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RSC Advances



REVIEW



Cite this: RSC Adv., 2019, 9, 39201

Received 5th August 2019 Accepted 31st October 2019

DOI: 10.1039/c9ra06086c

rsc.li/rsc-advances

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 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: ²²⁶Ra, ²²⁸Ra, ²²²Rn, ²¹⁰Pb, ⁴⁰K 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 Bq g^{-1} , and for more fly ash and some oil/gas scales, the radioactivity can reach 1000 Bq g^{-1} . 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: ²²⁸Ra, ²²⁶Ra and ²²²Rn^{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 TEN-ORMs 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: NOR48-50 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. HINAR51,52 is the abbreviation for the definition of High Natural Radioactivity, which is interpreted by a focus on areas affected by high natural radioactivity. NARM53-55 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. ENOR19,56 is the abbreviation for a definition of Enhanced Naturally Occurring Radioactivity, which is interpreted by naturally occurring radioactivity technologically enhanced. TENR56-61 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. TEN-ORM^{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 oilrelated 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 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 226 Ra 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 226 Ra was in the average of 11 512.8 Bq kg $^{-1}$. Thus, 232 Th, 40 K, 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

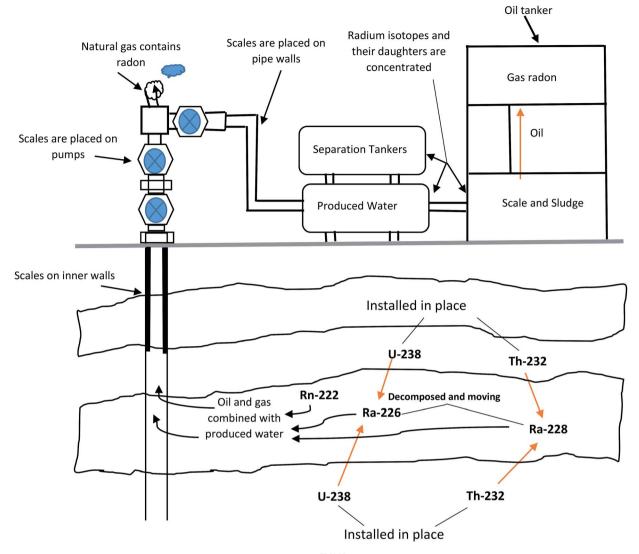


Fig. 1 The presence of natural radioactive materials in the oil industry. 78,82,83

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 reinjection 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

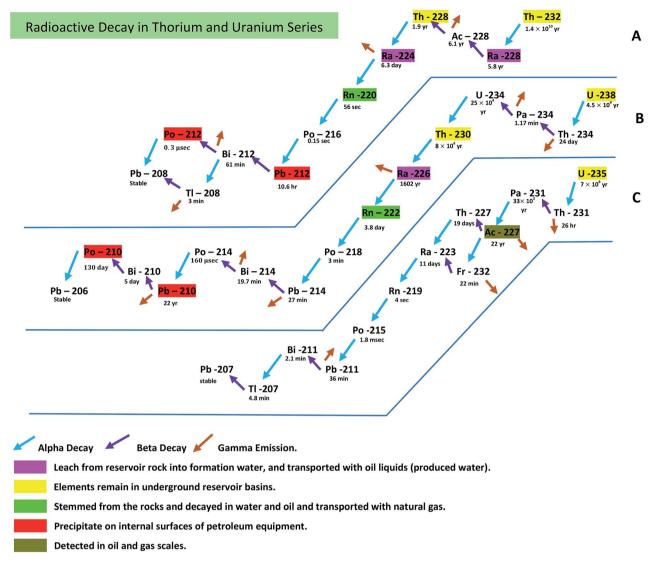


Fig. 2 Primordial radioactive decay series (A) ²³²Th, (B) ²³⁸U and (C) ²³⁵U.^{81,89}

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 sea water, 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 Bg 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 ²²⁶Ra and ²²⁸Ra. Numerous researchers, such as, ¹⁵³ reported the presence of ²²⁴Ra isotope in addition to ²²⁶Ra and ²²⁸Ra 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, 40K is one of the abundant elements in produced water and reported in many studies. For example, 40K was expressed in produced water in the Egyptian,93 Tunisian,23 and Nigerian141,142 petroleum industries. 137Cs 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 $^{-1}$	96 and 97
	238 U (0.0001–10), 226 Ra (0.1–40), 210 Po (0–10), 232 Th (0.03–2) mBq g ⁻¹	105
	222 Rn (5–200 000), 210 Pb (0.005–0.02) 210 Po (0.002–0.08) Bq m ⁻³	105
Saudi Arabia	238 U LD $^{\rm a}$, 232 Th $^{\rm c}$ 10.30 $^{\rm a}$, 226 Ra $^{\rm c}$ 26.5a, 224 Ra $^{\rm c}$ 10a, 40 K 4.3a Bq kg $^{\rm -1}$	1
Egypt	226 Ra $^{3.02^{a}}$, 228 Ra 200 , 40 K 40 K 40 R 40 Bq kg $^{-1}$	93
	226 Ra (31–2669), 232 Th (<dl–913.4), <math="">^{40}K (<dl–98) bq="" l<math="">^{-1}</dl–98)></dl–913.4),>	98
	226 Ra (ND-21.30), 232 Th (ND-10.50), 40 K (56.6-126) Bq kg ⁻¹	99
Tunicina	226 Ra (<0.106-0.38), 232 Th (0.02-0.12), 40 K (0.057-1.308) Bq kg ⁻¹	23
Iraq	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	27 and 41
	226 Ra 33.6 $^{\rm a}$, 232 Th 13.0 $^{\rm a}$, 40 K 197.0 $^{\rm a}$ Bq kg $^{-1}$	100
	226 Ra (2.30–5.80), 232 Th (2.90–5.50), 40 K (5–36) Bq kg ⁻¹	101
Turkey	²²⁴ Ra (3 to <11) 7.75 ^a , ²²⁶ Ra (5-16) 9.25 ^a , ²²⁸ Ra (2-10) 7 ^a Bq kg ⁻¹	94
Nigeria	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	102
	238 U (0.0000001–0.01), 226 Ra (0.0001–0.04), 210 Po (0–0.01) 232 Th (0.00003–0.002) Bq L $^{-1}$	103
Ghana	²²⁶ Ra (2.42–10.14), ⁴⁰ K (21.08–34.39), ²³² Th (11.5–12.65), Bq kg ⁻¹	104
Sudan	238 U $^{64.11^a}$, 232 Th $^{63.69^a}$, 40 K $^{22.13^a}$ Bq kg $^{-1}$	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 ²¹⁰Po, ²¹⁰Pb, and ²²⁸Th in produced water is scarce compared with Ra isotopes. ¹⁴⁴ In 1997, the presence of radionuclides ²¹⁰Po, ²¹⁰Pb, ²²⁸Th, ²²⁶Ra, and ²²⁸Ra 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 BaSO₄, SrSO₄, and CaCO₃.37,68,129,155 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:

