Th (0.71–6.41), ^{224}Ra (0.78–7), ^{40}K (5.90–23.90) Bq L^{-1}	82 and 140
Nigeria	^{226}Ra 8.9 ^a , ^{228}Ra 8.1 ^a , ^{40}K 39.8 ^a Bq L^{-1}	141
	^{228}Ra (0.75–12.30) 5.18 ^a , ^{226}Ra (2.01–13.19) 6.04 ^a , ^{40}K (9.08–155.22) 48.78 ^a , Bq L^{-1}	142
Argentina	U (<10.0 to 33.0) $\mu\text{g L}^{-1}$, ^{226}Ra (<1.7 $\times 10^{-3}$ to 26.8), ^{228}Ra (<1.1 $\times 10^{-3}$ to 9.6) Bq L^{-1}	143



Table 4 (Contd.)

Country	Concentrations of the TENORM in produced water with their range, averages and units	Ref
USA	^{226}Ra (56–1494) 367.5 ^a , ^{228}Ra (69–600) 275.75 ^a , pCi L ⁻¹ ^{226}Ra (30–2690), ^{228}Ra (35–763) Bq L ⁻¹ ^{226}Ra (<0.002 to 58) 11.7 ^a , ^{228}Ra (0.02–59) 15.5 ^a Bq L ⁻¹ ^{226}Ra (548–3970), ^{228}Ra (83–1080), pCi L ⁻¹ ^{238}U (0.0003–0.10), ^{226}Ra (0.002–1200), ^{210}Pb (0.05–180), ^{232}Th (0.0003–0.001), ^{228}Ra (0.30–180) Bq L ⁻¹ ^{226}Ra DL ^a , ^{228}Ra (DL–97.30), ^{238}U DL ^a , ^{235}U DL ^a , ^{137}Cs DL ^a , ^{40}K (DL–265)], pCi L ⁻¹ ^{226}Ra (ND–13 033), ^{228}Ra (ND–1485) Bq L ⁻¹ ^{226}Ra (0.02–0.24), ^{228}Ra (0.02–0.17) pCi ML ⁻¹ ^{226}Ra (0.40–70.30) pCi gm ⁻¹ U (Ra) (ND–101.07), Th (ND–13.71), K (26.1–194.5) Bq L ⁻¹	144 145 146 147 114 148
Azerbaijani		149 150 151 152
UK	^{226}Ra (0.16–90), ^{228}Ra (0.5–12) Bq L ⁻¹	48
Ukraine	^{226}Ra (0.002–1200), ^{228}Ra < 0.001 ^a Bq L ⁻¹ ^{224}Ra (1.57–5.51), ^{226}Ra (27.40–39.80), ^{222}Rn (16.87–19.77), ^{228}Ra (3.20–5.57), ^{230}Th (0.06–0.19), ^{232}Th < 0.001 ^a , ^{234}U 0.01 ^a , ^{238}U 0.01 ^a Bq dm ⁻³	154 153

^a () the data between the brackets means the range/[/]is the averages of concentration/LD = lower than the detection limits.

the region, which has high radioactivity.^{183,185} From safety risk assessment and management point of view, this is still a debatable matter, whereas, still if the likelihood is low then risk's consequences might be high taking in to account the volume of the oil & gas industry's plants distributed around the world *vs.* number of routine maintenance jobs and people involved. Unfortunately, there are many industries that do not carry out survey or dose assessment. Routine maintenance of the facilities where the scales are generated should be carried out while considering the dose values to minimize the risk of contamination to the workers and the environment. In the case of scales containing carbonate, the tubes with hydrochloric acid should be efficiently washed. Meanwhile, the wet mechanical treatment should be used in the case of scales containing sulfates, and the workers must wear special respirators and anti-pollution clothing.

5.3 Sludge

Sludge is a viscous substance consisting of a complex mixture containing various amounts of waste mineral, waste oil, wastewater, sand, *etc.*^{92,184} This substance often contains silica compounds but may also exhibit substantial amounts of barium.^{83,155} Sludge from petroleum industries accumulates in crude oil and natural gas reservoirs, refineries, desalination plants, transport pipelines, and other places during the production processing and transportation of oil and natural gas.^{83,92,148,155} Such accumulated sludge from the petroleum industry is considered hazardous waste according to the regulations set forth by the Environmental Protection Law and

Hazardous Waste Handling Regulations. Radionuclides were reported in the scale and sludge found in machinery and equipment in the oil and gas fields in northern Germany by Kolb and Wojcik. However, some radioactive elements develop the ^{238}U and ^{232}Th series.²² Many researchers worldwide studied the radioactivity of the sludge produced in the oil and gas industry. Several radioactive elements, particularly the highly abundant and long-range Ra isotopes, such as ^{226}Ra , ^{228}Ra , and ^{224}Ra , and other radioactive nuclides, were reported in the natural decay of ^{238}U and ^{232}Th series and ^{40}K element.

The survey included several studies on the sludge generated in the oil and gas industry in different parts of the world.^{1,30,129,173,185} However, many sludge components are naturally dangerous. Thus, different types of wastes, including sludge of all types (chemical, biological, and oily sludge) and of particular environmental importance, are created during the treatment of crude oil. Risky wastes are generated in bulk at oil refineries worldwide and vary from one place to another. For example, petroleum facilities in India annually produce approximately 20 000 tons of oily sludge. Meanwhile, the volume of sludge from oil and gas production in Australia is relatively small (approximately 200 m³ per year) compared with Europe and the USA. Sludge is disposed of in landfills or directly dumped into the sea from the production platform. The latest assessment by the (EPA, 2017) indicated that the average concentration of radionuclide of Ra in the sludge is 2.775 Bq g⁻¹ or approximately 2775 Bq kg⁻¹. The manner by which to safely dispose of such oily sludge is one of the major problems faced by oil refineries. To date, approximately 9 billion tons of



Table 5 Shows the typical ranges or mean values of the TENORM in scales in the oil and gas industry in different regions of the world, including recent data^a

