A review about radioactivity in TENORMs of produced water waste from petroleum industry and its environmental and health effects

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Abstract. Produced water that detached from petroleum production process is often reimbursed into the surrounding environment. This might have negative impact for environment; it is usually infiltrate agricultural soil and contaminate groundwater with toxic elements. These waters are often contaminated by technologically enhanced naturally occurring radioactive materials (TENORMs). It may create specific environmental issues and health radiological threats to the inhabitants, especially inside the petroleum facilities or the local population that live in areas and use the groundwater for the usual living purposes. The aim of this work is to provide an intensive review for the previous studies on the (TENORMs) that associating with the produced water from petroleum industry, and its environmental and health impacts in many areas around the globe. It is true that there is long term hazard associated with widespread behaviour of releasing produced water into the environments for both workers and local inhabitants as well as the environment itself. However, there is many studies suggesting that currently the situation could be considered in low hazard level in most of petroleum fields, but with increases in the extraction and use of petroleum, the discharge of produced water into the environment increases accordingly, this might be end up with negative impacts on the environment and health such as radiological contamination of aquatic and agricultural life.

1. Introduction

Petroleum reservoirs are considered to be main sources for fusel fuel energy in those years. It is true that the natural petroleum reservoirs are contained in their formation water beside the oil and gas. The formation water is reported to be mixed since millions of years with petroleum in the geologic reservoirs consisting of sedimentary porous rocks formation between stratum of impervious rocks inside the earth’s crust [1-3]. Moreover, sometime water and chemical materials are injected into the old reservoirs to keep the pressure level reasonable and to enhance both the safety and recovery rates of processes. These injected water and chemical materials might be break throughout the production area and recovered from the different components of petroleum through production operations [1][4-6]. This water has been substantive as one of the biggest quantity tailings stream in the production and exploration processes of petroleum, the quantity of it, in some cases, override the quantity of petroleum produces [7-10], this water is produced with oil and gas during extraction operations as
shown in Figure 1 below. Furthermore, the produced water might represent around 80% of the residuals waste generated from petroleum industry processes and its size generally increases as petroleum products decreases with the age of the well progresses [10-12]. Produced water includes melted hydrocarbons like monocyclic dispersed oil and aromatics, melted types such as Na and Cl with high concentration and a vast group of mineral combinations, as well as, contains naturally occurring radionuclides alongside with artificial radionuclides which enhanced as a result of technological and human activities. These radionuclides called technologically enhanced naturally occurring radioactive materials (TENORM), may be caused by uranium and thorium chains and may be separate elements [3] [13-14]. The water that used in the production process of oil from the field is actually considered to be consist a certain amounts of radioactive materials that created by the oil industries, such as 232Th, 238U, 226Ra, 228Ra, 210Pb, 224Ra, and etc., [3][8][9][14]. It is shepherded by discharges of TENORMs raising worries over the possible radioactivity effects of produced water on environment components including living organisms. The issue of the leakages of TENORMs from those water may lead to notable effects on environment components, therefore it has received valuable effort and great attention from both scientist and technical staff in the recent years in different areas around the globe [9][11][12][15]. Many researchers have been reported the mostly appearing radioisotopes in produced water and its values in several regions of the world. The continuous expansions of the activities of the petroleum industries and prediction of potential expansions have increased the needs for a responses and continuously examining the levels of radioisotopes. In this work we attempt to present an extensive overview of previously published studies on the effects of TENORMs from produced water by petroleum industries on the workers, public and the environment.