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3$$
,
 $Sr^{2+} + SO_4^{2-} \rightarrow SrSO_4$,
 $Ba^{2+} + SO_4^{2-} \rightarrow BaSO_4$.

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

	Concentrations of the TENORM in petroleum	
Country	productions with their range, averages and units	Ref
Saudi Arabia	²³⁸ U (<ld-8.5), <sup="">232Th (<ld-0.62), <sup="">226Ra (<ld-< td=""><td>1</td></ld-<></ld-0.62),></ld-8.5),>	1
	0.45), ²²⁴ Ra (<ld-0.598), <sup="">40K (0.13-57.05) Bq</ld-0.598),>	
	kg^{-1}	
Syria	²²⁴ Ra (9–14) 11.43 ^a , ²²⁶ Ra (4–11) 7 ^a , ²²⁸ Ra (3–13)	30
	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–	107
	36), 226 Ra (77–135), 228 Ra (24–31) Bq kg ⁻¹ 238 U (13–40) 29.44 ^a , 232 Th (37–48) 42.56 ^a , 226 Ra	
Malaysia	(13-40) 29.44°, ⁵⁵ Th (37-48) 42.56°, ⁵⁵ Ra (104-167) 141.76°, ²²⁸ Ra (117-158) 135.84° Bq	108
Formt	kg ⁻¹ ²²⁶ Ra (<0.2 to 46.78), ²²⁸ Ra (<0.2 to 10.50), ⁴⁰ K	93
Egypt	(<0.2 to 17.80) Bq kg ⁻¹	93
	226 Ra (1.13–15.70) 8.04 ^a , 232 Th (3.31–23.32)	109
	8.62 ^a , ¹¹⁰ Pb (17.16–48.55) 35.86 ^a , ⁴⁰ K (19.24–	109
	211.25) 72.84^{a} Bq kg ⁻¹	
	²²⁶ Ra (21–38), ²³² Th (9–15), ⁴⁰ K (303–154) Bq	110
	kg^{-1}	
	²³⁸ U 2.7 ^a , ²³² Th 9284.1 ^a , ²²⁶ Ra 11 512.8 ^a , ²¹⁰ Pb	111
	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)	99
	Bq kg ⁻¹	
Kuwait	²²⁶ Ra (ND-17.0), ²³² Th (ND-10.5), ⁴⁰ K (13.6-126)	99
	$\mathrm{Bq}\ \mathrm{kg}^{-1}$	
Iraq	²³⁸ U (1.59–6.63), ²³² Th (DL–35.47), ⁴⁰ K (14.16–	27 and 41
	208.50), 137 Cs (1.30–11.16) Bq L ⁻¹	
	²²⁶ Ra 31.6 ^a , ²³² Th 14.4 ^a , ⁴⁰ K 387.6 ^a Bq kg ⁻¹	100
Tunicina	²²⁶ Ra (0.07–0.64), ²³² Th (<ld-0.16), <sup="">40K (0.142–</ld-0.16),>	23
_	3.7) Bq kg^{-1}	
Ghana	226 Ra (<0.12 to 17.81), 40 K (<0.15 to 43.46), 232 Th	104
V	(<0.11 to 18.19) Bq kg ⁻¹	440
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 ,	112
	2^{7} ,Bi 16.7°,Bi 70°,Ac 14°,Th 0.48°, 232 Th 38°, 235 U 4 × 10 ^{-4a} , 238 U 0.65°, 40 K 20° Bq	
	kg ⁻¹	
	кg	