Country	Concentrations of the TENORM in scales with their range, averages and units	Ref
Brazil	^{226}Ra (19.1–323) 106 ^a , ^{228}Ra (4.21 to 235) 78 ^a kBq kg ⁻¹ ^{226}Ra (120.80–3500), ^{228}Ra (147.90–2195) kBq kg ⁻¹ ^{226}Ra (77.90–2110) 897.50 ^a , ^{228}Ra (101.50–1550) 679.50 ^a , ^{40}K (16–144.60) 65.10 ^a kBq kg ⁻¹ ^{226}Ra (109.60–2110) Bq kg ⁻¹ ^{226}Ra (16.10–93.20) 37 ^a , ^{228}Ra (4.04–36.8) 23 ^a , ^{228}Th (4.46–18.5) 10.50 ^a kBq kg ⁻¹ ^{226}Ra (120.80–955), ^{228}Ra (131.40–792) kBq kg ⁻¹ ^{224}Ra (27–115), ^{226}Ra (147–1050), ^{228}Ra (43–181), ^{210}Po (224–2371), ^{210}Pb (<12 to 174) Bq g ⁻¹ ^{226}Ra (0.3–1520) 174, ^{224}Ra (0.1–780) 67, ^{228}Ra (0.6–868) 91 ^a Bq kg ⁻¹ ^{226}Ra (5.90–215), ^{224}Ra (1.90–96.10), ^{228}Ra (2.30–83.30), Bq kg ⁻¹ ^{226}Ra (2–2922) 818 ^a , ^{228}Ra (0.3–254) 64 ^a , Bq kg ⁻¹ ^{226}Ra (226–306), ^{210}Pb (263–42) Bq g ⁻¹ ^{226}Ra (1317–2039), ^{210}Pb (843–1036) Bq g ⁻¹ ^{226}Ra (58.80–131.60), ^{214}Pb (47.50–119.70), ^{214}Bi (49.90–127.20), ^{210}Pb (6.30–11.50), ^{228}Ra (17–59.60), ^{212}Pb (18.70–49.10), Bq g ⁻¹ , ^{238}U (0.94–5.17), ^{232}Th (0.18–0.86) Bq kg ⁻¹ ^{226}Ra (1–15 000), ^{232}Th (0.001–0.002), Bq kg ⁻¹ ^{238}U (0.9–7.6), ^{232}Th (0.8–5.1), ^{226}Ra (31–1189) Bq kg ⁻¹ ^{226}Ra (51–136) 76.5 ^a , ^{232}Th (59–177) 98 ^a , ^{40}K (5.1–105) 25.03 ^a Bq kg ⁻¹ ^{238}U (0.9–53.8) 13.21 ^a , ^{232}Th (0.8–18.9) 3.39 ^a , ^{226}Ra (2.7–1126) 351.1 ^a Bq kg ⁻¹ ^{238}U (0.9–2.7) 1.8 ^a , ^{232}Th (0.8–2.2) 1.5 ^a , ^{226}Ra (97–151) 124 ^a Bq kg ⁻¹ ^{238}U < LD ^a , ^{232}Th (0.14–3.10) 1.80 ^a , ^{226}Ra (0.80–1.50) 1.10 ^a , ^{40}K (4.30–9.60) 7.40 ^a , ^{235}U < LD ^a , ^{224}Ra (1.40–2.71) 2.06 ^a Bq kg ⁻¹ ^{238}U (4500–11 800) 7100 ^a , ^{226}Ra (43 000–120 000) 68.9 ^a , ^{214}Pb (41 900–97 500) 66 500 ^a , ^{214}Bi (42 500–99 700) 66 800 ^a , ^{210}Pb (4200–6500) 4400 ^a , ^{223}Ra (1700–5700) 2700 ^a , ^{228}Ac (19 100–43 700) 24 000 ^a , ^{212}Bi (21 400–42 500), ^{212}Pb (17 000–38 500) 22 400 ^a , ^{208}Tl (20 300–39 000) 252 000 ^a , ^{40}K (1100–4800) 1300 ^a , ^{228}Ra 24 000 ^a Bq kg ⁻¹ ^{226}Ra (7541–143 262), ^{228}Ra (35 460–368 654), ^{214}Pb (18 215–322 604), ^{214}Bi (17 627–320 008), ^{212}Bi (LDL–368 654), ^{208}Tl (LDL–8615), ^{40}K (2914–45 882) Bq kg ⁻¹ ^{226}Ra 37.28 ^a , ^{232}Th 45.70 ^a , ^{210}Pb 34.77 ^a , ^{40}K 621.79 ^a , Bq kg ⁻¹ ^{238}U (9140–285 823) 147 481.5 ^a , ^{232}Th (427–34 339) 17 383 ^a , ^{40}K (51–1031) 541 ^a Bq kg ⁻¹ ^{226}Ra (493–519) 506 ^a , ^{223}Th (35.38–54.08) 44.7 ^a , ^{40}K (LD–0.177) 0.059 ^a Bq kg ⁻¹ ^{226}Ra (7–314.8), ^{223}Th (<DL–177.3), ^{40}K (<DL–223.0), Bq kg ⁻¹ ^{238}U 2500 ^a , ^{226}Ra 11 700 ^a , ^{210}Pb 12 800 ^a , ^{228}Ra 4200 ^a , ^{224}Ra 3900 ^a Bq kg ⁻¹ ^{226}Ra (13–31), ^{228}Ra (5–9), ^{40}K (0.271–0.324) kBq kg ⁻¹ ^{226}Ra (14.1–8.9), ^{228}Ra (5.0–9.5) kBq kg ⁻¹	158–160 160 160 and 161 154 154 162 163 163 132 164 and 165 166 167 168 and 169 154 129 23 129 129 1 20 and 43 32 170 2 171 98 134 77 43



Table 5 (Contd.)

Country	Concentrations of the TENORM in scales with their range, averages and units	Ref
Malaysia	^{226}Ra 68.9 ^a , ^{228}Ra 24 ^a Bq g ⁻¹ ^{238}U (568–729) 660 ^a , ^{226}Ra (2874–2197) 1979 ^a , ^{210}Pb (997–1643) 1399 ^a , ^{228}Ra (574–696) 654 ^a , ^{224}Ra (678–894) 794 ^a , ^{40}K (456–687) 556 ^a , ^{235}U (23.3–29.9) 27 ^a , Bq g ⁻¹ ^{226}Ra (550–434 000) 114 300 ^a , ^{228}Ra (900– 479 000) 130 120 ^a (Bq kg ⁻¹)	172 16
Norway	^{226}Ra (4.9–100), ^{228}Ra (0.40–28.90), ^{210}Pb (<0.2– 49) Bq g ⁻¹	174
German	^{226}Ra (850–100 000), ^{228}Ra (LD–240 000), ^{210}Pb (1400–70 000), ^{228}Th (LD–48 000), ^{227}Ac (LD– 2500) Bq kg ⁻¹	22
Iraq	^{238}U (11.22–32.62) 19.15 ^a , ^{232}Th (6.28–26.56) 18.10 ^a , ^{40}K (51.22–277.59) 195.98 ^a , ^{137}Cs (DL– 3.05) 2.33 ^a Bq kg ⁻¹ ^{226}Ra (137.6–152.4), ^{232}Th (34.4–49.4), ^{40}K (462– 750.3) Bq kg ⁻¹ ^{226}Ra (61.80–128.40), ^{232}Th (12.80–26.80), ^{40}K (82.60–83.10) Bq kg ⁻¹	41 100 101
USA	^{210}Pb 1370 ^a , ^{226}Ra 6230 ^a , ^{228}Ac (^{228}Ra) 565 ^a Bq kg ⁻¹ ^{226}Ra (15 400–76 100) Bq kg ⁻¹ , ^{222}Ra (0.037– 0.153) emanation fraction ^{40}K (ND–0.87), ^{226}Ra (ND–1.90), ^{228}Ra (ND–4.17) pCi g ⁻¹ ^{238}U (0.001–0.50), ^{226}Ra (0.1–15 000), ^{210}Pb (0.02–2000), ^{210}Po (0.02–1.5), ^{232}Th (0.001–0.07), ^{228}Ra (0.05–2800) Bq g ⁻¹ ^{210}Pb 19 250 ^a , ^{226}Ra 90 190 ^a , ^{228}Ra 23 286 ^a Bq kg ⁻¹ ^{226}Ra 13.3 ^a , ^{210}Pb 13.3 ^a , ^{210}Po 13.3 ^a , ^{228}Ra 4.44 ^a , ^{228}Th 4.44 ^a Bq kg ⁻¹ ^{226}Ra (0.84–7.38), ^{228}Ra (0.16–2.283) pCi g ⁻¹ ^{224}Ra (<1 to 520) 67.95 ^a , ^{226}Ra (13–132 000) 20 614.24 ^a , ^{228}Ra (<1 to 453) 63.71 ^a Bq kg ⁻¹ ^{226}Ra (20–9260) 3200 ^a Bq kg ⁻¹ ^{226}Ra (5900–215 000) 119 830, ^{228}Ra (2300–4000) 4200 ^a , ^{224}Ra (1900–96 100) 45 230 ^a Bq kg ⁻¹ ^{226}Ra (38.5–58.3) 43.9 ^a , ^{210}Pb (0.20–0.60) 0.36 ^a , ^{228}Ra (26.8–39.2) 30.3 ^a , ^{228}Th (6–15.9) 11.2 ^a , ^{224}Ra (8.8–15.4) 11.2 ^a , ^{40}K (1.3–2.3) 1.8 ^a , kBq kg ⁻¹ ^{226}Ra (28.22–47.54), ^{40}K (42.65–44.05), ^{232}Th (16.90–40) Bq kg ⁻¹	175 and 176 177 148 105 178 179 147 94 137 22 180 104 143 95 181 182 183 106
Turkey	^{226}Ra (1 to 520) 67.95 ^a , ^{226}Ra (13–132 000) 20 614.24 ^a , ^{228}Ra (<1 to 453) 63.71 ^a Bq kg ⁻¹	
Romania	^{226}Ra (20–9260) 3200 ^a Bq kg ⁻¹	
Syria	^{226}Ra (5900–215 000) 119 830, ^{228}Ra (2300–4000) 4200 ^a , ^{224}Ra (1900–96 100) 45 230 ^a Bq kg ⁻¹	
Ghana	^{226}Ra (38.5–58.3) 43.9 ^a , ^{210}Pb (0.20–0.60) 0.36 ^a , ^{228}Ra (26.8–39.2) 30.3 ^a , ^{228}Th (6–15.9) 11.2 ^a , ^{224}Ra (8.8–15.4) 11.2 ^a , ^{40}K (1.3–2.3) 1.8 ^a , kBq kg ⁻¹ ^{226}Ra (28.22–47.54), ^{40}K (42.65–44.05), ^{232}Th (16.90–40) Bq kg ⁻¹	
Argentina	^{226}Ra (<0.4 to 1.9) $\mu\text{g g}^{-1}$, ^{226}Ra (<0.1 to 1270), ^{228}Ra (115–1670) Bq g ⁻¹	
Algeria	^{226}Ra (1–950) Bq g ⁻¹	
Kazakhstan	^{226}Ra (510–51 000) 18 101 ^a , ^{228}Ra (200–10 000) 5005 ^a , ^{212}Pb (100–11 000) 6000 ^a , ^{214}Pb (510– 49 000) 17 944.3 ^a , ^{212}Bi (100–12 000) 5144.3 ^a , ^{208}Tl (100–10 100), ^{234}Th (<2000 to <3000) < 2285.7 ^a Bq kg ⁻¹	
Russia	^{226}Ra (0.03–7.93), ^{232}Th (0.02–5.09), ^{40}K (ND– 2.28), kBq kg ⁻¹	
Indonesia	^{226}Ra (78–36 106), ^{228}Ra (127–222 974), ^{228}Th (144–90 887), ^{238}U (LD–13 030), ^{232}Th (LD–6024) ^{40}K (20–5496), Bq kg ⁻¹	
Sudan	^{238}U (358.05–4106.9), ^{232}Th (375.88–2736.7), ^{40}K (16.07–9294) Bq kg ⁻¹	