2. Produced water wastes from petroleum industry
The natural petroleum reservoirs have oil and gas alongside with water in the pores of the earth rocks, where water quantities tend to vary according to the geological structure of the area were the reservoirs. The extraction of petroleum is usually accompanied with amounts of water that either injected or it exist already in holes with petroleum. In a general these water is called produced water, composition water, or saltwater in oil fields [15-17]. 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 dissolved and
dispersed hydrocarbons) and inorganic compounds (e.g., suspended particles, trace minerals, and dissolved salts). These organic compounds are composed in several groups, such as polycyclic aromatic hydrocarbons and alkylate phenols which produce different effects in various organisms, so that it can ease the transportation of radioactive nuclides [15][18-23]. 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 [15][24-27]. 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 [23][28-29]. Thus, the main source of TENORMs in the petroleum industries is the dissolved radionuclide in fluxes of produced water that is then transported to the surface with petroleum. [15][21][30][31-36].

| Country | Concentrations of the TENORMs in produced water with range, averages and units | Ref |
|---------|--------------------------------------------------------------------------------|-----|
| Algeria | 226Ra (5.1–14.8) Bq·g⁻¹                                               | [30]|
| Brazil  | 137Ba (0.36–25.7) 8.80a mg·L⁻¹, 226Ra (0.012–6.0)1.95a, 228Ra (<0.05–12.0) 2.91a Bq·L⁻¹ | [31]|
| Congo   | 238U < 4.5x 10-3, 232Th < 4.5 x 10-3, 226Ra 5.1a, Bq·dm⁻3                  | [32]|
| Italy   | 238U (7.3 x 10³ – 1.5 x 10²), 232Th< 4.5 x 10-3, 226Ra (0.06 – 20) Bq·dm⁻3 | [32]|
| Norway  | 228Ra 3.3a, 228Ra 2.8a Bq·L⁻¹, 210Pb < 10a, 210Po < DLa mbq·L⁻¹              | [33]|
| Norway  | 226Ra (0.5–16), 228Ra (0.5–21), 210Pb DLa Bq·L⁻¹                         | [34]|
| Syria   | 226 Ra (13.8–111.2) 51.9a, 228Ra (12.4–67.4) 37.5a, 224 Ra (0.2–3.7) 1.1a Bq·L⁻¹ | [35-36]|
| Syria   | 226Ra 186.2a, 232Th 19.2a, 40K 1460.8a Bq·kg⁻¹                          | [37]|
| Egypt   | 226Ra (1.0 – 34.1 ) 16a, 228Ra (< 0.02 – 13.26) 43.5a, 40K (3.6 – 15.37) 7.37a Bq·kg⁻¹ | [38]|
| Egypt   | 226Ra (5 – 40) 15.92a, 214Po 0.8 – 27) 15.93a, 214Bi (1.3 – 27) 15.43a, 226Ra (1.1 – 59) 38.37a | [39]|
| Egypt   | 238U (9.47 – 25.2), 232Th(7.33 – 22.6), 40K (632.5 – 1448.7) Bq·L⁻¹     | [40]|
| Tunicina| 226 Ra 19a, 232Th 39.9a, 40K 66a Bq·kg⁻¹                                  | [41]|
| Iraq    | 238U 4.12a, 232Th D.La, 40K 14.16a, 137Cs 11.16a Bq·L⁻¹                  | [42]|
| Iraq    | 226Ra 20.3a, 232Th 9.4a, 40K 66.4a Bq·kg⁻¹                               | [43]|
| Iran    | 142.97a, 214Bi (<DL – 471) 178.78a, 40K (4.4 – 43.7 43.7) 24.29a, 238U (30 – 211) 126.88a, 208Ti (4 – 44.3) 21.36a, Bq·L⁻¹ | [44]|
| Poland  | 40K (5 - 499) 75a, 238U <30 a, 226Ra <2a, 210Pb <5a, 228Ra <2a, 228Ra <2a, 232Th <2a, Bq·L⁻¹ | [45]|
| Turkey  | 224Ra (<1 – 4) 2.83a, 226Ra (< 3 – 10) 6a, 228Ra (<1 – 4) 3.17a Bq·L⁻¹ | [46]|
| Romania | 238U (0.043 – 1.1), 226Ra(23 – 45), 232Th (0.2 – 8), 40K (221 – 899) Bq·L⁻¹ | [47]|
| Omán    | 228Ac (1019 – 1040) 1030a, 226Ra(514 – 529), 40K(1522– 1535) 1528a, Bq·L⁻¹ | [48]|
| Ghana   | 238U (0.11 – 1.03) 0.54a, 232Th (0.21 – 0.56) 0.41a, 40K (1.65 – 11.99) 7.76a Bq·kg⁻¹ | [49]|
| Ghana   | 226Ra (6.20–22,30), 228Ra (6.40 – 35.50), 228Th (0.71 – 6.41), 224Ra(0.78 – 7), 40K(5.90 – 23.90) | [50]|
| Nigeria | 226Ra 8.9a, 228Ra 8.1a, 40K 39.8a Bq·L⁻¹                                 | [51]|
| Nigeria | 228 Ra (0.75 – 12.30) 5.18a, 226Ra(2.01 – 13.19) 6.04a, 40K(9.08 – 155.22) 48.78a, Bq·L⁻¹ | [52]|
| Argentina| 238U (< 10.0 – 33.0) µg·L⁻¹,  226Ra (<1.7 E–3 – 26.8), 228Ra (< 1.1 E–3 – 9.6) Bq·L⁻¹ | [53]|
| US      | 226Ra (56 - 1494) 367.5a, 228Ra (69 - 600)275.75a, 228Th (11.4 – 120.7)62.76a, 210 Po (0.6 – 0.6 – 2.3)1.39a, 210Pb (2.6 – 16.7)9.66a pCi·L⁻¹ | [54]|
| US      | 226Ra (30 – 2690), 228Ra (35 – 763) Bq·L⁻¹                              | [55]|
| US      | 226Ra (<0.002–58) 11.7a, 228Ra (0.02–59) 15.5a Bq·L⁻¹                  | [56]|
| Azerbaijani | 226Ra (ND – 101.07), 232Th (ND – 13.71), 40K- (26.1 - 194.5) Bq·L⁻¹ | [57]|