^a () the data between the brackets means the range/^a is 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 BaSO₄, SrSO₄, and CaSO₃. The most common scale is BaSO₄, 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 $(0.012-6.0)$	128
	1.95^{a} , 228 Ra (<0.05 to 12.0) 2.91^{a} Bq L ⁻¹	
Congo	238 U < 4.5 × $^{10^{-3a}}$, 232 Th < 4.5 × $^{10^{-3a}}$ 226 Ra	129
The lea	$5.1^{ m a}, { m Bq} { m dm}^{-3}$ $^{238}{ m U}$ (7.3 $ imes$ 10 $^{-3}$ to 1.5 $ imes$ 10 $^{-2}$), $^{232}{ m Th}$ < 4.5 $ imes$	120
Italy	10^{-3a} 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 <	119
riorivaj	DL^a mBq L^{-1}	113
	226 Ra (0.5–16), 228 Ra (0.5–21), 210 Pb–DL Bq L $^{-1}$	130
Syria	²²⁶ Ra (13.8–111.2) 51.9 ^a , ²²⁸ Ra (12.4–67.4) 37.5 ^a ,	24 and 26
	224 Ra (0.2–3.7) 1.1 ^a Bq L ⁻¹	
	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	132
Egypt	$(8.80-67.40)$, Bq L ⁻¹ 226 Ra $(1.07-34.15)$ 16^{a} , 228 Ra $(<0.02$ to $13.26)$	93
Едург	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	37
	(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}	
	²²⁶ Ra (7.98–17.82) 11.98 ^a , ²³² Th (4.55–9.60)	109
	7.62 ^a , ²¹⁰ Pb (4.03-41.49) 21.98 ^a , ⁴⁰ K (31.55-	
	166.88) 98.14 ^a Bq kg ⁻¹ ²³⁸ U (9.47–25.2), ²³² Th (7.33–22.6), ⁴⁰ K (632.5–	122
	$0 (9.47-23.2), 111 (7.33-22.0), R (032.3-1448.7) \text{ Bq L}^{-1}$	133
	²²⁶ Ra 8.04 ^a , ²³² Th 8.62 ^a , ²¹⁰ Pb 35.86 ^a , ⁴⁰ K 72.84 ^a ,	109
	$\mathrm{Bq}~\mathrm{kg}^{-1}$	
	$^{22\hat{6}}$ Ra (29.8–46.3), 228 Ra (8.5–10.2) Bq L $^{-1}$	134
Tunicina	²²⁶ Ra 19 ^a , ²³² Th 39.9 ^a , ⁴⁰ K 66 ^a Bq kg ⁻¹	23
Iraq	²³⁸ U 4.12 ^a , ²³² Th DL ^a , ⁴⁰ K 14.16 ^a , ¹³⁷ Cs 11.16 ^a Bq	41
	L ⁻¹	100
	²²⁶ Ra 20.3 ^a , ²³² Th 9.4 ^a , ⁴⁰ K 66.4 ^a Bq kg ⁻¹ ²²⁶ Ra 1.20 ^a , ²³² Th 9.4 ^a , ⁴⁰ K 66.4 ^a Bq kg ⁻¹	100
Iran	²²⁶ Ra (<dl-28) 12.35<sup="">a, ²²⁸Ac (<dl-10) 4.88<sup="">a, ²³⁵U</dl-10)></dl-28)>	101 135
iiaii	(<dl-1.6) 0.71<sup="">a, ²¹⁴Pb (<dl-334.1) 142.97<sup="">a, ²¹⁴Bi</dl-334.1)></dl-1.6)>	133
	(<dl-471) 178.78<sup="">a, ⁴⁰K (4.4-43.7) 24.29^a, ²³⁸U</dl-471)>	
	(30-211) 126.88 ^a , ²⁰⁸ Ti $(4-44.3)$ 21.36 ^a , Bq L ⁻¹	
Poland	40 K (5-499) 75 ^a , 238 U < 30 ^a , 226 Ra <2 ^a , 210 Pb < 5 ^a ,	136
	228 Ra $< 2^{a}$, 228 Th $< 2^{a}$ Bq L ⁻¹	
Гurkey	²²⁴ Ra (<1 to 4) 2.83 ^a , ²²⁶ Ra (<3 to 10) 6 ^a , ²²⁸ Ra (<1	94
Domania	to 4) $3.17^{\rm a}$ Bq L ⁻¹ 238 U (0.043–1.1), 226 Ra (23–45), 232 Th (0.2–8), 40 K	127
Romania	0 (0.043-1.1), Ra $(23-43)$, III $(0.2-8)$, K $(221-899)$ Bq L ⁻¹	137
Omán	²²⁸ Ac (1019–1040) 1030 ^a , ²²⁶ Ra (514–529), ⁴⁰ K	138
Oman	$(1522-1535)$ 1528^{a} , Bq L ⁻¹	100
Ghana	²³⁸ U (0.11–1.03) 0.54 ^a , ²³² Th (0.21–0.56) 0.41 ^a ,	139
	40 K (1.65–11.99) 7.76° Bq kg ⁻¹	
	²³⁴ U (<dl-6.10), <sup="">238U (<dl-5.50), <sup="">210Po (22-145), ²³⁰Th (2.9-15), ²³²Th (1.6-5.6) Bq L⁻¹</dl-5.50),></dl-6.10),>	140
	230 Th (2.9–15), 232 Th (1.6–5.6) Bq L ⁻¹	
	²²⁶ Ra (6.20–22,30), ²²⁸ Ra (6.40–35.50), ²²⁸ Th	82 and 140
	(0.71-6.41), ²²⁴ Ra $(0.78-7)$, ⁴⁰ K $(5.90-23.90)$ Bq L ⁻¹	
Nigeria	$^{ m L}$ $^{226}{ m Ra~8.9}^{ m a}, ^{228}{ m Ra~8.1}^{ m a}, ^{40}{ m K~39.8}^{ m a}~{ m Bq~L}^{-1}$	141
ivigeria	²²⁸ Ra (0.75–12.30) 5.18 ^a , ²²⁶ Ra (2.01–13.19)	141
	6.04° , 40 K (9.08–155.22) 48.78 $^{\circ}$, Bq L ⁻¹	172
Argentina	U (<10.0 to 33.0) μ g L ⁻¹ , ²²⁶ Ra (<1.7 × 10 ⁻³ to	143
-	26.8), 228 Ra (<1.1 × 10 ⁻³ to 9.6) Bq L ⁻¹	

Table 4 (Contd.)

