NUKLEONIKA 2009;54(1):3–9

REVIEW PAPER

Technologically enhanced naturally occurring radioactive materials in the oil industry (TENORM). A review

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Abstract. A large amount of naturally occurring radioactive materials in the form of by-products or waste is produced annually by the growing activity of the oil and gas industry. Solid scale, sludge and produced water are typical residues contaminated with natural radionuclides from the uranium and thorium series, particularly ^{226}Ra and ^{228}Ra. The observed specific activities of these radionuclides are in the ranges up to 3700 kBq/kg and up to 168 kBq/kg for solid scale and sludge, respectively. The average activities of both radionuclides exceed the exemption level of 10,000 Bq/kg recommended by IAEA safety standards. This means that TENORM wastes from the oil industry may generate radiation exposure levels which require attention and continuous monitoring during some routine operations in this industry. This exposure is mostly caused by external γ-radiation coming from the ^{226}Ra radionuclide and its progenies.

Key words: oil industry • radiation exposure • radionuclides

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Received: 18 July 2008
Accepted: 10 October 2008

Introduction

Radionuclides of natural origin are present in the whole environment in which we live. It has even been suggested that the long term emission of alpha particles from natural radionuclides could be one of the possible sources of energy associated with the transformation of organic matter into petroleum [39]. In recent decades, the development of new technologies has resulted also in the production of by-products and waste with the so-called technologically enhanced naturally occurring radioactive materials (TENORM). Therefore, human technical activity can increase radiation exposure, not only to the person directly involved in these activities, but also to the local or even whole population. The first evaluations of occupational radiation exposure in the oil and gas industries were reported a few decades ago [19, 32, 47]. The management of the waste from these industries containing TENORM and the evaluation of a potential radiation hazard have been the subject of continuous activity of radiological protection specialists in recent years [27, 34, 48].

If low levels of radiation are proven to be carcinogenic, or have some other detrimental effects, then current regulatory efforts must protect the public and workers. Therefore, any regulations for the optimisation of radiological protection from TENORM should take into account the additional risk over and above exposure to local natural radiation. The International Atomic Energy Agency (IAEA) published comprehensive radiation safety standards based on the recommendations of

Table 1. EC Clearance levels

Radionuclide	Quantity	Concentration
^{40}K	106 Bq (27 mCi)	100 Bq/g (2.7 nCi/g)
^{226}Ra and progeny	10,000 Bq (270 nCi)	10 Bq/g (270 pCi/g)
^{232}Th (secular equilibrium)	1000 Bq (27 nCi)	1 Bq/g (27 pCi/g)

the International Commission on Radiation Protection, which have been recommended for adoption as Basic Safety Standards (BSS) by all European Community Countries [26].

On the basis of these recommendations, several countries have introduced their own regulation for NORM and classification of TENORM by setting clearance or exemption levels for selected radionuclides for discharge. These regulations were presented in the IAEA's workshop materials [28].

In the BSS there are recommended exposure limits and exemptions from various sources of radioactivity, including NORM. The most important limits are as follows:

- Maximum annual dose limit of 1 mSv (100 mrem) to members of the public, with a provision for allowing higher doses in any single year, provided that the average over five consecutive years does not exceed 1 mSv per year.
- The limit on an effective dose for exposed workers shall be 100 mSv (10 rem) in a consecutive five-year period, subject to a maximum effective dose of 50 mSv (5 rem) in any single year.
- Establishing the so-called clearance levels for releasing materials and items with concentrations and total activity below specific levels.

The clearance levels for some important radionuclides occurring in the oil industry are shown in Table 1.

Observed levels of natural radionuclides in the oil industry

Natural radionuclides from the uranium and thorium series are also present in geological formations containing crude oil or gas. Average concentrations of uranium and thorium in the earth's crust are around 4 and 12 ppm, respectively. This corresponds to the mean value of ~ 40 Bq/kg in the specific radioactivity units. Both parent radionuclides, ^{232}Th and ^{238}U, and the majority of their radioactive decay products, present in the crust, are insoluble in adjacent organic fluids. Therefore, the concentrations of radionuclides from these series in the crude oil layers, except for gaseous radon isotopes, are much lower in comparison to average values observed in soil. The radionuclides of primary concern to the oil and gas industry are ^{226}Ra (^{238}U decay) and ^{228}Ra (^{232}Th decay) due to their radiotoxicity and relatively long half-lives (1620 and 5.75 years, respectively). For example, reported values of the activity levels for ^{226}Ra in US, Algerian and Nigerian crude oils are in the ranges of 0.1 ÷ 40 Bq/kg [46], 6 ÷ 20 Bq/kg [22] or 0.4 ÷ 1.3 Bq/kg [1], respectively. Radon radionuclides escaping from the adjacent geological formations are soluble in crude oil, but due to its half-life (3.825 days) only ^{222}Rn is present in the pumped oil in varying concentrations from 10 to 800 Bq/kg [23]. Much broader ranges of the radon concentrations may occur in natural gas (NG) including natural gas liquids (NGL). Many previous data on the distribution of the main radon nuclide ^{222}Rn in different gas fields and processing plants were collected in the UNSCEAR report [56]. For example, the radon concentrations in North Sea fields are relatively low and vary between 74 and 148 Bq/m^3, whereas the highest concentration were observed for British Columbia (~ 20,000 Bq/m^3) and US fields (up to 54,000 Bq/m^3). The typical activities of ^{222}Rn in Algerian and Middle East NG are in the range 15 to 1200 Bq/m^3 [3, 23].