( ) The data between the brackets means the range, ° is the averages of concentration, LD = lower than the detection limits.
3. Concentrations of TENORMs in produced water

The radioactivity levels in the produced water measured in what known by radioactivity concentration, which is the amounts of specific radionuclide in given volume of water. The reported values for all types of radionuclides are varied from region to another, this clearly obvious because of the geographical structure. A summary of several previous studies that conducted a measurement of TENORMs generated from produced water of petroleum industry is given Table 1. From Table 1, we can notice that the radioisotopes and their radioactivity concentrations have wide range of spectrum, from natural to artificial and therefore their environmental and health effects also vary from region to region.

From the table above, we can clearly identify the results of previous studies [26][42-50][52][54-59][60-67] have shown that radium isotopes are mostly present among all others radioactive elements in produced water, especially $^{226}$Ra, $^{228}$Ra, and $^{224}$Ra. $^{226}$Ra which are produced from disintegrates of $^{238}$U into $^{222}$Rn by alpha and beta particles decay beside the gamma rays to reach aground state over 1,600 years of half-life. $^{228}$Ra and $^{224}$Ra originate from $^{223}$Th which decay to $^{228}$Ac and $^{220}$Rn respectively. $^{228}$Ra is emitting beta particles and gamma ray in attempt to reach aground state from excited state over around 5.75 years of half-life. $^{224}$Ra emits alpha particles and gamma rays with estimated half-life of 3.7 days. From analysis of the previous studies’ results can show that about 36% of radium isotopes in produced water are less than 5 Bq.L$^{-1}$, while approximately 49% of its where ranged between 5 Bq.L$^{-1}$ and 60 Bq.L$^{-1}$, and about 15% were more than 60 Bq.L$^{-1}$. The human body process of radium are similar in it a manner to the process calcium, therefore any inhalation or ingestion of small amounts of radium lead to accumulate them with time in the body and thereby causing dangerous harms in the long-term future. The ultimate result of continuous internal exposure to radium always considered resulting to a bone and sinus cancer [44][48-49][58-59]. Moreover, it is also potentially causing widely spread diseases such as Nasal mucosa tumors and bone tumors. $^{226}$Ra bioaccumulation in the food series is a possible risk for live organism, thereof the ecosystem is needed to be protected from this threat. That is, many studies alarm the need for a primary concern for protection of the ecosystems [52][54-55]. Therefore, appropriate preventative policy must be put in plan to protect the environment against such radiations.