	Concentrations of the TENORM in produced	
Country	water with their range, averages and units	Ref
JSA	²²⁶ Ra (56–1494) 367.5 ^a , ²²⁸ Ra (69–600) 275.75 ^a ,	144
	$pCi L^{-1}$	
	$^{\hat{2}26}$ Ra (30–2690), 228 Ra (35–763) Bq L $^{-1}$	145
	²²⁶ Ra (<0.002 to 58) 11.7 ^a , ²²⁸ Ra (0.02–59) 15.5 ^a	146
	$\rm Bq~L^{-1}$	
	226 Ra (548–3970), 228 Ra (83–1080), pCi L $^{-1}$	147
	²³⁸ U (0.0003–0.10), ²²⁶ Ra (0.002–1200), ²¹⁰ Pb	114
	(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 ,	148
	137 Cs DL ^a , 40 K (DL–265)), pCi L ⁻¹	
	226 Ra (ND-13 033), 228 Ra (ND-1485) Bq L $^{-1}$	149
	226 Ra (0.02–0.24), 228 Ra (0.02–0.17) pCi ML $^{-1}$	150
	²²⁶ Ra (0.40–70.30) pCi gm ⁻¹	151
zerbaijani	U (Ra) (ND-101.07), Th (ND-13.71), K (26.1-	152
	194.5) Bq L^{-1}	
K	226 Ra (0.16–90), 228 Ra (0.5–12) Bq L $^{-1}$	48
	226 Ra (0.002–1200), 228 Ra < 0.001 a Bq L $^{-1}$	154
kraine	²²⁴ Ra (1.57–5.51), ²²⁶ Ra (27.40–39.80), ²²² Rn	153
	(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}	

^a () the data between the brackets means the range/^a 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 ²³⁸U and ²³²Th 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 ²²⁶Ra, ²²⁸Ra, and ²²⁴Ra, and other radioactive nuclides, were reported in the natural decay of ²³⁸U and ²³²Th series and ⁴⁰K 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	158-160
	kg^{-1}	
	$^{226}{ m Ra}~(120.80-3500),~^{228}{ m Ra}~(147.90$ –2195) kBq kg $^{-1}$	160
	^{kg} ²²⁶ Ra (77.90–2110) 897.50 ^a , ²²⁸ Ra (101.50–1550)	160 and 161
	679.50 ^a , ⁴⁰ K (16–144.60) 65.10 ^a kBq kg ⁻¹	100 and 101
	226 Ra (109.60–2110) Bq kg $^{-1}$	154
	²²⁶ Ra (16.10–93.20) 37 ^a , ²²⁸ Ra (4.04–36.8) 23 ^a ,	154
	228 Th (4.46–18.5) 10.50 $^{\rm a}$ kBq kg $^{-1}$ 226 Ra (120.80–955), 228 Ra (131.40–792) kBq kg $^{-1}$	162
Syria	²²⁴ Ra (27–115), ²²⁶ Ra (147–1050), ²²⁸ Ra (43–181),	163
5,114	²¹⁰ Po (224–2371), ²¹⁰ Pb (<12 to 174) Bq g ⁻¹	100
	²²⁶ Ra (0.3–1520) 174, ²²⁴ Ra (0.1–780) 67, ²²⁸ Ra	163
	$(0.6-868) 91^{a} \text{ Bq kg}^{-1}$	
	²²⁶ Ra (5.90–215), ²²⁴ Ra (1.90–96.10), ²²⁸ Ra (2.30–	132
	83.30), Bq kg ⁻¹	4641465
	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	164 and 165
	Ra (226–306), Pb (263–42) Bq g 226 Ra (1317–2039), 210 Pb (843–1036) Bq g $^{-1}$	166 167
UK	²²⁶ Ra (58.80–131.60), ²¹⁴ Pb (47.50–119.70), ²¹⁴ Bi	168 and 169
	(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	154
Tunicina	238 U (0.9–7.6), 232 Th (0.8–5.1), 226 Ra (31–1189) Bq kg ⁻¹	129
	ьц ку ²²⁶ Ra (51–136) 76.5 ^a , ²³² Th (59–177) 98 ^a , ⁴⁰ К	23
	$(5.1-105) 25.03^{a} \text{ Bq kg}^{-1}$	23
Italia	²³⁸ U (0.9–53.8) 13.21 ^a , ²³² Th (0.8–18.9) 3.39 ^a ,	129
	²²⁶ Ra (2.7–1126) 351.1 ^a Bq kg ⁻¹	
Congo	²³⁸ U (0.9–2.7) 1.8 ^a , ²³² Th (0.8–2.2) 1.5 ^a , ²²⁶ Ra	129
	(97–151) 124 ^a Bq kg ⁻¹	
Saudi Arabia	238 U < LDa, 232 Th (0.14–3.10) 1.80a, 226 Ra (0.80–1.50) 1.10a, 40 K (4.30–9.60) 7.40a, 235 U < LDa,	1
	1.50) 1.10, $\frac{1}{1}$ K (4.30–9.60) 7.40, $\frac{1}{1}$ C < LD,	
Egypt	²³⁸ U (4500–11 800) 7100 ^a , ²²⁶ Ra (43 000–	20 and 43
-5J PC	120 000) 68.9 ^a , ²¹⁴ Pb (41 900–97 500) 66 500 ^a ,	20 4114 10
	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	
	$252\ 000\ , \mbox{K}\ (1100-4800)\ 1300\ , \mbox{Ra}\ 24\ 000$ Bq kg $^{-1}$	
	²²⁶ Ra (7541–143 262), ²²⁸ Ra (35 460–368 654),	32
	²¹⁴ Pb (18 215–322 604), ²¹⁴ Bi (17 627–320 008),	02
	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	170
	621.79 ^a , Bq kg ⁻¹	2
	²³⁸ U (9140 ⁻ 28 ⁻ 5 823) 147 481.5 ^a , ²³² Th (427- 34 339) 17 383 ^a , ⁴⁰ K (51-1031) 541 ^a Bq kg ⁻¹	2
	²²⁶ Ra (493–519) 506 ^a , ²²³ Th (35.38–54.08) 44.7 ^a ,	171
	40 K (LD-0.177) 0.059 $^{\rm a}$ Bq kg $^{-1}$	1,1
	²²⁶ Ra (7–314.8), ²²³ Th (<dl-177.3), <sup="">40K (<dl-< td=""><td>98</td></dl-<></dl-177.3),>	98
	223.0), $\mathrm{Bq} \ \mathrm{kg}^{-1}$	
	²³⁸ U 2500 ^a , ²²⁶ Ra 11 700 ^a , ²¹⁰ Pb 12 800 ^a , ²²⁸ Ra	134
	$4200^{\rm a}$, 224 Ra $3900^{\rm a}$ Bq kg ⁻¹	
	226 Ra (13–31), 228 Ra (5–9), 40 K (0.271–0.324) kBq kg $^{-1}$	77
	кд ²²⁶ Ra (14.1–8.9), ²²⁸ Ra (5.0–9.5) kBq kg ⁻¹	43
	na (14.1-0.3), na (3.0-9.3) kdy kg	43

