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Review About NORM Concentration and Behavior in Produced Water from Oilfield Activities and Its Assessment Methodology for Human Exposure

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***Abstract*—Produced water that is considered as the harmful waste result from petroleum extraction process according to the significant concentration of Naturally Occurring Radioactive Material (NORM) is frequently disposed into the nearby areas, leading to the contamination that increase the possibility of human exposure to NORM in various pathways. This paper aims to provide a comprehensive review of NORM reported in produced water associated with the oilfield activities from previous studies and the methodology often followed for assessing radiation risks of human exposure. The comparison of NORM values in oilfields worldwide provides more understanding of radionuclides behaviors and the oil extraction operation. The results indicate Ra isotopes as the most radionuclides present in produced water; especially, radiological indices noticed to vary from one region to another in a high average. Oil extraction is increasing, which leads to more produced water being disposed of, which consist of genuine concern for human health, 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 and other environmental protection agencies.**

***Index Terms*- NORM, Oilfield waste, Produced water, radium isotopes, radioactivity assessment**

I. Introduction

When the well has been drilled into the oil zone of the reservoir, where there is an aquifer underneath, and usually a layer of gas setting on the top, to get the flow 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 enter the well was inevitably mixed with a considerable 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 particular chemical and physical treatment technologies[1]-[2]-[3]. This water is generally comprised of the organic and inorganic compounds, NORM radionuclides, in addition to other chemical elements. So that its disposal process is one of the global concerns, consequently leading to human exposure in different pathways Figure 1.

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Naturally, radiation presents due to the cosmic ray or the naturally occurring radioactive materials (NORMs) that

originated in the earth's crust and are present everywhere in the environment [4]. Produced water has been reported to Contain significant values of NORM [5]-[6]-[7]-[8]. Almost elements are constituents from stable nuclides; however, U, Th are unstable by nature [9] and will fade in time by disintegrations into other radioactive elements by emitting Alpha and Beta particles accompanied by gamma rays. A uranium-235 nucleus goes through 11 transformations to become stable lead-207. A thorium-232 nucleus goes through 10 transformations, counting Ra^{228} and Ra^{224} to become stable lead-208. Moreover, a uranium-238 nucleus goes through 14 transformations, including Ra^{226} and Rn^{222}, to become stable lead-206, in addition to K^{40}, one of the three isotopes of K, which is widely distributed identically with its isotopes, this implies that the presence of K will be accompanied with K^{40} as well. K^{40} disintegrates once into either Ca^{40} or Ar^{40} emitting β-particle (89%) or γ-photon (11%) respectively.

The radiation hazard indicators were identified as assessment indices of human exposure to NORM different studies. Radium equivalent activity Bq, absorbed gamma dose rate AGDR (nGy) annual effective dose rate AEDR (mSv), external hazard index, internal hazard index, annual gonadal dose equivalent AGDE (mSv), activity utilization index AUI, excess lifetime cancer risk ELCR, activity concentration index, and alpha index [10]-[11]-[12].

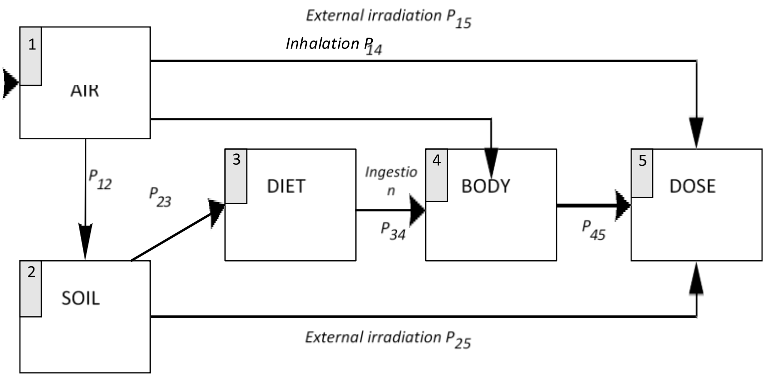

Figure I. Terrestrial pathways of transfer of radionuclides and dose to humans. (4)