In interstitial rock spaces, in addition to oil or gas, water is also present in varying amounts (so-called formation water). Crude oil is usually pumped to the surface together with formation water containing dissolved or partially precipitated mineral salts, together with radon and mainly radium radionuclides since uranium and thorium usually do not go into solution. However, in contrast to various cations such as barium, calcium, strontium or sodium, together with anions such as sulphate, chloride or the bicarbonate solubility of radon in water is lower, and the observed ^{222}Rn concentrations in the petroleum formation water are in the range up to 18.5 Bq/dm^3 [23]. Under the reducing conditions in the formation waters, U and Th are also present in very low concentrations. Radium nuclides released by alpha recoil from the surrounding minerals or leaching processes are usually scavenged by sorption and their activities are also generally low, except for saline waters with high concentration of chloride anions – Cl^- [11].

In many offshore oil fields sea water is additionally injected to maintain pressure, and it mixes with the formation water. In such cases, in the exploited oil/water mixture, the content of the production water can reach even 95%. For this reason, the produced waters are typically saline and rich in Cl^- anions forming aqueous complexes with Ra that enhance the mobility of Ra nuclides from adjacent geological rocks into these waters [16].

Comprehensive older literature reviews of radium nuclide concentrations in formation and produced water indicated an average radium nuclide concentration in waters in excess of 1.85 Bq/dm^3 and exceptionally up to ~ 1000 Bq/dm^3 [29, 46, 59]. As ^{226}Ra originated from the radioactive decay of ^{238}U, while ^{228}Ra from ^{232}Th, the $^{226}Ra/^{228}Ra$ ratio in the oil-field brines depends on the U/Th ratio of the reservoir rock and ranges from 0.1 to 2.0, but for the most cases its activities are comparable. Typical ranges or average values of the radium radionuclide concentrations in the formation or produced water from different oil fields, including the recent data, are listed in Table 2.

A critical review of the intense studies of the activity concentrations of ^{226}Ra, ^{228}Ra as well as ^{210}Pb and ^{210}Po in produced water in 2003 from Norwegian oil and gas platforms located in the North Sea were also reported [36]. The concentrations of ^{226}Ra and ^{228}Ra in

Table 2. Ranges of activity levels in produced water from the oil fields

Field	Sample	^{226}Ra (Bq/dm^3)	^{228}Ra (Bq/dm^3)
Algeria [22]	Formation water	5.1–14.8	
Australia [21]	Produced water	17[a]	23[a]
Brazil [58]	Produced water	0.01–6	0.05–12
Congo [54]	Produced water	5.1[a]	
Egypt [42]	Formation water	5–40	1–59
Italy [54]	Produced water	0.2–2	
Norway [52]	Formation water	0.3–10.4	–
Norway [36]	Produced water	3.3[a]	2.8[a]
Norway [13]	Produced water	0.5–16	0.5–21
Syria [2]	Produced water	9.9–111.2	8.8–60.4
UK [55]	Produced water	1.7[a]	–
USA [46]	Produced water	0.1–60	–
USA [51]	Produced water	0.15–21.6	0.7–1.7
USA [53]	Oilfield brine	12.6[a]	15.1[a]
USA [61]	Produced water	22–30	25–30

[a] Mean activity concentration.

produced water discharged from these offshore platforms vary between 0.1 Bq/dm^3 and about 200 Bq/dm^3 with the average values estimated to be 3.3 Bq/dm^3 and 2.8 Bq/dm^3, respectively. Slightly higher radium activities ~ 10 Bq/dm^3 have been found for produced water outfalls in the Gulf of Mexico [50].