Table 5 (Contd.)

	Concentrations of the TENORM in scales with	
Country	their range, averages and units	Ref
	226 Ra $68.9^{\rm a},^{228}$ Ra $24^{\rm a}$ Bq ${ m g}^{-1}$	172
	²³⁸ U (568–729) 660 ^a , ²²⁶ Ra (2874–2197) 1979 ^a ,	16
	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}	
Malaysia	²²⁶ Ra (550–434 000) 114 300 ^a , ²²⁸ Ra (900–	173
	479 000) 130 120 ^a (Bq kg ⁻¹)	
Norway	²²⁶ Ra (4.9–100), ²²⁸ Ra (0.40–28.90), ²¹⁰ Pb (<0.2–	174
	49) Bq g ⁻¹	
German	²²⁶ Ra (850–100 000), ²²⁸ Ra (LD–240 000), ²¹⁰ Pb	22
	(1400–70 000), 228 Th (LD–48 000), 227 Ac (LD–2500) Bq kg $^{-1}$	
irod	²³⁸ U (11.22–32.62) 19.15 ^a , ²³² Th (6.28–26.56)	41
Iraq	18.10 ^a , ⁴⁰ K (51.22–277.59) 195.98 ^a , ¹³⁷ Cs (DL–	41
	3.05) 2.33^{8} Bq kg ⁻¹	
	²²⁶ Ra (137.6–152.4), ²³² Th (34.4–49.4), ⁴⁰ K (462–	100
	750.3) Bq kg ⁻¹	100
	²²⁶ Ra (61.80–128.40), ²³² Th (12.80–26.80), ⁴⁰ K	101
	(82.60–83.10) Bq kg $^{-1}$	
USA	²¹⁰ Pb 1370 ^a , ²²⁶ Ra 6230 ^a , ²²⁸ Ac (²²⁸ Ra) 565 ^a Bq	175 and 176
	$ m kg^{-1}$	
	²²⁶ Ra (15 400-76 100) Bq kg ⁻¹ , ²²² Ra (0.037-	177
	0.153) emanation fraction	
	⁴⁰ K (ND-0.87), ²²⁶ Ra (ND-1.90), ²²⁸ Ra (ND-4.17)	148
	$ m pCi~g^{-1}$	
	²³⁸ U (0.001–0.50), ²²⁶ Ra (0.1–15 000), ²¹⁰ Pb	105
	(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	178
	kg ⁻¹ ²²⁶ Ra 13.3 ^a , ²¹⁰ Pb 13.3 ^a , ²¹⁰ Po 13.3 ^a , ²²⁸ Ra 4.44 ^a ,	470
	Ra 13.3 , PD 13.3 , PO 13.3 , Ra 4.44 , ²²⁸ Th 4.44 ^a Bq kg ⁻¹	179
	¹¹¹ 4.44 в q кд ²²⁶ Ra (0.84-7.38), ²²⁸ Ra (0.16-2.283) pCi g ⁻¹	147
Γurkey	²²⁴ Ra (<1 to 520) 67.95 ^a , ²²⁶ Ra (13–132 000)	94
Turkey	20 614.24 ^a , ²²⁸ Ra (<1 to 453) 63.71 ^a Bq kg ⁻¹	74
Romania	²²⁶ Ra (20–9260) 3200 ^a Bq kg ⁻¹	137
Syria	²²⁶ Ra (5900–215 000) 119 830, ²²⁸ Ra (2300–4000)	22
.,,	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 ,	180
	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	
	$ m kg^{-1}$	
	²²⁶ Ra (28.22–47.54), ⁴⁰ K (42.65–44.05), ²³² Th	104
	$(16.90-40) \text{ Bq kg}^{-1}$	
Argentina	U (<0.4 to 1.9) $\mu g g^{-1}$, ²²⁶ Ra (<0.1 to 1270), ²²⁸ Ra	143
	(115–1670) Bq g ⁻¹	
Algeria	²²⁶ Ra (1–950) Bq g ⁻¹	95
Kazakhstan	²²⁶ Ra (510–51 000) 18 101 ^a , ²²⁸ Ra (200–10 000)	181
	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	²²⁸ Ra (0.03–7.93), ²³² Th (0.02–5.09), ⁴⁰ K (ND–	182
Nussia	$^{\text{Ra}}$ (0.03–7.93), $^{\text{Th}}$ (0.02–5.09), $^{\text{R}}$ (ND–2.28), kBq kg $^{-1}$	182
ndonesia	^{2.28} f, kbq kg ²²⁶ Ra (78–36 106), ²²⁸ Ra (127–222 974), ²²⁸ Th	102
HUOHESIA	(144-90 887), ²³⁸ U (LD-13 030), ²³² Th (LD-6024)	183
	(144-90 887), W (LD-13 030), W III (LD-6024) 40 K (20-5496), Bq kg ⁻¹	
Sudan	²³⁸ U (358.05–4106.9), ²³² Th (375.88–2736.7), ⁴⁰ K	106