The potassium isotope, $^{40}$K is naturally occurs radioactive element. The pollution from $^{40}$K could be originated by the persistent use of the earth’s crust components containing radionuclides. It is estimated to be 0.012 percent of natural potassium and has concentrations of about 0.5 Bq.g$^{-1}$ or 1.8 mg.kg$^{-1}$ in the earth's crust [51-53][55-56][58-60]. $^{40}$K undergoes a manifold disintegration into $^{40}$Ca by releasing a beta particle or upgraded to $^{40}$Ar by capturing electron and emitting gamma ray, with a half-life of 1.3 billion years. However, there is no chemical or physical evidences about it acts in the environment manner except like all the other potassium isotopes. It has also being assimilated during normal biological operations into the all animal and plant tissues [55-56][58-60]. Therefore, from released quantities of produced water the biosphere may be affected by abnormal levels of $^{40}$K this because, it is also among the mostly appearing radionuclide in produced water [48-53][55-56][58-65]. We can see from the analysis of previous studies’ results around 34% of the of $^{40}$K isotope activity concentrations in produced water were found to be lower than 20 Bq.L$^{-1}$, and about 25% of the reported values ranged between 20 Bq.L$^{-1}$ and 60 Bq.L$^{-1}$. Approximately around 41 % or slightly lower than this was over 60 Bq.L$^{-1}$. All these reported values are above the recommended value for drinking water which is no more than 0.2 Bq.L$^{-1}$ according to UNSCER and the IAEA recommendations. Furthermore, when produced water is consist a high level of radioactivity concentration of any radioisotope this might be serve as an evidence of the potential level of radioactive contamination for agricultural soil and groundwater.

From the reported results of previous studied [26][43-46][50][55][66]. We can see that there is certain amounts of lead isotopes in produced water, especially $^{214}$Pb and $^{210}$Pb isotopes these are originated from $^{238}$U by alpha decay into $^{214}$Bi and $^{210}$Bi respectively. $^{214}$Pb emits beta particles and gamma rays with about 27 minutes of half-life. $^{210}$Pb also emits beta particles and gamma rays in around 22.3 years of half-life. The analysis showed that the activity concentrations of all lead isotopes
in the produced water are less than 20 Bq.L\(^{-1}\) [26][45][50][55-56][66]. However, during process of petroleum industry lead might be moved to the earth surface, and then stick to the soil. From the surface soil could easily move to groundwater. All these process above might be affected by quality of the chemical composition that formed by lead and the soil characteristics.

The results of these previous studies [44][51][53][55-56][58][61][65] found that uranium isotopes were, appeared in produced water, especially \(^{238}\)U and \(^{235}\)U isotopes. \(^{238}\)U disintegrate into \(^{234}\)Th by emitting alpha particle and gamma rays with about 4.5 billion years of half-life. \(^{235}\)U disintegrate into \(^{231}\)Th by emitting alpha particle and gamma rays to achieve aground state for certain isotope within 70 million years of half-life. While another studies, [45][48][51-54][56-51][59-62][66], have shown that thorium isotopes were detected in produced water, especially \(^{232}\)Th and \(^{226}\)Th isotopes. \(^{232}\)Th disintegrate into \(^{228}\)Ra by emitting alpha particle and gamma rays with a half-life around 14 billion years. Furthermore, \(^{226}\)Th isotope also detected, which is a daughter of \(^{232}\)Th that disintegrate into \(^{224}\)Ra by emitting alpha particle and gamma rays in a period of time around 1.9 years known as the half-life. An analysis of the results of previous studies showed that most of the activity concentrations of uranium and thorium isotopes in the produced water are less than 20 Bq.L\(^{-1}\). Water pollution introduced by depleted uranium or thorium will certainly causes many diseases to the consumer of product that used in it process in some way, these are either an immediate or nearby future consequences [48][52-54][56][60]. Moreover, if the soil is contaminated with depleted uranium or thorium, this has very harming consequences that could affect everything including human beings, animals, plants, and insects because its suspend on the air and penetrated into the water and soil, and here lies the danger [44][51][57][59-61][62].

