

REVIEW OF NORM DISTRIBUTION AND BEHAVIORS IN PRODUCED WATER FROM OILFIELD INDUSTRIES

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Abstract

According to the noteworthy concentration of naturally occurring radioactive material (NORM), produced water that is considered a damaging waste resulting from the oil extraction process according to the noteworthy concentration of naturally occurring radioactive material (NORM), is regularly disposed into nearby areas, resulting in environmental contamination that raises the possibility of human exposure NORM in various pathways. This paper purposes to provide an extensive examination of the NORM reported in produced water linked with oilfield activities from previous reports. The comparison of NORM values in oilfields around the world demonstrates a vivid understanding of radionuclide behaviors associated with oil extraction operations. The results illustrate that Radium isotopes are the most radionuclides abundant in produced water, especially Radium-226, activity concentrations of U-238 and Th-232 were remarkable to be low in the water. Oil extraction is increasing, leading to more produced water being disposed of, which is a real concern for human health, so a depth study is recommended to focus on minimization and other management methods of produced water instead of being disposed of. Furthermore, the NORM waste and its influence could be minimized by obeying the recommended standard set by IAEA and other environmental protection agencies.

Keywords: Environment contamination, NORM, Human exposure, Produced water, Radiation risks.

Introduction

The major challenge in the recent oil industry is the unwanted production of water and gas; “every day, approximately 300 million barrels of water are brought up to the surface together with oil and gas” (Liang et al., 2018). produced water represents an enormous waste stream because of several oilfield company operations. During the oil extraction process, a huge amount of water comes out of the wells to the surface with the crude oil, including both formation water and injected water into the wells to enhance the oil and gas recovery (Altare et al., 2006). The first source of produced water follows the oil and gas extraction process, while formation water that exists below the oil layer then enters through the porous reservoir and comes out of the well mixing with the crude oil. This process destabilizes the pressure of the reservoir and resolves this problem by injecting water again into the system to sustain the hydraulic pressure. (A Elbrir et al., 2018).

This injected water represents the second source of produced water based on the fact that the more oil extraction there is, the more produced water. Furthermore, “the origin of that unwanted water involves saline water that exists and resides in the layer below oil and gas due to its high density compared to those hydrocarbons. Generally, there are two sources of saline water: flow from the same hydrocarbon zone due to hydrocarbon production and flow from other hydrocarbon zones due to hydrocarbon migration” (Altare et al., 2006). This Saline water is called formation water and becomes produced water when it is brought up in addition to oil to the surface as a mixture. In some other cases and due to the reduction of oil extraction, this water will be injected again to keep the hydraulic pressure and enhance the oil recovery. The injected water is usually from injector wells towards the formation, which directs oil to another well-called producer well, while the formation water or the injected water arrives in the producer wells. These wells start extracting hydrocarbons as well as producing water. This mixture contains, in addition to the water and oil, metals that have been reported in various studies, including Cr, Ba, Ni, Zn, Mg, Fe, Ni, Pb, and K (Johnson et al., 2008), furthermore “lead, nickel, zinc, cadmium, and copper usually exist as heavy metals in oil and gas field-produced water” (Liang et al., 2018). Heavy metals are transformed from a dissolved state to particles in water under oxygenated conditions (Azetsu-Scott, Yeats, Wohlgeschaffen, Dalziel, Niven, & Lee, 2007), along with radium and radon, treating chemicals, salt, and dissolved oxygen. The outflow of produced water is considered the main waste with regard to size subsequent to the oil and gas facilities. (Al-Masri, 2006; Eriksen et al., 2006; Committee on Evaluation of EPA Guidelines for Exposure to Naturally Occurring Radioactive Materials et al., 1999.; M. S. Hamlat et al., 2001; Jerez Vegueria et al., 2002; Othman & Al-Masri, 2008; Testa et al., 1994)