 $[^]a$ () the data between the brackets means the range/ a is 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 ,	163
Indonesia	(Bq kg ⁻¹) ²³⁸ U (12.40-98), ²³² Th (4-148), ²²⁶ Ra (40-6129), ⁴⁰ k (105-268), ²²⁸ Ra (515-7993), ²²⁸ Th (135-	185
Tunicina	2198) Bq kg ⁻¹ ²³⁸ 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	1
Malaysia	¹ ²²⁶ 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	$^{\mathrm{Ag}}$ $^{226}\mathrm{Ra}$ (<0.2 to 44.3) 20.71 $^{\mathrm{a}}$, $^{228}\mathrm{Ra}$ (<0.02 to 43.89) 7.96 $^{\mathrm{a}}$, $^{40}\mathrm{K}$ (<0.2 to 19.44) 9.58 $^{\mathrm{a}}$ BBq Lq kg $^{-1}$	93
	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	171
	226 Ra 18 032 ^a , 214 Pb 19 394 $^{\overline{a}}$, 214 Bi 18 324 a , 228 Ac 13 257 a , 212 Bi 7398 a , 208 Tl 5105 a , 40 K 1261 a Bq kg $^{-1}$	32
	$^{18}_{238}$ U (12.3–29.6), 232 Th (14.63–28.22), 40 K (789.4–1680.5) Bq kg $^{-1}$	133
	238 U 108 ^a , 226 Ra 11 960 ^a , 226 Ra 0.009 ^a , 228 Ra 1750 ^a Bq kg ⁻¹	186
	226 Ra (13.10–94.20), 232 Th (1.50–27.70), 40 K (ND–81.80) Bq kg $^{-1}$	99
	²²⁶ Ra (5.50–1785.80), ²³² Th (<dl-885), <sup="">40K (<dl-880) bq="" kg<sup="">-1</dl-880)></dl-885),>	98
	²²⁶ Ra 14.67 ^a , ¹²³² Th 9.99 ^a , ²¹⁰ Pb 68.04 ^a , ⁴⁰ K 112.82 ^a Bq L ⁻¹	170
	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	187 171
	$ m kg^{-1}$ 238 U 108^{a} , 226 Ra $11~963^{a}$, 210 Pb 2290^{a} , 228 Ra	134
Norway	$1747^{\rm a}, ^{224}{ m Ra}~1904^{\rm a}~{ m Bq}~{ m kg}^{-1}$ $^{226}{ m Ra}~(<0.1~{ m to}~4.7)~2.5^{\rm a}, ^{228}{ m Ra}~(<0.1~{ m to}~4.6)~2.1^{\rm a},$ $^{210}{ m Pb}<0.7^{\rm a}~{ m Bq}~{ m g}^{-1}$	188
Brasil	²²⁶ Ra (50–167.80), ²²⁸ Ra (48.60–152.40), kBq kg ⁻¹	36
	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)	158–160 162 160 and 161
Albania	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	189
	(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-	5
Brazil	25.60), 228 Th 23a , 137 Cs 7a Bq kg $^{-1}$ 226 Ra $^{(0.20-265)}$ 107a , 228 Ra $^{(0.11-244)}$ 77a , 228 Th $^{(<0.1)}$ to 172) 77a kBq kg $^{-1}$	190
Iraq	²³⁸ U 35.04 ^a , ²³² Th 15.22 ^a , ⁴⁰ K 149.01 ^a , ¹³⁷ Cs 1.82 ^a Bq kg ⁻¹	27 and 41
	1.82 Eq kg ²²⁶ Ra (235.4–321.8), ²³² Th (48.9–140.8), ⁴⁰ K (502.7–800.8) Bq kg ⁻¹	100
	$^{(502.7-800.8)}$ Bq kg 226 Ra (1.80–38.10), 232 Th (6.80–14.40), 40 K (12.10–242.10) Bq kg $^{-1}$	101
USA	(12.10-242.10) Bq kg ²¹⁰ Pb 5148 ^a , ²²⁶ Ra 59 000 ^a , ²²⁸ Ac (²²⁸ Ra) 28 501 ^a Bq kg ⁻¹	175 and 176

Table 6 (Contd.)