^a () the data between the brackets means the range/^ais the averages of concentration/LD = lower than the detection limits.



Table 6 shows the typical ranges or mean values of TENORM in sludge in the oil and gas industry in different regions of the world, including recent data^a

Country	Concentrations of the TENORM in sludge with their range, averages and units	Ref
Syria	^{224}Ra (7620–27 700) 15 680 ^a , ^{226}Ra (10 050–45 350) 24 870 ^a , ^{228}Ra (10 800–29 100) 17 630 ^a , (Bq kg ⁻¹)	163
Indonesia	^{238}U (12.40–98), ^{232}Th (4–148), ^{226}Ra (40–6129), ^{40}K (105–268), ^{228}Ra (515–7993), ^{228}Th (135–2198) Bq kg ⁻¹	185
Tunisia	^{238}U (5–6.6) 5.6 ^a , ^{232}Th (2.6–10) 6.2 ^a , ^{226}Ra (66–453) 229.3 ^a Bq kg ⁻¹	129
Saudi Arabia	^{238}U (10.3–52.8) 30.53 ^a , ^{232}Th (6.3–47.6) 28.28 ^a , ^{226}Ra (6.8–59.4) 37.80 ^a , ^{40}K (79.9–594) 447.4 ^a , ^{235}U (0.46–7.3) 3.80 ^a , ^{224}Ra (3.57–37.7) 27.31 ^a Bq L ⁻¹	1
Malaysia	^{226}Ra (6–560) 51 ^a , ^{228}Ra (4–520) 58 ^a , (Bq kg ⁻¹) ^{238}U (13–40) 29.44 ^a , ^{232}Th (37–48) 42.56 ^a , ^{226}Ra (104–167) 141.76 ^a , ^{228}Ra (117–158) 135.84 ^a Bq kg ⁻¹	173 108
Egypt	^{226}Ra (<0.2 to 44.3) 20.71 ^a , ^{228}Ra (<0.02 to 43.89) 7.96 ^a , ^{40}K (<0.2 to 19.44) 9.58 ^a BBq Lq kg ⁻¹ ^{226}Ra (5.27–8.68) 6.99 ^a , ^{223}Th (1.08–2.09) 1.37 ^a , ^{40}K (LD–0.677) 0.148 ^a Bq kg ⁻¹ ^{226}Ra 18 032 ^a , ^{214}Pb 19 394 ^a , ^{214}Bi 18 324 ^a , ^{228}Ac 13 257 ^a , ^{212}Bi 7398 ^a , ^{208}Tl 5105 ^a , ^{40}K 1261 ^a Bq kg ⁻¹ ^{238}U (12.3–29.6), ^{232}Th (14.63–28.22), ^{40}K (789.4–1680.5) Bq kg ⁻¹ ^{238}U 108 ^a , ^{226}Ra 11 960 ^a , ^{226}Ra 0.009 ^a , ^{228}Ra 1750 ^a Bq kg ⁻¹ ^{226}Ra (13.10–94.20), ^{232}Th (1.50–27.70), ^{40}K (ND–81.80) Bq kg ⁻¹ ^{226}Ra (5.50–1785.80), ^{232}Th (<DL–885), ^{40}K (<DL–880) Bq kg ⁻¹ ^{226}Ra 14.67 ^a , ^{232}Th 9.99 ^a , ^{210}Pb 68.04 ^a , ^{40}K 112.82 ^a Bq L ⁻¹ ^{226}Ra 8908 ^a , ^{228}Ra 933 ^a , Bq kg ⁻¹ ^{238}U (66–1567) 816.5 ^a , ^{40}K (787–1544) 1165.5 ^a Bq kg ⁻¹ ^{238}U 108 ^a , ^{226}Ra 11 963 ^a , ^{210}Pb 2290 ^a , ^{228}Ra 1747 ^a , ^{224}Ra 1904 ^a Bq kg ⁻¹ ^{226}Ra (<0.1 to 4.7) 2.5 ^a , ^{228}Ra (<0.1 to 4.6) 2.1 ^a , ^{210}Pb < 0.7 ^a Bq g ⁻¹ ^{226}Ra (50–167.80), ^{228}Ra (48.60–152.40), kBq kg ⁻¹ ^{226}Ra (0.36–367.0), ^{228}Ra (0.25–343.0) kBq kg ⁻¹ ^{226}Ra (2.4–3500), ^{228}Ra (35.5–2052) kBq kg ⁻¹ ^{226}Ra (8.10–413.40) 42.70 ^a , ^{228}Ra (9.40–117.90) 40.50 ^a , ^{40}K (16.20–53.70) 21.50 ^a kBq kg ⁻¹ ^{226}Ra (18–20) 19 ^a , ^{228}Ra (21–22) 22.66 ^a , ^{238}Th (13–25) 20.33 ^a , ^{40}K (175–348) 279 ^a Bq kg ⁻¹ ^{40}K 348 ^a , ^{226}Ra (15.40–22.80), ^{228}Ra (17.20–25.60), ^{228}Th 23 ^a , ^{137}Cs 7 ^a Bq kg ⁻¹ ^{226}Ra (0.20–265) 107 ^a , ^{228}Ra (0.11–244) 77 ^a , ^{228}Th (<0.1 to 172) 77 ^a kBq kg ⁻¹ ^{238}U 35.04 ^a , ^{232}Th 15.22 ^a , ^{40}K 149.01 ^a , ^{137}Cs 1.82 ^a Bq kg ⁻¹ ^{226}Ra (235.4–321.8), ^{232}Th (48.9–140.8), ^{40}K (502.7–800.8) Bq kg ⁻¹ ^{226}Ra (1.80–38.10), ^{232}Th (6.80–14.40), ^{40}K (12.10–242.10) Bq kg ⁻¹ ^{210}Pb 5148 ^a , ^{226}Ra 59 000 ^a , ^{228}Ac (^{228}Ra) 28 501 ^a Bq kg ⁻¹	93 171 32 133 186 99 98 170 187 171 134 188 36 158–160 162 160 and 161 189 5 190 27 and 41 100 101 175 and 176
Norway		
Brasil		
Albania		
Brazil		
Iraq		
USA		



Table 6 (Contd.)