The worldwide average concentration of these radionuclides in produced water discharged to the environment is estimated at 10 Bq/l. These concentrations are approximately three orders of magnitude higher than natural concentrations of radium in drinking or sea water. Because the radium radionuclide concentrations in that waste water are usually below the clearance levels (see Table 1), it is recognized as a low specific activity waste and they may be injected into underground formations or disposed into the sea.

A comprehensive evaluation of discharges from the oil industry to the sea was done for European waters during the European Commission Marina Project [7]. The annual release of ^{226}Ra and ^{228}Ra with produced water from off-shore fields in Europe in the 1990s stabilized at around 5 TBq ($\times 10^{12}$ Bq) per year and 2.5 TBq per year, respectively. The commonly used two-step model of the radionuclide dispersing and diluting in the water in the vicinity of the oil platforms predicts a diluting factor up to 10^3 within minutes and within a few meters of the discharge source [10]. Therefore, additional radium nuclide concentrations in seawater of the local zone could be estimated as equal to around 5–10 Bq/m^3, in comparison with the natural concentration of around 1 Bq/m^3 for ^{226}Ra.

Unfortunately, there is no reported summary data of even approximate values concerning oil and gas industry related radionuclide discharged to the very important aquatic system of the Gulf of Mexico. However, on the same basic assumptions as for European water (a reference ratio between the volumes of produced oil and water equal to 0.33 and a ratio of 5×10^{-5} between the water production and standard cubic metre of produced gas), such a calculation can be done for radium nuclide discharges with produced water and their activity concentrations for the Gulf of Mexico water. Taking into account the mean daily oil production rate of 0.16 million m^3 (1.4 million barrels) and 0.23 billion m^3 of gas (8 billion cubic feet) in 2007 [31], one can simply calculate the total amount of the released produced water in this region in 2007 equal to 0.18 trillion m^3, and average radium nuclide activity (^{226}Ra or ^{228}Ra) annual release at around 2 TBq, very close to data for European waters. Assuming the same mean radium activities in the produced water as for the North Sea, a yearly release of 0.65 TBq for ^{226}Ra and 0.33 TBq for ^{228}Ra was appraised for offshore oil production from Argentina and Brazil [24].

In conclusion, it can be affirmed that discharges of NORM with produced water from the offshore oil and gas industry contribute usually at a very low level to the total concentration of the α-emitters in the marine environment, and only slightly enhanced levels of radioactivity in marine biota components can be observed in small local vicinities around the dispersing sites.

On the other hand, sea water injected into oil or gas containing geological formations disturbs the chemical equilibriums leading to the precipitation of some carbonate or sulphate salts. As a consequence of the physical and chemical processes during the extraction of oil, besides the production water, an additional waste product called scale is obtained. Scale production in gas and oil field equipment is due to precipitation of alkaline earth metal sulphates or carbonates according to the following chemical reactions:

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

Radium, strontium and barium are chemically similar and radium nuclides co-precipitate together with alkaline earth carbonates or sulphates, replacing calcium, barium or strontium cations in the crystal structures. The formation of scale is a complex phenomenon and it can be explained by the variation of the solubility of sulphates or carbonates by: temperature and pressure changes, evaporation in the gas extraction pipes and first of all, by water injection into the reservoirs, to maintain proper pressure during oil field exploitation.

Therefore, radium and radon concentrations in the pipe scale and waste sludge are dependent on the

Table 3. Ranges of activity levels of ^{226}Ra and ^{228}Ra in different scale and sludge samples

Oil field	Sample	^{226}Ra (Bq/kg)	^{228}Ra (Bq/kg)
Algeria [22]	Scale	1000–950,000	
Australia [21]	Scale	21,000–250,000	48,000–300,000
Brazil [20]	Scale	19,100–323,000	4210–235,000
Brazil [17]	Scale	121,000–3,500,000	148,000–2,195,000
Brazil [18]	Scale	77,900–2,110,000	101,500–1,550,000
Congo [54]	Scale	97–151	
Egypt [12]	Scale	68,900	24,000
Egypt [42]	Scale	7541–143,262	35,460–368,654
Italy [54]	Scale	< 2.7–2890	
Kazakhstan [30]	Scale	510–51,000	200–10,000
Malaysia [38]	Scale	114,300–187,750	130,120–206,630
Norway [33]	Scale	300–32,300	300–33,500
Saudi Arabia [4]	Scale	0.8–1.5	
Tunisia [54]	Scale	31–1189	
Tunisia [25]	Scale	4300–658,000	
UK [15]	Scale	1000–1,000,000	
USA [40]	Scale	up to 3,700,000	
USA [60]	Scale	15,400–76,100	
Australia [21]	Sludge	25,000	30,000
Brazil [20]	Sludge	50,000–168,000	49,000–52,000
Brazil [17]	Sludge	< LLD–413,000	< LLD–117,900
Egypt [42]	Sludge	18,000	13,250
Malaysia [38]	Sludge	6–560	4–520
Norway [33]	Sludge	100–4700	100–4600
Tunisia [54]	Sludge	66–453	

amount of Ra present in the subsurface soil, formation water components, and treatment processes applied during oil or gas production.