	Concentrations of the TENORM in sludge with	
Country	their range, averages and units	Ref
	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 $^{-1}$	105
	Ra (0.5-50) Bq g 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	179
	⁴⁰ K 22.20 ^a , ²²⁶ Ra 2.34 ^a , ²²⁸ Ra 3.60 ^a , pCi g ⁻¹	148
	226 Ra (121–872) pCi g $^{-1}$	149
Poland	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	136
Turkey	224 Ra (<1 to 178) 3 0 .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) μ 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) Bg 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 $(<0.20 \text{ to } <0.30)$, 228 Ra $(0.0094-0.0636)$, 228 Th $(<0.11 \text{ to } <0.15)$ 40 K $(0.0492-0.154)$ Bq g $^{-1}$	191
	²²⁸ Ac (1019–1040) 1030 ^a , ²²⁶ Ra (514–529), ⁴⁰ K (1522–1535) 1528 ^a , Bq L ⁻¹	138
Qatar	226 Ra (394.49–27 884.9), 232 Th (30.6–94.07), 40 K (62.8–110.6) Bq kg $^{-1}$	192
Irán	²²⁶ Ra 41 ^a , ²¹⁰ Pb 70 ^a , ²¹⁰ Po 92 ^a	193
	226 Ra (6.80–466.30), 232 Th (ND–47.6), 40 K (ND–154.70), Bq kg $^{-1}$	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)	179
	210 Pb $815^{\rm a}, ^{226}$ Ra $1~481^{\rm a}, ^{228}$ Ac $(^{228}$ Ra $)~251^{\rm a}$ Bq kg $^{-1}$	175 and 176
	226 Ra (0.15–120), 228 Ra (ND–17) Bq L $^{-1}$	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 $^{0.037^{\rm a}}$, 232 Th $^{0.027^{\rm a}}$, 40 K $^{0.22^{\rm a}}$ 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 $^{-1}$	170
	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 $^{-1}$	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
Country		Rei
Saudi Aribia	²³⁸ U (19.0-62.8) 39.0, ²²⁶ Ra (8.68-156) 23.2 ^a ,	196
	²³² Ra (2.76–12.8) 7.73 ^a , ⁴⁰ K (108–446) 278 ^{a 40} Cs (<mda–3.83) 1.42<sup="">a Bq kg⁻¹</mda–3.83)>	
Povint	(SMDA-3.83) 1.42 Bq kg ²²⁶ Ra (7541–194 489), ²¹⁴ Pb (18 215–437 960),	165
Egypt	²¹⁴ Bi (17 627–434 435), ²²⁸ Ac (35 460–897 803),	103
	²¹² Bi (LDL–500 476), ²⁰⁸ Tl (LDL–8645), ⁴⁰ K	
	$(LDL-45~882)~Bq~kg^{-1}$	
Гипісіпа	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 , ²²⁸ Ra (14–25) 20.7 ^a , ²³⁸ Th	189
	$(12-25) 20.3^{a}, {}^{40}K (326-549) 413.7^{a} \text{ Bq kg}^{-1}$	
raq	²²⁶ Ra (18.4–97.6), ²³² Th (11.5–42.7), ⁴⁶ K (176.9–	100
	485.6) Bq kg ⁻¹ ²²⁶ Ra (13.2–15.90), ²³² Th (25–35.20), ⁴⁰ K (385.5–	
	432.2) Bg kg ⁻¹	
	²²⁶ Ra 33.588 ^a , ²³² Th 20.647 ^a , ⁴⁰ K 511.604 ^a Bq	203
	kg^{-1}	200
Turkey	²²⁴ Ra (<10-535) 223.67 ^a , ²²⁶ Ra (24.79-70.48)	94
·	46.47^{a} , 228 Ra (<11-423) 186.67 a Bq kg $^{-1}$	
Rusia	TPH (1.6–880.3) 223.23 ^a g kg ⁻¹ , ²²⁶ Ra (0.03–	204
	7.93) 2.62^{a} , 232 Th $(0.02-5.09)$ 1.28^{a} , 40 K $(0.03-$	
	$2.28) 0.56^{a} \text{ Bq kg}^{-1}$	
Domania	226 Ra 2 2 2 2 Th 3 2 40 K 311 8 8 1 2 8 1 238 U $^{2.4}$ $^{-1}$ 20 Ra 20 Ra 20 Ra 20 Ra 20 Th 20 Rh 20 N 20	182
Romania	(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	204
Jilalia	(60.4-248.9) 157.0° Bq kg ⁻¹	204
	238 U (1.60–21.3) 8.65 ^a , 232 Th (2.78–32.2) 12.5 ^a ,	205
	⁴⁰ K (111–528) 214 ^a Bq kg ⁻¹	
Nigeria	²²⁶ Ra (19.2–94.2) 41.0 ^a , ²³² Th (17.7–47.5) 29.7 ^a ,	206
	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–	207
2-1	41.37) Bq kg ⁻¹	400
Qatar Canadá	226 Ra 2 Ca 5 Ra 232 Th 1 Ch 4 Ra 1 Ra 40 K 21 Ca 69 Ra 1 Ca 226 Ra 200 Ra 238 Ra 238 Ra 200 Ra 228 Th 2	192 208
Janada	$^{\text{Ra}}$ (0.01–3.84), $^{\text{Ra}}$ (0.01–0.007), $^{\text{TH}}$ (ND–0.05) Bq $^{\text{g}}$	208
USA	²¹⁰ Pb 815 ^a , ²²⁶ Ra 1481 ^a , ²²⁸ Ac (²²⁸ Ra) 251 ^a Bq	175 and 176
	kg^{-1}	170 4114 17
	²²⁸ Ra (9–1883), ²²⁶ Ra (12–2802), ²¹⁰ Pb (ND–209),	209
	228 Th (11–628) Bq kg $^{-1}$	
	⁴⁰ K (10–19.70), ²³⁸ U (ND–1.11), ²²⁶ Ra (ND–1.43),	184
	²²⁸ Ra (0.82–1.22), pCi g ⁻¹	
	²¹⁰ Pb 1370 ^a , ²²⁶ Ra 6230 ^a , ²²⁸ Ac (²²⁸ Ra) 565 ^a Bq	178
Zuwoit	$ m kg^{-1}$ 232 Th (8.70–27.90) 226 Ra (9.80–42.30), 40 K	210
Kuwait	(191.2-632) Bq kg $^{-1}$	210
	²³² Th 12.53 ^a , ²²⁶ Ra 10.65 ^a , ⁴⁰ K 300 ^a Bq kg ⁻¹	111
	²³² Th (10.52–21.96) 13 ^a , ²²⁶ Ra (33.7–250.6) 85 ^a ,	46
	40 K (331–449.4) 406 $^{\rm a}$ Bq kg $^{-1}$	
Syria	²²⁶ Ra (18.90–210), ²³² Th (16.80–55.90), ⁴⁰ K (44–	131
	213) Bq kg $^{-1}$	
	²²⁶ Ra (1030–7780) Bq kg ⁻¹	212
	²²⁶ Ra (1.34–7.75), ²²⁸ Ra (0.34–2.42), ²²⁴ Ra (0.30–	213
hina	2) Bq kg ⁻¹ ²²⁶ Ra (16–82) 57 ^a Bq kg ⁻¹	014
China Omán	²²⁶ Ra (30.4-41.2) 34.2 ^a , ²²⁸ Ra (5.7-11.3) 8.1 ^a ,	214 213
/111a11	228Th (5.7–9.1) 7.1 ^a , ⁴⁰ K (93–293) 151 ^a , ¹³⁷ Cs	213
	$(<0.12 \text{ to } 1.02) \text{ Bq kg}^{-1}$	