Country	Concentrations of the TENORM in sludge with their range, averages and units	Ref
Poland	^{238}U (0.005–0.01), ^{226}Ra (0.05–800), ^{210}Pb (0.1–1300), ^{210}Po (0.004–160), ^{232}Th (0.002–0.01), ^{228}Ra (0.5–50) Bq g^{-1} ^{226}Ra 2.07 ^a , ^{210}Pb 2.07 ^a , ^{210}Po 2.07 ^a , ^{228}Ra 0.7 ^a , ^{228}Th 0.7 ^a Bq kg^{-1} ^{40}K 22.20 ^a , ^{226}Ra 2.34 ^a , ^{228}Ra 3.60 ^a , pCi g^{-1} ^{226}Ra (121–872) pCi g^{-1} ^{40}K (116–272) 183 ^a , ^{238}U (14–393) 271 ^a , ^{226}Ra (15–415) 278 ^a , ^{210}Pb (12–391) 263 ^a , ^{228}Ra (8–516) 340 ^a , ^{228}Th (5–515) 337 ^a , Bq kg^{-1}	105 179 148 149 136
Turkey	^{224}Ra (<1 to 178) 30.6 ^a , ^{226}Ra (1.01–988) 213.29 ^a , ^{228}Ra (<1 to 188) 35.95 ^a Bq kg^{-1}	94
Ghana	^{226}Ra (2.84–36.09), ^{40}K (26.76–189.87), ^{232}Th (2.60–55.90) Bq kg^{-1}	104
Romania	^{226}Ra (21–330) 120 ^a Bq kg^{-1}	137
Argentina	U (<0.4 to 0.7) $\mu\text{g g}^{-1}$, ^{226}Ra (1.9×10^{-3} to 18.7), ^{228}Ra (2.1×10^{-3} to 65.4) Bq g^{-1}	143
Ukraine	^{226}Ra (0.012–1270), ^{214}Bi (0.105–1270), ^{214}Po (0.107–1275), ^{228}Ac (0.003–9.83), ^{212}Bi (0.012–1350), ^{212}Pb (0.07–1.85) kBq kg^{-1} , ^{234}U (0.98–8.50), ^{238}U (0.77–8.90), ^{228}Th (63–1570), ^{232}Th < 1.0 ^a Bq kg^{-1}	153
Libya	^{226}Ra (5.00–19.00), ^{232}Th (2.00–12.00) Bq kg^{-1}	154
UK	^{226}Ra (0.08–27.70), ^{214}Pb (0.06–22.20), ^{214}Bi (0.03–22.50), ^{210}Pb (0.06–3.95), ^{228}Ra (0.28–8.58), ^{212}Pb (0.32–8.98), Bq g^{-1} , ^{238}U (1.56–9.42), ^{232}Th (0.03–0.94) Bq kg^{-1}	168 and 169
Sudan	^{238}U (23.30–655.36), ^{232}Th (16.19–396.34), ^{40}K (16.07–238.65) Bq kg^{-1}	106
Omán	^{226}Ra (0.07–0.203), ^{210}Pb (<0.20 to <0.30), ^{228}Ra (0.0094–0.0636), ^{228}Th (<0.11 to <0.15) ^{40}K (0.0492–0.154) Bq g^{-1}	191
Qatar	^{228}Ac (1019–1040) 1030 ^a , ^{226}Ra (514–529), ^{40}K (1522–1535) 1528 ^a , Bq L^{-1}	138
Irán	^{226}Ra (394.49–27 884.9), ^{232}Th (30.6–94.07), ^{40}K (62.8–110.6) Bq kg^{-1} ^{226}Ra 41 ^a , ^{210}Pb 70 ^a , ^{210}Po 92 ^a ^{226}Ra (6.80–466.30), ^{232}Th (ND–47.6), ^{40}K (ND–154.70), Bq kg^{-1}	192 193 194

^a () the data between the brackets means the range/^a is the averages of concentration/LD = lower than the detection limits.

Table 7 Shows the range and mean values of TENORM in water samples found in the oil and gas industry and production facilities in different regions of the world, including recent data^a

Country	Concentrations of the TENORM in water samples with their range, averages and units	Ref
Iraq	^{238}U (2.41–43.18), ^{232}Th (0.13–7.43) Bq kg^{-1}	197
Azerbaiyán	^{226}Ra (ND–101.70), ^{232}Th (ND–13.71), ^{40}K (26.10–194.50), Bq L^{-1}	154
USA	^{226}Ra (425–1840), ^{228}Ra (616–1630) ^{210}Pb 815 ^a , ^{226}Ra 1 481 ^a , ^{228}Ac (^{228}Ra) 251 ^a Bq kg^{-1} ^{226}Ra (0.15–120), ^{228}Ra (ND–17) Bq L^{-1}	179 175 and 176 199
Irán	^{232}Th (8.7–403), ^{40}K (82–815), ^{226}Ra (0.1–30.3) Bq L^{-1}	198
Nigeria	^{238}U (2.9–13.65), ^{232}Th (0.34–3.89), ^{40}K (48.39–109.39), Bq L^{-1}	200
Tunicina	^{226}Ra 0.037 ^a , ^{232}Th 0.027 ^a , ^{40}K 0.22 ^a Bq L^{-1}	23
Egypt	^{226}Ra 4.895 ^a , ^{232}Th 2.247 ^a , ^{210}Pb 32.87 ^a , ^{40}K 31.852 ^a , Bq L^{-1} ^{226}Ra (5–40), ^{214}Pb (0.8–27), ^{214}Bi (1.3–27), ^{228}Ac (1.1–59), ^{212}Bi (0.7–12), ^{208}Tl (1.1–4), ^{40}K (19–29) Bq L^{-1}	170 32
Turkey	^{224}Ra (<1 to 4), ^{226}Ra (<3 to 10), ^{228}Ra (<1 to <4) Bq kg^{-1}	94
Romania	^{238}U (0.043–18.5), ^{226}Ra (1.8–45), ^{232}Th (0.2–12.2), ^{40}K (25–899) mBq L^{-1}	137

^a () the data between the brackets means the range/^a is the averages of concentration/LD = lower than the detection limits.