Produced water components

When the well has been drilled into the oil zone of the reservoir, where there is an aquifer underneath, and normally a layer of gas setting on the top, to get the stream from the reservoir and into the wellbore based on the pressure drawdown, taking into account the form of the reservoir either sandstone or carbonate, the oil that enters the well was certainly mixed with a massive amount of water in addition to the gas, this water that comes out with oil due to low velocity which leads to high flowrate in compare with oil that has a high velocity, this fact according to the Darcy Low, this water called the produced water, which then separated from oil using special chemical and physical treatment technologies (Al-Ghouti et al., 2019; Igunnu & Chen, 2014; Sathe & Munavalli, 2019). This water is generally comprised of organic and inorganic compounds such as chloride and sodium were the most abundant in produced water; however, sulfate was the lowest inorganic ions, additionally produced water comprises of metals including Chromium, Barium, Nickel, Zink in addition to heavy metals, Total dissolved solids (TDS), Total suspended solids (TSS), chemical additives to the wells while drilling operation to treat or prevent possible operation problems while wells oil drilling and enhance the separation of oil and water after extracting oil, in addition to Dissolved gas counting Oxygen, Hydrogen sulfide, and carbon dioxide. (John, n.d.). therefore, its disposal process is one of the global concerns, consequently leading to human exposure in different pathways and environmental contamination. **Table 1** displays information about the Concentrations of metals in produced water and other parameters.

Table 1: « Concentrations of metals, hardness, salinity, TDS, and density of produced water ».
 (Dórea et al., 2007; Xiao et al., 2021)

Parameters	Range ($mg.L^{-1}$)
Na	8800-189000
Cl	16000-19500
Co	0.003-0.004
Pb	0.003-0.003
Cu	0.001-16.9
Fe	4310-4770
Mn	0.058-17.2
Ni	0.015-0.017
Zn	0.027-10.1
K	3100-9530
Mg	1530-3790
Al	<PL-12.5
Sr	709-2450
TDS	237.680
The hardness of water, $CaCO_3$	4890-44778
Density. 20°	1010-1070

NORM concentration in produced water

“Naturally, radiation exposure exists due to cosmic rays or naturally occurring radioactive materials (NORMs) that originate in the Earth's crust and are present everywhere in the environment” (Nations Scientific Committee on the Effects of Atomic Radiation, 2000). Produced water has been reported to contain significant values of NORM. (Al-Masri, 2006b; Bakr, 2010; Khodashenas et al., 2012; Shams et al., 2017). Practically all elements are constituents from stable nuclides; however, U and Th are unstable by nature and will fade in time by disintegrations into other radioactive elements by emitting alpha and beta particles accompanied by gamma rays. To become stable lead-207, a uranium-235 nucleus goes through 11 changes. While To reach the stable lead-208, a thorium-232 nucleus goes through ten transformations, counting ^{228}Ra and ^{224}Ra .

A uranium-238 nucleus decays through 14 transformations, including ^{226}Ra and ^{222}Rn to become stable lead-206, in addition to ^{40}K , one of the three isotopes of K, which is widely distributed identically with its isotopes, implying that the presence of K will be accompanied by ^{40}K which disintegrates once into either Ca^{40} or Ar^{40} emitting β -particles (89%) or γ -photons (11%), respectively.

The radioactivity concentration in a given volume of water, “represents the levels of radioactivity in produced water” (Al-Masri, 2006); the distribution of the reported levels in different areas are varied from one region to another due to the geological characteristics in each region. **Table 2** resumes the values of radionuclides associated with produced water that has been reported in several regions in the world.

According to the results shown in table 2, we notice that Ra isotopes are the dominant radionuclides in produced water, specifically ^{226}Ra , ^{228}Ra , and ^{224}Ra . ^{226}Ra which results from ^{238}U , decays into ^{222}Rn by emitting alpha and beta particles, in addition to gamma radiation, to end up in a stable state concluded 1600 years of half-life, on the other hand, “ ^{228}Ra and ^{224}Ra are daughters products of the ^{232}Th decays chain, which decays into ^{228}Ac , ^{220}Rn respectively; ^{228}Ra reach the ground state by emitting beta particles and gamma rays with an estimated half-life of 5.75 years, while ^{224}Ra end up in the stable state through decays by emitting alpha particles and gamma rays over 3.7 days of half-life” (Ali et al., 2020). The ^{224}Ra is noticed as the lowest radium isotopes present in produced water because it reveals in produced water without its immediate parent ^{228}Th , so that will die out within two weeks of secular equilibrium, the same period for ^{226}Ra to reach its secular equilibrium with ^{222}Rn , ^{218}Po , ^{214}Po , ^{214}Bi , and ^{210}Pb , while ^{228}Ra is considered as the quick radium isotopes that reach its equilibrium with ^{228}Ac withing two days.