The studies such as [50][55][59] also showed the appearances of the \(^{228}\)Ac isotope which is the daughter of \(^{228}\)Ra. Actinium is secularly balanced with its parent, and its existence in the environment at high levels poses a considerable long-term health risk due to its well-known radiological toxicity [50][55][59]. The artificial radionuclide such cesium \(^{137}\)Cs are also detected produced water of the Iraqi oil, this might be due to the first and second Gulf War [42]. The results of previous studies also showed that other radioisotopes such a \(^{137}\)Ba [31], \(^{210}\)Po [26][66], \(^{212}\)Bi [39], \(^{214}\)Bi [50][55] and \(^{208}\)Ti [50][55] are tend to be in detectable levels in produced water, but the low activity concentrations compared to the above mentioned radioactive elements.

If the above mentioned radioisotopes are detected this might be used as indicator of the existence of \(^{228}\)Ra, and therefore, this study might provide a beneficial review to sustainable development of what ongoing by providing an overview of polluted produced water around the world which might indirectly contaminate the soil, water etc.

4. Risks of exposure to TENORMs

4.1. Risks of TENORMs of produced water on the environment

The concept of environment protection from low level radiation is highly applicable to the TENORMs more than nuclear material industry because the TENORMs levels in the petroleum industries are low compared with those in the nuclear industry. Radiation protection policy for animals and plants are less restricted even though there are certain amounts of doses are recommended, while human being is highly restricted and protected by certain radiological standards. For this reason, the International Commission on Radiological Protection (ICRP, 1991) recommended the adoption of human protection standards as criteria for the protection of living organisms. This condition indicates that if the overall exposure to radiation from TENORMs of produced water, or the disposal of this TENORMs wastes is less than the general public limit of 1 mSv/y, then the radioactive doses received by organisms are considered to be acceptable [55][58-61]. Using contaminated TENORMs or radioactive waste materials and their improper handling, storing, and transferring without effective control can contaminate multiple-land areas in the surrounding environment which can lead to the potential exposure of the public [58][62-63]. The environmental effects of the TENORMs resulting from produced water in the petroleum industries can be summarized in the land and marine environment as
follows: (1) discharge of produced water, into the facilities of the petroleum industries may lead to the transfer of TENORMs to the surrounding environment, accordingly the content of radionuclides in rock and soil deposits into river water, and other components of the surrounding environment which may be increases a cause of potential contamination. These radionuclides may be transported by water and soil to animals and plants. However, the produced water of petroleum industry contains low radioactivity of TENORMs and therefore low dose generation in environment. For this reason, radiological exposures and radioactivity the force a negative effect on the environment are unlikely to be insignificant due to discharges of small volumes of inorganic materials, but these wastes accumulate over time and thus attitude radiological hazards [40][55][59][64-65].

4.2. Risks of TENORMs on human health
Produced water may contains TENORMs, which could release all kinds of radiation (alpha particles beta particles, and gamma ray). TENORMs is therefore might be consider potential source of radiation exposure and inhalation, where a person can be exposed into it from the well two known ways of exposure:

4.2.1. External exposure (irradiation). The External exposure or irradiation the source remains outside the body. When workers perform routine operations, they might be exposed to gamma radiation or beta and alpha from short distances. The gamma ray radiation could tunnel throughout steel walls of the pipes or concrete wall that containing the produced water or any other gamma sources. The dose rate can be varying from fraction of micro-Si evert per hour for weak sources to several micro-Sievert on the surface of highly contaminated pipelines and utensils. The workers may have been exposed to inhaling radon, TENORMs dust, and gamma radiation during shutdown and maintenance periods of petroleum plan. In many studies such as [41][42][49][60][66-67] conducted in oil and gas fields found that the rate of external radiation exposure dose depend on the area where the measurement have been conducting. For example, in an area closed to the machinery and equipment the radiation dose found to be higher than that in the outer walls, this because steel walls are unprotected from the external gamma radiation while the outer wall is highly fabricated with materials that could attenuate and decreases the intensity of photons [50][62].