Table 8 Shows the range and mean values of TENORM in soil samples found in the oil and gas industry and production facilities in different regions of the world, including recent data^a

Country	Concentrations of the TENORM in soil samples with their range, averages and units	Ref
Saudi Aribia	^{238}U (19.0–62.8) 39.0, ^{226}Ra (8.68–156) 23.2 ^a , ^{232}Ra (2.76–12.8) 7.73 ^a , ^{40}K (108–446) 278 ^a ^{40}Cs (<MDA–3.83) 1.42 ^a Bq kg ⁻¹	196
Egypt	^{226}Ra (7541–194 489), ^{214}Pb (18 215–437 960), ^{214}Bi (17 627–434 435), ^{228}Ac (35 460–897 803), ^{212}Bi (LDL–500 476), ^{208}Tl (LDL–8645), ^{40}K (LDL–45 882) Bq kg ⁻¹	165
Tunisia	^{226}Ra 9.11 ^a , ^{232}Th 11.1 ^a , ^{40}K 176 ^a Bq kg ⁻¹	23
Albania	^{226}Ra (12–23) 18.3 ^a , ^{226}Ra (14–25) 20.7 ^a , ^{238}Th (12–25) 20.3 ^a , ^{40}K (326–549) 413.7 ^a Bq kg ⁻¹	189
Iraq	^{226}Ra (18.4–97.6), ^{232}Th (11.5–42.7), ^{40}K (176.9– 485.6) Bq kg ⁻¹ ^{226}Ra (13.2–15.90), ^{232}Th (25–35.20), ^{40}K (385.5– 432.2) Bq kg ⁻¹ ^{226}Ra 33.588 ^a , ^{232}Th 20.647 ^a , ^{40}K 511.604 ^a Bq kg ⁻¹	100
Turkey	^{224}Ra (<10–535) 223.67 ^a , ^{226}Ra (24.79–70.48) 46.47 ^a , ^{228}Ra (<11–423) 186.67 ^a Bq kg ⁻¹	94
Russia	TPH (1.6–880.3) 223.23 ^a g kg ⁻¹ , ^{226}Ra (0.03– 7.93) 2.62 ^a , ^{232}Th (0.02–5.09) 1.28 ^a , ^{40}K (0.03– 2.28) 0.56 ^a Bq kg ⁻¹	204
Romania	^{226}Ra 21 ^a , ^{232}Th 32 ^a , ^{40}K 311 ^a , Bq kg ⁻¹ ^{238}U (2.4–120), ^{226}Ra (60–330), ^{232}Th (8–87), ^{40}K (53–960) Bq kg ⁻¹	137
Ghana	^{238}U (7.7–25.5) 15.2 ^a , ^{232}Th (8.5–67.2) 26.9 ^a , ^{40}K (60.4–248.9) 157.0 ^a Bq kg ⁻¹ ^{238}U (1.60–21.3) 8.65 ^a , ^{232}Th (2.78–32.2) 12.5 ^a , ^{40}K (111–528) 214 ^a Bq kg ⁻¹	204
Nigeria	^{226}Ra (19.2–94.2) 41.0 ^a , ^{232}Th (17.7–47.5) 29.7 ^a , ^{40}K (107.0–712) 412.5 ^a Bq kg ⁻¹ ^{232}Th (2.36–33.67), ^{40}K (1.79–395) ^{226}Ra (3.52– 41.37) Bq kg ⁻¹	206
Qatar	^{226}Ra 20.05 ^a , ^{232}Th 16.43 ^a , ^{40}K 216.69 ^a Bq kg ⁻¹	192
Canada	^{226}Ra (0.01–3.84), ^{228}Ra (0.01–0.007), ^{228}Th (ND– 0.05) Bq g ⁻¹	208
USA	^{210}Pb 815 ^a , ^{226}Ra 1481 ^a , ^{228}Ac (^{228}Ra) 251 ^a Bq kg ⁻¹ ^{228}Ra (9–1883), ^{226}Ra (12–2802), ^{210}Pb (ND–209), ^{228}Th (11–628) Bq kg ⁻¹ ^{40}K (10–19.70), ^{238}U (ND–1.11), ^{226}Ra (ND–1.43), ^{228}Ra (0.82–1.22), pCi g ⁻¹ ^{210}Pb 1370 ^a , ^{226}Ra 6230 ^a , ^{228}Ac (^{228}Ra) 565 ^a Bq kg ⁻¹	175 and 176
Kuwait	^{232}Th (8.70–27.90) ^{226}Ra (9.80–42.30), ^{40}K (191.2–632) Bq kg ⁻¹ ^{232}Th 12.53 ^a , ^{226}Ra 10.65 ^a , ^{40}K 300 ^a Bq kg ⁻¹ ^{232}Th (10.52–21.96) 13 ^a , ^{226}Ra (33.7–250.6) 85 ^a , ^{40}K (331–449.4) 406 ^a Bq kg ⁻¹	210
Syria	^{226}Ra (18.90–210), ^{232}Th (16.80–55.90), ^{40}K (44– 213) Bq kg ⁻¹ ^{226}Ra (1030–7780) Bq kg ⁻¹ ^{226}Ra (1.34–7.75), ^{228}Ra (0.34–2.42), ^{224}Ra (0.30– 2) Bq kg ⁻¹	131
China	^{226}Ra (16–82) 57 ^a Bq kg ⁻¹	214
Omán	^{226}Ra (30.4–41.2) 34.2 ^a , ^{228}Ra (5.7–11.3) 8.1 ^a , ^{228}Th (5.7–9.1) 7.1 ^a , ^{40}K (93–293) 151 ^a , ^{137}Cs (<0.12 to 1.02) Bq kg ⁻¹	213

^a () the data between the brackets means the range/^a is the averages of concentration/LD = lower than the detection limits.



Table 9 Shows the range and mean values of TENORM in sand samples found in the oil and gas industry and production facilities in different regions of the world, including recent data^a

Country	Concentrations of the TENORM in sand samples with their range, averages and units	Ref
Egypt	^{226}Ra 3.4 ^a , ^{232}Th 3.1 ^a , ^{40}K 37.8 ^a Bq kg ⁻¹ ^{226}Ra (4.29–18.52) 11.63 ^a , ^{232}Th (4.56–18.65) 11.41 ^a , ^{238}U (5.31–17.46) 10.86 ^a , ^{40}K (145.85– 441.15) 327.65 ^a , Bq kg ⁻¹	99 47
Indonesia	^{226}Ra (4686–211 310), ^{228}Ra (7548–170 430), ^{228}Th (4636–177 790), ^{238}U (LD–4810), ^{232}Th (11.60–6493) ^{40}K (297–4930), Bq kg ⁻¹ ^{238}U (13.89–2807.36), ^{232}Th (14.60–3466.24), ^{40}K (16.6–196.28) Bq kg ⁻¹	183
Sudan	^{40}K (0.338–0.514), ^{210}Pb (0.015–0.007), ^{232}Th (0.030–0.040), ^{226}Ra (0.012–0.0145) Bq g ⁻¹	106
Sri Lanka	^{226}Ra (<0.1 to 22) 4 ^a , ^{228}Ra (<0.1–13) 2.5 ^a , ^{210}Pb < 0.5 ^a Bq g ⁻¹	215 and 216
Norway	^{226}Ra (3.8–55.3), ^{232}Th (3.2–5.4), ^{40}K (43–183) Bq kg ⁻¹	188
Kuwait		99

^a () the data between the brackets means the range/^ais the averages of concentration/LD = lower than the detection limits.

oily sludge wastes are found on the planet.^{83,92,155,171,195} This volume is increasing every minute and may vary from one place to another. Although the concentration of radiation is less in the sludge than in the standards, sludge is more soluble in the environment. Accordingly, sludge is formed with produced water, and is considered a form of petroleum wastes extremely hazardous to humans and the environment in case of over-exposure.^{35,83,92,169–195} Sludge is classified as hazardous waste because radium isotopes and their progenies are strong gamma emitters and other radioactive nuclides that emit beta and gamma such as ^{210}Pb , and also those emitting alpha such as ^{210}Po . Therefore, the external radiation dose and associated risks in the vicinity of oil and gas facilities increases as sludge builds up.^{138,191} Sludge may be classified as hazardous wastes (more than the solid wastes) because it is often in liquid form, so when disposed of, it always seeps through the soil and gradually pollutes groundwater and shallow groundwater.

6 TENORM in non-oil samples from oil industry fields and facilities

Many researchers studied TENORM in non-oil samples, such as soil, sand, water, and plants. These samples were selected from the surrounding environments of the oil and gas industry. Thus, appropriate treatments in the cases with indicators of possible future hazards must be used to identify the long-term effect of radiation from the oil and gas industry and determine the extent to which the TENORM moves from oil and gas products and residues onto the surrounding environments. These researchers studied the effect of TENORM on workers, the public, and the environment. Many researchers were quick to take samples of the environment surrounding oil and gas facilities that are likely to be contaminated with TENORM. Then, these samples were studied, after which the transmission

of radionuclides from the oil and gas industry was measured to avoid their effects on health and the environment.