Table 2: Activity concentrations (Bq.L^{-1}) of ^{238}U , ^{226}Ra , ^{232}Th , ^{40}K , and ^{228}Ra and ^{224}Ra in produced water in different oilfields worldwide

Radionuclides	^{238}U	^{232}Th	^{40}K	^{226}Ra	^{228}Ra	^{224}Ra	Ref
Congo(Bq.dm^{-3})	$<4.5 \times 10^{-3}c$	$<4.5 \times 10^{-3}c$	-	5.1c	-	-	(Testa et al., 1994)
Egypt	-	39.9c	66c	19c	-	-	(Zakaria KM et al., 2018)
Iraq	-	9.4c	66.4c	20.3c	-	-	(Ali et al., 2017)
Romania	(0.043-1.1)	(0.21-8)	(221-899)	(23-45)	-	-	(Botezatu et al., 2004)
Syria	-	19.2c	1460c	186.2c	-	-	(Ghafar et al., 2017)
Ghana	(0.11-1.03)	(0.21-0.56)	(1.65-11.99)	-	-	-	(Faanu et al., 2010)
Ghana	-	-	(5.90-23.90)	(6.20-22.30)	(6.40-35.50)	(0.78-7)	(Kpeglo, 2015)
Nigeria	-	-	39.8c	8.9c	8.1c	-	(Ezekiel et al., 2012)
Nigeria	-	-	(9.08-155.22)	(2.01-13.19)	(0.75-12.30)	-	(Ezekiel et al., 2013)
US(pCi.L^{-1})	-	-	-	(56-1494)	(69-600)	-	(Lagera et al., 1999)
US	-	-	-	(30-2690)	(35-763)	-	(Mcdevitt et al., 2019)
US	-	-	-	(<0.002-58)	(0.02-59)	-	(Kraemer et al., 1984)
Azerbaijan	-	(ND-13.71)	(26.1-194.5)	(ND-101.7)	-	-	(Khalilovai et al., 2016)
Poland	<30	-	75c	<2	<2	-	(Jodłowski et al., 2017)
Texas	-	-	-	(0.1 – 5,150)	ND	-	(Spoonamore, 2011)
Brazil	-	-	-	(0.012-6)	(<0.05-12)	-	(Jerez Vegueria et al., 2002)
Norway	-	-	-	3.3c	2.8c	-	(Eriksen et al., 2005)
Norway	-	-	-	(0.5-16)	(0.5-21)	-	(Eriksen et al., 2006)
Syria	-	-	-	51.9c	37.5c	(0.2-3.7)	(Othman & Al-Masri, 2008)
Oman	-	-	(1522-1535)	(514-529)	-	-	(Pillay et al., 2010)
Turkey	-	-	-	6c	3.17c	2.83c	(Parmaksız et al., 2015.)

(): the range of the concentration, c: the average concentration, ND: below the detectible limits

^{226}Ra noticed as the most Ra isotopes present in produced water in different studies from different areas, (K. Ali, Shafik, et al., 2017a; Botezatu et al., 2004; Ghafar et al., 2017; Spoonamore, 2011; Zakaria KM et al., 2018.; Testa et al., 1994), one of the reasons could be its high solubility in water and its behavior preferring the aqueous state. Furthermore, ^{226}Ra is chemically similar to barium Ba, strontium Sr, calcium Ca, and magnesium Mg, High precipitation of ^{226}Ra has been reported with strontium and barium, which are taken part in the metals present in produced water. This result is following various previous experiments that aim to find the correlation between radium isotopes and metals. (Azetsu-Scott, Yeats, Wohlgeschaffen, Dalziel, Niven, Lee, et al., 2007; Bzowski et al., 2015; Zhang et al., 2014)