 $[^]a$ () 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

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

^a () the data between the brackets means the range/^a is 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 overexposure. 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 210Pb, and also those emitting alpha such as ²¹⁰Po. 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	147
	²⁰⁸ Tl (0.21–0.49), ²²⁶ Ra (0.83–2.25), ²²⁸ Ra (0.63–	
	11 (0.21–0.49), Ra (0.83–2.25), Ra (0.03–1.63), 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	217
Poland	⁴⁰ K (680–999), ²³² Th (23.30–31) Bq kg ⁻¹	218
	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	136
	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	136

 $[^]a$ () the data between the brackets means the range/*is 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
	Samples	their range, averages and annes	
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),	219
Romania	Spontaneous vegetation	⁴⁰ K (66.80–70.20) Bq kg ⁻¹ ²³⁸ 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 ÷ 990 a Bq kg $^{-1}$	154
Nigeria Azerbaiyán	Clay Rock	226 Ra 28.7 ^a , 232 Th 67.1 ^a Bq kg ⁻¹ 226 Ra (ND–96.70), 232 Th (ND–18.94),	220
China	Limestone Shale Sediments	⁴⁰ K (115.40–467.30), Bq kg ⁻¹ ²²⁶ Ra (13–29) 20 ^a Bq kg ⁻¹ ²²⁶ Ra (114–183) 149 ^a Bq kg ⁻¹ ²²⁶ Ra (306–396) 351 ^a Bq kg ⁻¹	214
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 Rock	²²⁶ Ra (0.2-1), ²²⁸ Ra (0.2-1) pCi ML ⁻¹ ²²⁶ Ra (0.1-1), ²²⁸ Ra (0.1-1) pCi ML ⁻¹ ²³⁸ U (0.1-70 000), ²³² Th (0.1-20 000)	222 221
	ROCK	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 TEN-ORM 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.82,133,227-230 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:

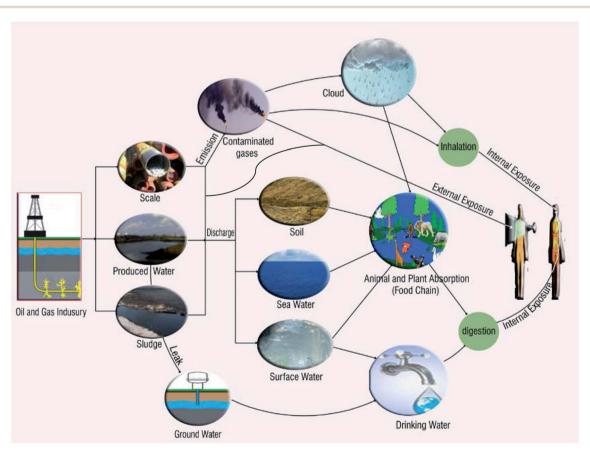


Fig. 3 The main forms of petroleum pollution of TENORMs.^{242,261}

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 TEN-ORMs 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 $\rm g^{-1}$. 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 (Ra_{eq}), dose-absorbed dose (D_{yr}), and effective annual dose rate $(D_{\rm eff})$. 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:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_{K}$$

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

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

where Ra_{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; $D_{\gamma r}$ 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 12).

Overall, when comparing studies done to measure TEN-ORMs 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	$Ra_{eq} (Bq kg^{-1})$	$D_{\gamma r} \left(\text{nGy h}^{-1} \right)$	$D_{\rm eff} m (mSv y^{-1})$	Ref
Saudi Arabia	(21.6–204) 62.1	(10.5-96.4) 29.3	(0.013-0.118) 0.038	196
	1.35-173	<ld-83.6< td=""><td>· · ·</td><td>1</td></ld-83.6<>	· · ·	1
Egypt	$(70.4 – 213.9) \times 10^3$	$31-72.7 \times 10^3$		42
	$(544-596)\ 570 \times 10^3$	(250-273) 262	(306-335) 321	171
Tunisian	0.09–398	0.043-177	5.28×10^{-5} to 3.64×10^{-4}	23
Algeria		$(0.1100) \times 10^3$	(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
	(10.01–128.92)	,	,	104
Nigeria	,	$(21-117.5) 54.6 \times 10^3$	(0.03-0.2) 0.07	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
	(5.21-79.84) 47.39	(2.49–37.53) 22.18	(0.012 - 0.184) 0.109	39
USA	,	,	(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 TEN-ORMs 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.

Acknowledgements

This paper is funded by the International Exchange Program of Harbin Engineering University, China Scholarship Council (CSC) for Innovation-oriented Talents Cultivation.

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