Concentrations of the highly radioactive elements in these samples in an oil-contaminated environment are relatively high compared with the non-oil ones. For example, concentrations of radioactive elements obtained from soil samples in Saudi Arabia taken from the Ras Tanura area (an oil polluted environment)¹⁹⁶ are greater than those of the same radioactive elements obtained in sand samples taken from Qassim area in Saudi Arabia (an environment that is non-polluted by oil).²²⁴ This finding indicates that the oil spill may adversely affect the environment in which it has been extracted. In general, the concentrations of radionuclide activity in soil, water, plants, drill cutting, and other samples taken from oil industry fields and facilities (Tables 7–11) are higher than those obtained for

Table 10 Shows the range and mean values of TENORM in drill cutting samples found in the oil and gas industry and production facilities in different regions of the world, including recent data^a

Country	Concentrations of the TENORM in Drill cutting samples with their range, averages and units	Ref
USA	^{226}Ra (0.835–7.38), ^{228}Ra (0.161–2.283) pCi g ⁻¹ ^{208}Tl (0.21–0.49), ^{226}Ra (0.83–2.25), ^{228}Ra (0.63– 1.63), ^{234}U (0.34–1.94), ^{235}U (0.010–0.123), ^{238}U (0.46–2.09), ^{228}Th (0.51–2.12), ^{230}Th (0.54–3.03), ^{232}Th (0.64–2.17) pCi g ⁻¹	147 217
Poland	^{40}K (680–999), ^{232}Th (23.30–31) Bq kg ⁻¹ ^{40}K (446–1092) 815 ^a , ^{238}U (35–99) 57 ^a , ^{226}Ra (29– 120) 62 ^a , ^{210}Pb (30–43) 69 ^a , ^{228}Ra (22–47) 37 ^a , ^{228}Th (19–47) 37 ^a , Bq kg ⁻¹ ^{40}K (289–3590) 966 ^a , ^{238}U (35–46) 37 ^a , ^{226}Ra (15– 44) 25 ^a , ^{210}Pb (15–54) 30 ^a , ^{228}Ra (13–31) 20 ^a , ^{228}Th (13–26) 18 ^a , Bq kg ⁻¹	218 136 136

^a () the data between the brackets means the range/^ais the averages of concentration/LD = lower than the detection limits.



Table 11 Shows the range and mean values of TENORM in other environmental samples found in the oil and gas industry and production facilities in different regions of the world, including recent data^a

Country	Samples	Concentrations of the TENORM in other environmental samples with their range, averages and units	Ref
Sudan	Grass	^{238}U 59.13 ^a , ^{232}Th 52.42 ^a , ^{40}K 224.7 ^a Bq kg ⁻¹	106
Nigeria	Grass	^{238}U (14.70–6.20), ^{232}Th (7–11.40), ^{40}K (66.80–70.20) Bq kg ⁻¹	219
Romania	Spontaneous vegetation	^{238}U (0.2–55), ^{226}Ra (3.7–59.2), ^{232}Th (0.05–0.12), ^{40}K (710–1100) Bq kg ⁻¹	137
Libya	Clay shale	^{226}Ra 1 \div 990 ^a Bq kg ⁻¹	154
Nigeria	Clay	^{226}Ra 28.7 ^a , ^{232}Th 67.1 ^a Bq kg ⁻¹	
Azerbaiyán	Rock	^{226}Ra (ND–96.70), ^{232}Th (ND–18.94), ^{40}K (115.40–467.30), Bq kg ⁻¹	220
China	Limestone	^{226}Ra (13–29) 20 ^a Bq kg ⁻¹	214
	Shale	^{226}Ra (114–183) 149 ^a Bq kg ⁻¹	
	Sediments	^{226}Ra (306–396) 351 ^a Bq kg ⁻¹	
USA	Sediment	^{40}K (590–239), ^{235}U (ND–6.32), ^{208}Tl (8.58–17.30), ^{212}Pb (25.90–47.80), ^{212}Bi (35.80–104), ^{224}Ra (47.30–26), ^{228}Ra (^{228}AC) (26.40–49.2), ^{210}Pb (ND–64.34), ^{214}Pb (27.30–47.10), ^{214}Bi (23.90–40.90), ^{234}Pa m (ND– 95.60), ^{234}Th (28.30–50.30) Bq kg ⁻¹	223
	Biota	^{226}Ra (0.2–1), ^{228}Ra (0.2–1) pCi ML ⁻¹	
	Rock	^{226}Ra (0.1–1), ^{228}Ra (0.1–1) pCi ML ⁻¹ ^{238}U (0.1–70 000), ^{232}Th (0.1–20 000) ppm	221

^a () the data between the brackets means the range/^a is the averages of concentration/LD = lower than the detection limits.

the same samples in non-polluted environments. For example, concentrations of radioactive elements of soil samples for the oil-bearing environment²²⁵ are higher than for soil samples of the non-oil-bearing environment.²²⁶ The results of an investigation done on samples in the oil environment in Kuwait⁴⁶ indicates that the discharge of produced water has an effect on the concentrations of TENORM in the environment of the lake of the produced water, thereby increasing their value compared with the values previously reported through the broader Kuwaiti environment. High concentrations of activity may be the result of oil spillage, uncontrolled storage or unsafe discharge, leakage of sludge, produced water or scales, and their interaction with the environmental components.

7 Risks of TENORMs on the environment

The principle of environmental protection of radiation applies extensively to the nuclear industry, but in the oil and gas industry, this protection seems to be relatively low. Radiation protection is unconstrained to animals or plants, while humans are protected by certain radiological standards.^{227,228} For this reason, the International Commission on Radiological Protection (ICRP, 1991) postulated the adoption of human protection standards as criteria for the protection of living organisms.^{227–229,278} This condition indicates that if the overall

exposure to radiation from TENORM discharges, or the disposal of TENORM wastes is less than the general limit of 1 mSv y⁻¹, then the radioactive doses received by organisms are considered acceptable.²²⁸ But it should take into account that it is still associated with high uncertainties. Using contaminated TENORM or waste media and their improper handling, storing, and transfer without effective control can contaminate multiple land areas in the surrounding environment which, in turn, can lead to the potential exposure of the public.^{92,133,227–230} The environmental effects of the TENORM resulting from the industry of oil and gas can be summarized as follows: (1) discharge of produced water, scale, and sludge into the offshore facilities of the oil and gas industry may lead to the transfer of TENORM to the surrounding environment. Accordingly, the content of radionuclides in rock and soil deposits, river water, and other components of the surrounding environment increases, which may be a cause of potential contamination of drinking water. These radionuclides may be transported through water and soil to the animals and plants. (2) The petroleum industry has a low TENORM activity and a low generation rate in the environment.^{230–232} For this reason, it may be unlikely that radioactive exposures and radiation impact on life are significant because of the discharge of small amounts of petroleum wastes, but these wastes accumulate over time, and thus may pose radioactive hazards to humans and their environment.



7.1 The main forms of petroleum pollution TENORMs

Disposal of hazardous waste from petroleum industries by burying them in the ground or dumping them into the sea leads to the possibility of pollution of air, soil, groundwater, surface and sea water, which leads to possible adverse effects on the environment, animals, plants and fish, and thus on the health of man himself.^{230–240} Perhaps the most prominent of these forms are:

Impact on water.^{236–241} The extractive oil industry is highly influential in water as it is reflected in groundwater, surface water, rivers, oceans and seas, as a result of the waste generated from the exploration and extraction (drilling fluids and waste water), as well as extraction of oil through water injection in order to increase the withdrawal of oil and improve productivity.²⁴² In this way, the water works to remove oil from the porous medium through the separation processes, which keeps oil in the reservoir.