During formation of the scale, radium radionuclides are efficiently concentrated from the water phase. Therefore, the observed levels of activity concentrations both in the separated sludge and solid scale are much higher than those observed in the produced water from the oil industry. In the case of the ^{226}Ra radionuclide, as a result of its decay, a transient radioactive equilibrium (after one month's storage) can be settled and several daughter radioactive nuclides are produced. Among them, the most dangerous is the gaseous ^{222}Rn nuclide.

Reported levels of the ^{226}Ra and ^{228}Ra activity concentrations observed in the solid scale and sludge are listed in Table 3.

As shown in Table 3, the concentration levels of radium nuclides in scale vary within a wide range being much higher than those of the sludge. According to the latest EPA estimation, the average radium nuclide concentration is around 18,000 Bq/kg and 2800 Bq/kg in scale and sludge, respectively [37]. Elevated concentration activities of both radionuclides, exceeding the exemption level of 10,000 Bq/kg recommended by IAEA safety standards, were frequently found in the scale samples.

A large uncertainty is observed in the estimations of the total amount of radioactive waste generated by oil industry, and the EPA assumes that 100 tons of scale per oil well are generated annually in the United States [37], while for the North Sea wells a somewhat lower value of 20 t is suggested [14] and only 2.25 t per year by one oil-producing well for Latin American oil producing countries [49]. It was also estimated that approximately 2.5×10^4 and 2.25×10^5 tons of contaminated scale and sludge, respectively, were generated each year from the petroleum industry in the middle of the previous decade [45].

This means that TENORM waste from the oil industry may generate radiation exposure levels which require attention and continuous monitoring during some routine operation in this industry. This exposure is caused by external γ-radiation coming from the ^{226}Ra radionuclide and its progenies: ^{214}Pb and ^{214}Bi as well as by inhalation of α-emitting radionuclides: ^{222}Rn as well as ^{218}Po and ^{214}Po formed from ^{222}Rn escaping into the air adjacent to scale deposits.

Assessment of radiological doses for workers and environmental impact of TENORM

In the past, some NORM-contaminated scale and sludge were disposed of via land spreading and shallow or underground burial, or simply stored in solid waste landfills. Now, the petroleum industry is adopting methods for managing and disposing of NORM contaminated waste that are more restrictive to provide better isolation of the radioactivity.

In general, handling and storage of NORM contaminated wastes can lead to the exposure of workers, while burial and land spreading is connected with potential exposure of the members of public both from external radiation and radon inhalation.

If the concentration of ^{228}Ra(Th), ^{226}Ra and ^{40}K in the deposited scale is known, the calculating of the so-called reference dose in outdoor air at 1 m above the ground can be done from the equation adopted by the United Nations Scientific Committee on the Effects of Atomic Radiation [57].

Table 4. Exposure rate levels in the oil industry

Country	Reported range (μSv/h)
Algeria [22]	Bkgd–100
United Kingdom [22]	10–300
Egypt [12]	50–100
Congo, Italy, Tunisia [54]	0.1–6
USA [29]	up to 300

$$D_\gamma = (0.417\ C_{Ra} + 0.604\ C_{Th} + 0.0417\ C_k) \times 10^{-3}$$

where: C_{Ra}, C_{Th} and C_K are the activities (Bq/kg) of ^{226}Ra, ^{228}Ra and ^{40}K, respectively and D_γ is the dose rate (μGy/h) due to gamma radiation.

Taking into account the median radium nuclide activities in the scale equal to 100,000 Bq/kg and ^{40}K ~ 100 Bq/kg one can get D_γ = 100 μGy/h. This value far exceeds (by more than 1000 times) gamma dose of ~ 0.07 μGy/h from terrestrial gamma and cosmic rays.

Numerous studies have been devoted to appraise the real radiation doses and risk for workers in the oil industry. Some results of these investigations are shown in Table 4.

The real occupational doses depend on the dose rates and the working time spent during normal activities. The crucial problem in the occupational effective dose evaluation is to assess the so-called occupancy factor. Usually, for typical activities and repair work, this value ranges from 10 to 20 h/year. Calculated on these assumptions the annual effective doses for normal activities in the oil industry should be in the range of up to 2 mSv/year. Similar results (see Table 5) were published in the report concerning the dose assessment in the American oil industry [44].