^{238}U and ^{232}Th concentrations are noticed in Table 1 and were measured according to their progenies ^{226}Ra and ^{210}Pb , respectively (Azetsu-Scott, Yeats, Wohlgeschaffen, Dalziel, Niven, Lee, et al., 2007; Kraemer et al., 1984; Parmaksız et al., 2015), The absence of U^{238} and Th^{232} in produced water in some studies, (Al-Masri, 2006b; Ezekiel et al., 2013; Kpeglo, 2015; Kraemer et al., 1984.; Lagera et al., 1999; Mcdevitt et al., 2019), according to their chemical characteristics, “makes them prefer the solid rock phase and do not dissolve in the aqueous or oily phase. As a result, both series remain with reservoir rock and may appear as a natural concentration just during drilling activities” (Managing Naturally Occurring Radioactive Material (NORM) in the Oil and Gas Industry | IOGP Publications Library, n.d.)

The results display a high value of ^{40}K activity concentration, ranging from 1.65-1460 Bq.L^{-1} in produced water because K isotopes are widely distributed in nature (abundance in the Earth’s crust is 2.1%), including ^{40}K (0.0117%). However, the ^{40}K concentrations in produced water are lower than the values found in soil samples around the oilfield area. (Agbalagba et al., 2012).

Figures 1-4 display the activity concentration of ^{226}Ra , ^{232}Th , ^{238}U and ^{40}K respectively, histograms confirmed the abundance of ^{226}Ra in produced water, Moreover, the graphs show an abnormal distribution (not bell-shaped) of these radionuclides, which reaffirms the effect of the geological characteristics on the distribution of NORM.