Impact on groundwater.^{238–243} Groundwater is polluted as a result of waste liquid and non-liquid drilling (basins Waste). These wastes contain high levels of salts such as barium, benzene, *etc.*,²⁴⁴ which are rich in radionuclides, where the liquid part of the waste is filtered to the ground water, causing serious radioactive contamination.^{232,245–248}

Impact on surface water.^{8,138,225,249} All effluents from the oil and gas industry such as produced water, washing water for rigs, drilling machines, maintenance and repair, oil grease

contaminated water, liquid sludge, *etc.*, may contain radioactive materials. Consequently, discharging into the environment and lead to the release of these materials into the surface of the earth and may cause unexpected damage due to increase the concentrations of radioactive materials by leakage to subsurface water.

Effect on air.^{249–252} Air pollution caused by the emission of gas associated with the drilling and extraction, resulting mainly from the combustion of fuel and the process of disposal of unwanted gases that appear with oil extracted and the evaporation of volatile parts of oil spread over the surface of water.^{253–255} ²²²Rn is one of the most important gases emitted from the exploration and extraction processes.

Impact on soil.^{211,236–238,254–261} It is known that the drilling process directly affects the soil because it causes large impacts in the ground, and the drilling machines may cause leaking the fuel through the pores of the soil, as well as the oil residues.^{242,258–261} Then, radionuclides are transferred to the soil and then to the plants and living cells. Existing methods for the disposal of radioactive petroleum wastes used by the oil and gas industry *e.g.* land farms, deep injection... *etc.* as well implementation of enhanced oil recovery technologies such produced water injection method that contains TENORM or hydraulic fracturing methods that use TENORM slurry, all also have a great impact to contaminate soil. The main forms of petroleum pollution TENORMs can be summarized in Fig. 3 as shown below:

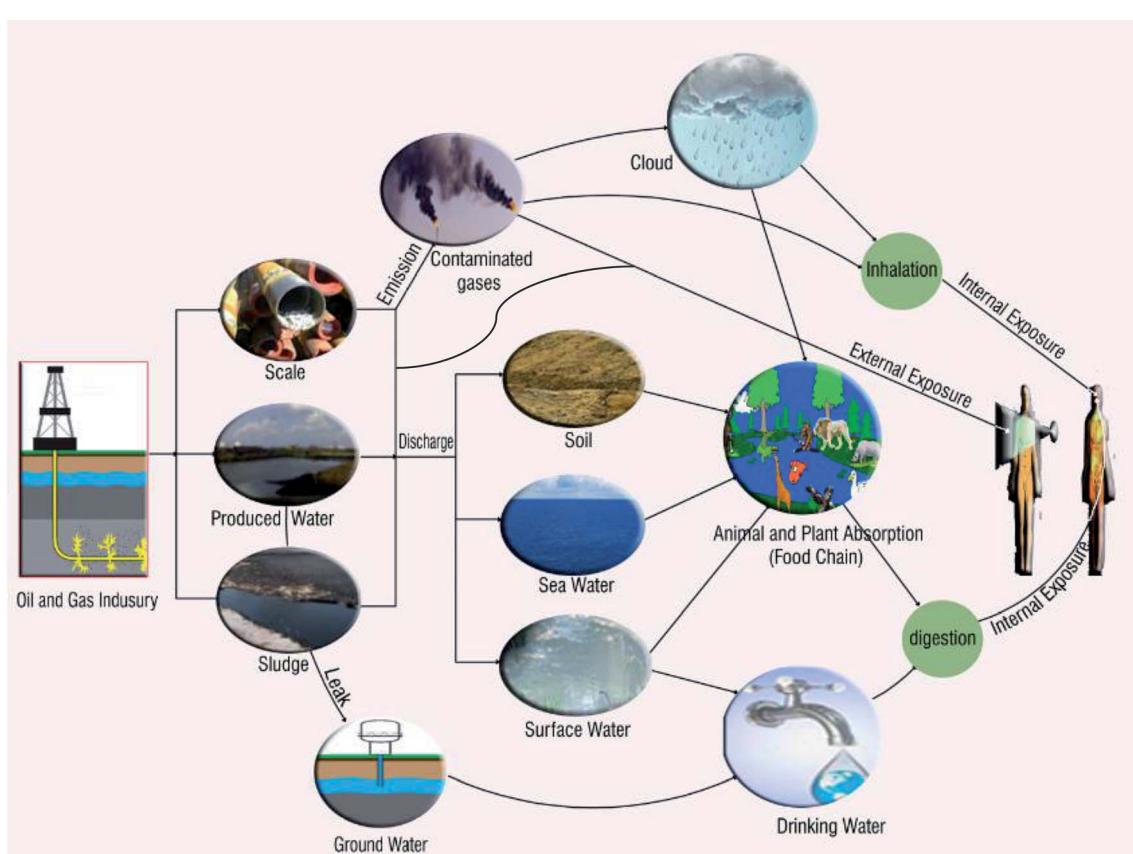


Fig. 3 The main forms of petroleum pollution of TENORMs.^{242,261}



Water and soil contamination with oil containing radionuclides has effects on plants and organisms (birds, cattle, reptiles, *etc.*).^{262–265} Thus, these radionuclides may pass through the food chain to humans, causing many health effects such as cancer and others.

8 Risks of TENORMs on human health

Oil and gas products and residues may contain TENORMs, which then emit alpha, beta, and gamma radiation. Thus, TENORMs can be a source of radiation. A person can be exposed to TENORMs in two ways: external and internal exposures (see Fig. 3).

8.1 External exposure (irradiation)

In this case, the source remains outside the body. Workers are exposed to gamma radiation during routine operations. The external gamma radiation passes through the steel walls of the tubes and vessels, and the dose rate on the surface of oil production pipelines and vessels can be within the tens of microSievert per hour. During closure and maintenance periods, workers may also be exposed to inhaled radon, a dust of the TENORM and gamma radiation. Studies, such as,^{23,39,42,139,171,202} on oil and gas found that the rate of external radiation dose within the separators in the machinery and equipment is higher than that in the outer walls because the external gamma radiation inside the separator is unprotected by its steel walls.

8.2 Internal exposure (contamination)

The radioactive material is transported to the body either by inhalation, ingestion, or absorption.^{82,100,133,226–229} The radium equivalent activity in the sulfate scale may be greater than 500 Bq g⁻¹. Thus, only 100 mg of inhaled dust is sufficient for an irradiation dose exceeding the annual general dose limit of 1 mSv. Although the amount of radon released is low, concentrations of radon in non-aerated vats of scale, sludge, and sand can cause increased exposure to radiation.

Many researchers and specialists addressed the health effects of ionizing radiation exposure. The results showed that the health effects associated with exposure to ionizing radiation vary depending on the total amount of energy absorbed, time period, dose rate, and body exposed to radiation. A key consideration in dealing with TENORM is that, exposures are generally low and below the internationally defined regulatory levels.²²⁸ Exposure to TENORM produces no sharp and severe effects similar to the effects associated with exposure for high levels of radiation from man-made sources. In most cases, exposure to low ionizing radiation exhibits no adverse health effects. However, many new epidemiological and laboratory studies have concluded that exposure to low doses of ionizing radiation may still cause a risk.^{230–233} A variety of cancers, including leukemia, lung cancer, stomach, esophagus, bone, thyroid, brain, and nervous system, have been associated with exposure to ionizing radiation. This indicates that exposure to TENORM over the limits of exposure to the general public or following inadequate safety precautions usually presents delayed effects, such as the development of some forms of