4.2.2. Internal exposure (contamination). The internal exposure is considered to be most dangerous, this because of the radioactive material transported into the human body either by absorption, ingestion. The radium equivalent activity concentration in the sulfate scale or sludge may be greater than 500 Bq.g−1. That is, only 100 mg with such concentration of inhaled dust is sufficiently enough for irradiation dose exceed the annual general public dose limit of 1 m Sv.y-1. Although the amount of radon released is low, concentrations of radon in non-aerated vats of produced water can cause

| Country  | Ra eq (Bq.kg⁻¹) | D pr (nGy.h⁻¹) | D eff (mSv.y⁻¹) | Ref |
|----------|----------------|----------------|----------------|-----|
| Saudi Arabia | 1.35 - 173 | <1D – 83.6 | | [68] |
| Egypt | (70.4 -164.9)×10³ | 31 - 72.7×10³ | | [67] |
| Egypt | (544 - 596)×10³ | (250 – 273) 262a | (306 – 335) 321a | [66] |
| Tunisia | 0.09 - 398 | 0.043 - 177 | 5.28×10⁻³ – 3.64×10⁻³ | [41] |
| Iraq | (5.21 -79.84)×10⁻¹ | (2.49 -37.53) 22.18a | (0.012 - 0.184) 0.109a | [42] |
| Algeria | 0.1(100)×10³ | (0.01 – 0.60) | | [30] |
| Italy | (0.1–6)×10³ | | | [32] |
| Turkey | (0.2–25.7)×10³ | | | [46] |
| GHANA | (10.0 – 57.0) | (12.6 – 83.4) | | [49] |
| Iraq | (41.75 - 117.61) | (21.08 - 57.10) | (0.025 - 0.280) | [60] |
| Iraq | (0.08 – 11.2) | | | [42] |

( ) The data between the brackets means the range / a is the averages of radiation indexes
increased exposure to radiation [40][43][61][62-64]. Several studies have attempted to measure the radioactivity doses and the true dangers in the produced water of petroleum industry, some of it results are selected and presented in Table 2.

These results are clearly indicate as an evidence for how match the produced water is contaminated with TENORMs, which produces harmful radiation doses to human being and the environment at different levels. The higher the level of radioactivity in waste (produced water) the greater the radiation effects occur, especially when one considers the possibility of exposing an operator throughout what is known by internal pollution. This tend to occur when dust absorbed during produced water treatment, and closeness to the radiation source shows a significant health effected of the radiation [58][62]. Since the radiation doses measured in accumulation manner, this un-accounted additional radiation might be exceeding the limits allowed to the public, therefore leading to health risks in these communities.

5. Conclusions
Petroleum industry are often linked to various types of environmental problems such as petroleum spillage and improper discharge of produced waste, along with pollution and environmental degeneration which become major issue facing human being with the increasing relaying on petroleum industry and its activities for energy purposes. This might become with non-compliance to environmental safety standards. These wastes contain TENORMs which are led to increase in the level of radioactivity on the earth surface and thereby such radiological wastes create environmental and human health problems. The release of some radiation materials like $^{226}$Ra, $^{228}$Ra, $^{222}$Rn, $^{210}$Po, $^{210}$Pb and other radionuclides are dangerous to the environment when it transferred to the drinking water and agricultural soil, which may be consumed by animals and plants through the food series, and therefore consumed by people causing neurological disorders, cancers and other complicated health issues. The results of previous studies clearly shown that the levels of radioactivity in produced water in some areas exceeded the limits set by the US EPA. It can be concluded that in order to avoid long-term radiological hazards that may affect the health of communities’ pre-medial steps must be taken to properly manage and dispose of the produced water. Although all radioactive material from waste to produced water, and its impact can be reduced by complying with the standards that recommended by the IAEA and other environmental agencies.

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