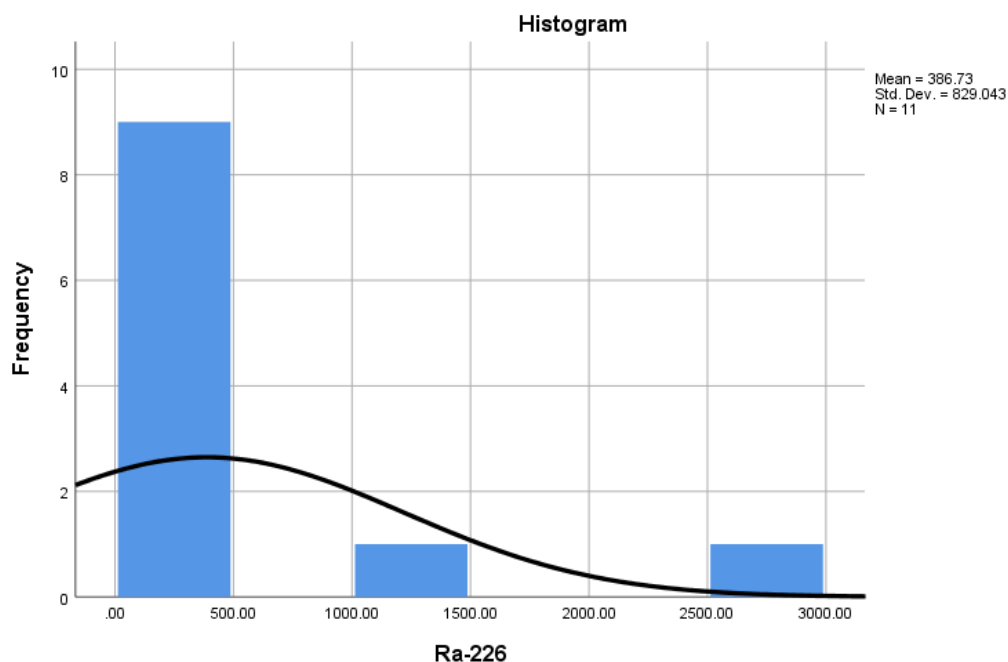


Fig.1: The distribution frequency of concentration of ^{226}Ra

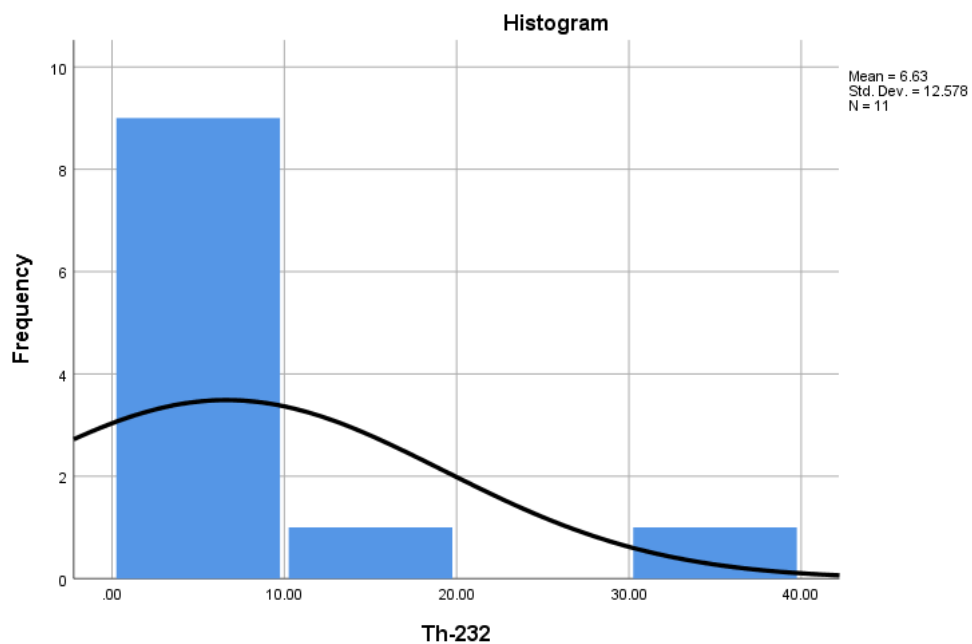


Fig.2: The distribution frequency of concentration of ^{232}Th

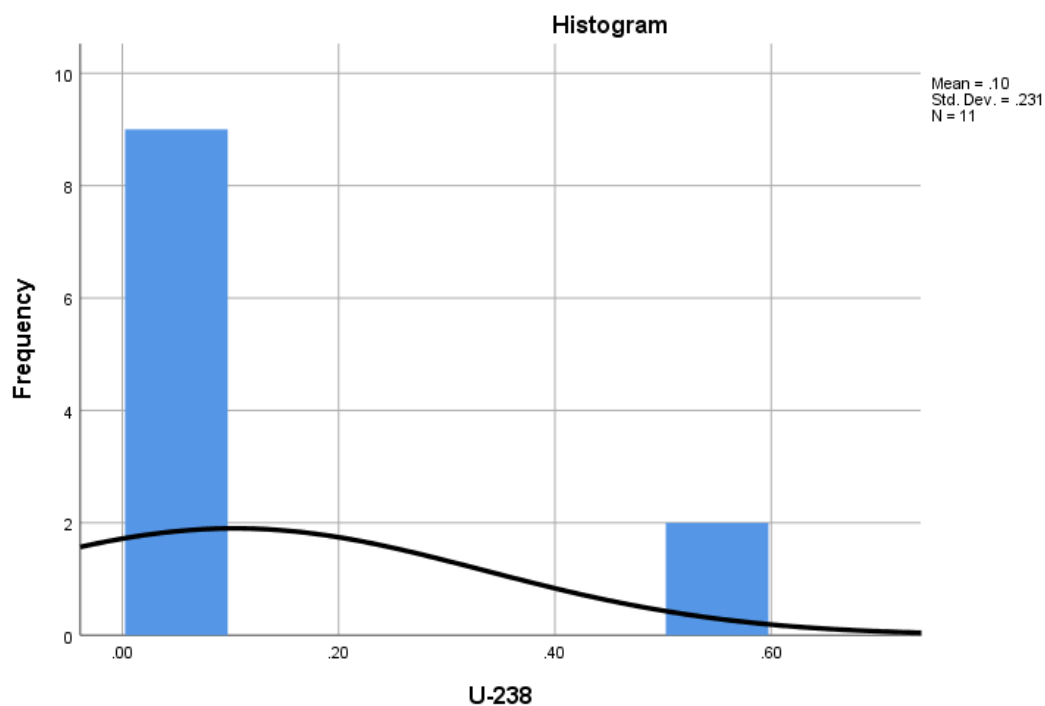


Fig.3: The distribution frequency of concentration of ^{238}U

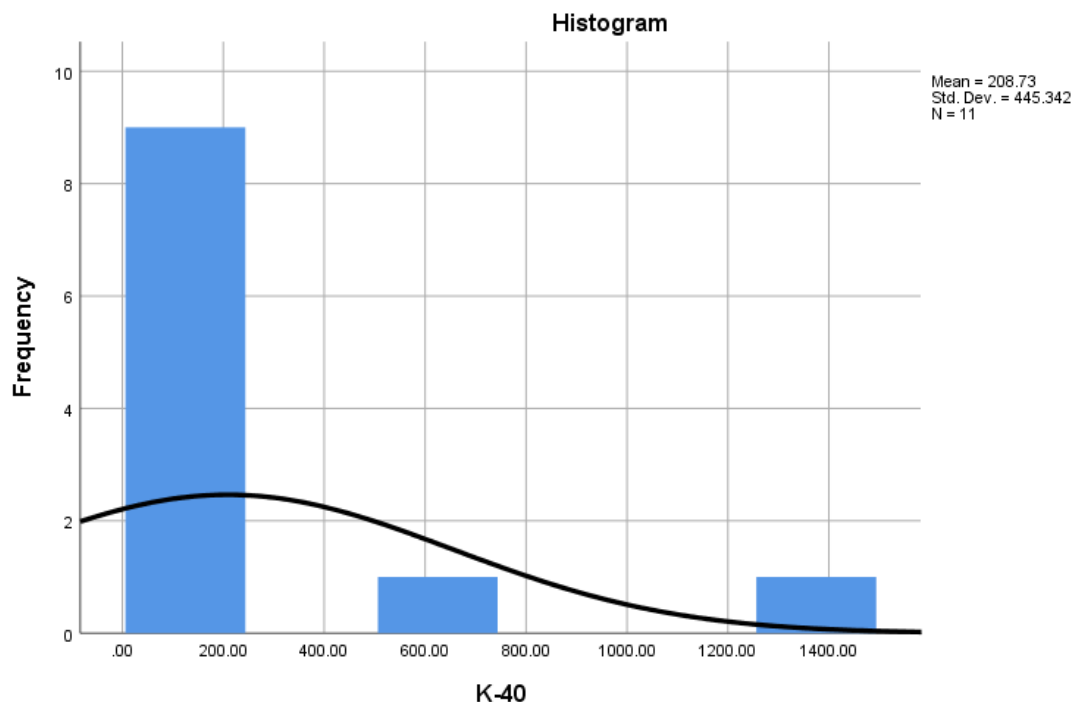


Fig.4: The distribution frequency of concentration of ^{40}K

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Conclusion

Oil and gas companies generate a vast amount of produced water; the management process consists of one of the global challenges for several petroleum facilities, produced water must be either re-used or disposed of. Re-use operation of produced water requires some treatment to remove oil residue. However, some produced water does not meet the criteria required for it re-used, so almost all of the produced water has been disposed of. According to the disposal of waste from petroleum industries, a significant amount of NORM is released into the soil or the seawater, NORM is in the form of different radionuclides including Ra isotopes primarily which decay into the Rn^{222} gas, and these progenies such as Po^{218} , Po^{214} , Bi^{214} , and Pb^{210} is transferred to the environment, and then act and result in a dangerous effect to the human, according to several pathways either internal exposure from inhalation of radon gas present in the air, and digestion from food that comes from the contaminated soil, or external exposure. The values of radionuclides are almost above the recommended limits set by the US EPA in different oilfield regions around the world, which lead to more concern about human health and environmental pollution, so a depth study is recommended focusing on minimization and other management methods of produced water instead of being disposed of. Furthermore, the NORM waste and its influence could be reduced by complying with the recommended standard set by IAEA

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