cancer. Potential health effects are strongly linked to dose, and radiation exposure is unassociated with all forms of cancer.^{117,228,229} In general, many researchers have studied the risks of exposure to radiation from the oil and gas industry in different parts of the world. The handling and storage of contaminated petroleum waste by TNORMs can expose workers, public, and environment to harmful radiation doses. Meanwhile, burial and spread of petroleum wastes in the ground are associated with potential exposure to external radiation and inhalation of radon. If the concentrations of ²²⁸Ra, ²²⁶Ra, and ⁴⁰K and the rest of the radioisotopes deposited in the scale, sludge, produced water, and other samples are determined, exposure can be measured through the highly common radiation indicators to assess the real doses of radiation, including the radon equivalent (R_{eq}), dose-absorbed dose (D_{γ}), and effective annual dose rate (D_{eff}).^{42,82,169,187} These aforementioned factors are indicators of reference doses in the outer air at the height of 1 m above the Earth's surface and can be calculated from the equations adopted by the United Nations Scientific Committee on the effects of atomic radiation:

$$R_{\text{eq}} = C_{\text{Ra}} + 1.43C_{\text{Th}} + 0.077C_{\text{K}}$$

$$D_{\gamma} = (0.462C_{\text{Ra}} + 0.604C_{\text{Th}} + 0.0417C_{\text{K}}) \times 10^{-3}$$

$$D_{\text{eff}} = D_{\gamma} \times 10^{-6} \times 8760 \times 0.20 \times 0.7$$

where R_{eq} is radium equivalent activity measured by Bq kg⁻¹, and it is a radiation index used to evaluate actual radioactivity in substances containing natural radionuclides; C_{Ra} , C_{Th} , and C_{K} are the activities for ²²⁶Ra, ²²⁸Ra, and ⁴⁰K, respectively; D_{γ} is the rate of dose resulting from gamma rays measured by (nGy h⁻¹); and D_{eff} is the annual effective dose and measured by mSv y⁻¹. Several studies have attempted to measure the radioactivity doses and the true dangers in the petroleum industry. Some survey results are presented in Table 6.

These results provide evidence that the oil and gas production areas may be contaminated with TENORM but generally low. But over time, the accumulation of oil and gas residues under improper management can produce harmful radiation doses to humans and the environment.^{23,39,95,104} The higher the level of radioactivity in waste, the greater the radiation effects, especially when considering the possibility of exposure to operators through internal pollution by absorbing dust during waste treatment.^{94,139,198,201,266} Immediacy to the source of radiation exhibits a significant part in the radioactivity effect. This condition may lead to the accumulation of additional radiation exceeding the limits allowed to the public, thereby leading to health risks in these communities (Table 6).

Overall, when comparing studies done to measure TENORMs in the oil and gas industries with global liquid fuel production per day, we find that there are large amounts of waste likely to contain radioactive materials, which are traditionally disposed of in the environment.^{267,268} All onshore and offshore drilling activities are extremely risky and have a serious potential to harm people, cause environmental damage or loss of assets, and negatively impact industry reputation. For this



Table 12 Radiation risk indicators in some oil industries

Country	Ra_{eq} (Bq kg $^{-1}$)	D_{yr} (nGy h $^{-1}$)	D_{eff} (mSv y $^{-1}$)	Ref
Saudi Arabia	(21.6–204) 62.1 1.35–173	(10.5–96.4) 29.3 <LD–83.6	(0.013–0.118) 0.038	196 1
Egypt	(70.4–213.9) $\times 10^3$ (544–596) 570 $\times 10^3$	31–72.7 $\times 10^3$ (250–273) 262	(306–335) 321	42 171
Tunisian	0.09–398	0.043–177	5.28×10^{-5} to 3.64×10^{-4}	23
Algeria		(0.1–100) $\times 10^3$	(0.01–0.60)	95
Argentina		(0.8–400) $\times 10^3$	(0.01–1.6)	143
Nigeria		(0.32–1.38)	(0.00281–0.0121)	102
Congo, Italy		(0.1–6) $\times 10^3$		129
Turkey		(0.2–25.7) $\times 10^3$		94
Ghana		(10.0–57.0)	(12.6–83.4)	139
Nigeria	(10.01–128.92)	(21–117.5) 54.6 $\times 10^3$	(0.03–0.2) 0.07	104 200
	(51.04–100.85) 74.71	(23–47) 35	(0.027–0.057) 0.043	201
	(40.96–108.58) 60.75	(20–61) 32		201
Hungary		(85–129) 180	(0.5–0.7) 0.6	266
Albania		(32–58) 38	(0.04–0.07) 0.05	189
Iraq	(41.75–117.61)	(21.08–57.10)	(0.025–0.280)	198
		(0.08–11.2)		100
USA	(5.21–79.84) 47.39	(2.49–37.53) 22.18	(0.012–0.184) 0.109	39
			(0.002–0.50)	68

reason, many researchers have taken the initiative to find safe ways to get rid of the tumor in the oil and gas industries. For example, Khalid Al Nabhani and others introduced a program to develop a larger TENORM risks management system in the oil and gas industries.^{269,270} This system emphasizes the importance of new methods of special personnel protective equipment protected by an effective and lightweight layer of lead-based materials, and introduces a new thermal chemical conversion technology (TCT) for the treatment of TENORM.²⁷⁰ This technology is designed to manage TENORM waste and ultimately turn it into renewable fuel and energy.^{19,271} This system also revealed that there is a strong correlation between the radioactive materials and presence of hydrocarbons, which identified knowledge and technical gaps related to TENORM in oil and gas production, and also rethink the interpretation of the theory of oil and gas formation based on logical scientific explanations.²⁷² This system provides an analysis of risk assessment methods commonly used in the oil and gas industry and TENORM waste disposal options.²⁷³ To evaluate its effectiveness, the system uses a fate model, exposure paths, and integrated exposure pathways.^{272,273} It also examines reasonable scenarios in which pollutants can travel through the biosphere and atmosphere, reaching the humans, animals and environment. The real state scenario of TENORM wastes disposed of in the evaporation pond was simulated using RESRAD (version 6.5).^{274,275} The system also introduces a new approach to dynamic modeling and quantitative risks assessment of TENORMs exposure in the oil and gas industries by use the SMART approach, which integrates the rationality theory (SMART approach) and SHIPP methodology (system risk identification, prediction and prevention). The SHIPP methodology is a general framework used to model, identify and evaluate potential TENORM occupational exposure incidents.^{276,277} They focused on the relationship between legislation and policy in

the oil and gas industry and laws related to the oil and gas industry that protect human health and environmental safety. Also highlighted the importance and activation of the role of public participation in drafting legislation that strives to balance the interests of the authorities with those of the public in a democracy. In spite of all the above, we find a big gap between the results of studies and scientific and technological research, which was concerned with the study and evaluation of risks related to the tumor in the oil and gas industry and how to safely dispose of it and the practice in the production facilities of the oil and gas industry. This requires that oil and gas production companies to adopt safer policies and turn the results of the studies into practice on the ground.

9 Conclusions

The review results of obtainable data relating to the incidence of TENORM in the oil and gas industry indicate that an initial conclusion can be strained on the necessity for further study in this field. Human and technological activities in the oil and gas industry can increase concentrations of naturally occurring radionuclides (TENORMs). An overall review was conducted to determine the concentrations of radioactive elements in petroleum products, petroleum residues (produced water, scales, and sludge) and in samples of oil environments (soil, water, and plant). The subsequent radioactive effects of the oil and gas industry on workers, public, and environment were also assessed. The activities of the observed elements showed varying ranges, as presented in the abovementioned tables. Evidently, the activities of some radionuclides exceed the 10 Bq kg^{-1} exemption level recommended in the safety standards of IAEA. This finding indicates that extended constant consideration and monitoring are required through most routine processes in the industry, because TENORM waste from the oil and gas industry may



produce high levels of radiation exposure. These exposures are generally caused by external γ -radiation that comes from the radionuclides and their offspring, which in turn, may lead to multiple environmental and health risks.

Conflicts of interest

The authors have no affiliation with any organization with a direct or indirect financial interest in the subject matter discussed in the manuscript.

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