Therefore, all of these calculated doses are not only below the 20 mSv/year limit on the effective dose for exposed workers but also below the limits for members of the public (1 mSv/year). It is worth underlining that because of the low radon emanation rates from solid scale, the annual occupational doses associated with radon daughter inhalation are in the range below 1 mSv.

During the EU Marina II project, extensive studies concerning the radiological impact of discharges on the European marine systems from NORM (including the oil and gas industry) to the European population have been done. The peak collective dose rate occurred in 1984 and was just over 600 manSv/year. At that time, the collected dose was almost entirely due to discharges from the phosphate industry. However, in 2000 the oil industry contributed about 39% (76 manSv/year) to the total (195 manSv/year) collective dose rate from NORM-industry discharges. The largest input to the estimated collective dose is due to ingestion of ^{210}Po in seafood [7]. Similar conclusions have been achieved after a probabilistic (fuzzy rule) modelling of possible human health effects after the discharge of produced water [41]. The risk in terms of predicted additional cancer incidence from radionuclides in produced water is within regulatory acceptable ranges.

Landspreading of low activity solid scale or sludge is practised in some countries. Potential doses associated with the disposal of petroleum industry NORM waste and further use of this area for industrial and residential purposes were evaluated using the RASRAD computer program [6, 43].

On the basis of these analyses (see Table 6), one can conclude that for residential houses, constructed on recultivated areas after scale disposal, the equivalent ^{226}Ra concentration in soil should be below 350 Bq/kg. However, the US EPA has issued another, non-mandatory guideline identifying radium concentration limits for disposal at landfills:

- 108÷1850 Bq/kg – for disposal in sanitary landfills, with limited access and no future development of the site.
- 1850÷74,000 Bq/kg – for disposal in TENORM or low-level radioactive waste facilities.
- Greater than 74,000 Bq/kg – for disposal according to Atomic Act regulations.

Ecological catastrophes occurred in the oil industry because the low content of the natural radionuclides in the crude oil did not influence environmental radioactivity levels, even in the local scale.

For example, Kuwait was heavily contaminated during the Gulf War due to large quantities of crude oil released in the vicinity of the oil fields and the atmospheric discharges from incomplete oil combustion

Table 5. Doses associated with equipment cleaning facilities [44]

Scenario	Pathway	Annual dose (mSv)
Pipe cleaner (wet process)	External gamma	0.04
Pipe cleaner (dry process)	External gamma	0.04
	Ingestion	0.35
	Inhalation	0.11
	Total	0.50
Vessel cleaner	External gamma	0.05
Storage yard worker	External gamma	0.40
Adjacent resident	External gamma	0.002

Table 6. Potential doses corresponding to various ^{226}Ra concentration after landspreading [6]

^{226}Ra concentration after landspreading (Bq/kg)	Receptor dose (mSv/year)		
	Worker	Residential	Industrial
185	0.007	0.3–0.6	0.15
370	0.014	0.6–1.2	0.30
555	0.022	0.9–1.8	0.45

from the burning of oil wells. Approximately 67 million tons of crude oil was burned over 250 days. According to our estimations, this resulted in the emission of approximately 6.7 GBq of ^{238}U, 10 GBq – ^{226}Ra and 5 GBq – ^{232}Th radionuclides [9]. Extensive environment radioactivity studies performed after that ecological disaster in this region have not showed any measurable increase in concentration of these radionuclides in the surface soil samples [8] nor the bottom sediment cores from Kuwait Bay [5].

Conclusions

Accumulated scale in tubes or stored in the open air, containing radium nuclides in concentration up to 3.5×10^6 Bq/kg, can pose an occupational hazard, mainly by external gamma radiation and escaping radon and its daughters' inhalation. In practice, because the times workers spend around these radiation sources are infrequent, it does not present a severe radiation protection problems. Reinjection or disposal of produced water into marine environment does not generally influence the levels of natural radioactivity in marine biota. However, disposal of solid radioactive waste or sludge from the oil industry to marine or to sanitary landfills should be carefully examined and is generally not recommended. Repositories situated within an underground rock formation seem to be costly but are the safest way for the final storage destination for the majority of solid scale waste with specific activities in the range of 10,000 to 100,000 Bq/kq. Such a repository for radioactive waste from the petroleum industry on the Norwegian continental shelf has been recently opened under authorisation of the Norwegian Radiation Protection Authority [35].

However, as it is evident from the papers presented at the latest IAEA Conference on NORM [28], there is an urgent need to establish clear rules and international regulations to deal with radioactive waste, not only from the oil industry.

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