II. PRODUCED WATER IN OIL COMPANIES

The major challenge in the oil industry recently 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 [13]. Produced water represents an enormous waste stream because of several oilfields' operations. During the oil extraction process, a tremendous 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 [14]. The first source of produced water follows the oil and gas extraction process, while formation water which exists below the oil layer then enter through the porous reservoir and comes out of the well mixing with the crude oil; this process leads to a reduction in reservoir pressure, resolve this problem by injected water again in the reservoir system to maintain the hydraulic pressure [15], this injected water present the second source of produced water based on the fact, the more oil extraction, 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 [14]. This saline water is called then formation water and becomes produced water when it is brought up and oil to the surface as a mixture. In some other cases and due to the reduction of pressure in the reservoir, this water will be injected again to maintain the hydraulic pressure and enhance the oil recovery, injected water usually from injectors wells towards the formation which directed 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 produced water, this mixture contains in addition of the water and oil, metals that have been reported in various studies including Cr, Ba, Ni, Zn, Mg, Fe, Ni, Pb and K [16], furthermore lead, nickel, zinc, cadmium, and copper usually exist as heavy metal in oil and gas field produced water heavy metals were transformed from a dissolved state to particles in water under oxygenated conditions[17], along with radium and radon, treating chemicals, salt and dissolved oxygen. The stream of the produced water is considered as the main waste in terms of size subsequent from the oil and gas facilities [18]-[19]-[20]-[21]-[22]-[23]-[24].

III. NORM CONCENTRATION IN PRODUCED WATER

The radioactivity concentration in a given volume of water represents the levels of radioactivity in produced water; 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 1 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 1, we notice that Ra isotopes are the dominant radionuclides in produced water, especially Ra^{226} , Ra^{228} and Ra^{224} . Ra^{226} , which results from U^{238} , decays into Rn^{222} by emitting alpha and beta particles, in addition to gamma radiation, to reach a stable state over 1600 years of half-life. *On the other hand*, Ra^{228} , Ra^{224} are daughters products of the Th^{232} decays chain, which decays into Ac^{228} , Rn^{220} respectively; Ra^{228} reach the ground state by emitting beta particles and gamma rays with an estimated half-life of 5.75 years, while Ra^{224} end up in the stable state through decays by emitting alpha particles and gamma rays over 3.7 days of half-life. The Ra^{224} noticed as the lowest radium isotopes present in produced water because Ra^{224} appears in produced water without its immediate parents Th^{228} , so that will die out within two weeks of secular equilibrium, the same period for Ra^{226} to reach its secular equilibrium with Rn^{222} , Po^{218} , Po^{214} , Bi^{214} , and Pb^{210} , while Ra^{228} considered as the quick radium isotopes that reach its equilibrium with Ac^{228} withing two days.

Ra^{226} noticed as the most Ra isotopes present in produced water in different studies from different areas [21],[25]-[26]-[27]–[28],[38], one of the reasons can be due to its high solubility in water and its behavior preferring the aqueous state, furthermore, Ra^{226} is chemically similar to Barium Ba, Strontium Sr, Calcium Ca. Magnesium Mg so that it becomes incorporated in group II sulfate or carbonate deposits and scale[9], high precipitation of Ra^{226} reported with strontium and barium which are taken part in the metals present in produced water, this result is according to various previous experiments that aim to find the correlation between radium isotopes and metals. [45]-[46]– [47].

U^{238} and Th^{232} concentrations are noticed from table 1 that were measured according to their progenies Ra^{226} and Ra^{228} respectively [35],[44],[45],The absent of U^{238} , Th^{232} in produced water in some studies [24],[30],[32]-[33]-[34]–[35]. according to their chemical characteristics, they 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.[9]

Results display a high value of K^{40} activity concentration, of range (1.65-1460) $Bq.L^{-1}$ in produced water owning to the fact that K isotopes are widely distributed in nature (abundance in the Earth's crust 2.1%), including K40 (0.0117%). However, K^{40} concentrations in produced water are lower than the values found in soil samples around the oilfield area [48].

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

Radionuclides	U^{238}	Th^{232}	K^{40}	Ra^{226}	Ra^{228}	Ra^{224}	Ref
Congo($Bq.dm^{-3}$)	<4.5×$10^{-3}c$	<4.5×$10^{-3}c$	-	5.1c	-	-	[21]
Egypt	-	39.9c	66c	19c	-	-	[25]
Iraq	-	9.4c	66.4c	20.3c	-	-	[26]
Romania	(0.043-1.1)	(0.21-8)	(221-899)	(23-45)	-	-	[27]
Syria	-	19.2c	1460c	186.2c	-	-	[28]
Ghana	(0.11-1.03)	(0.21-0.56)	(1.65-11.99)	-	-	-	[29]
Ghana	-	-	(5.90-23.90)	(6.20-22.30)	(6.40-35.50)	(0.78-7)	[30]
Nigeria	-	-	39.8c	8.9c	8.1c	-	[31]
Nigeria	-	-	(9.08-155.22)	(2.01-13.19)	(0.75-12.30)	-	[32]
US ($pCi.L^{-1}$)	-	-	-	(56-1494)	(69-600)	-	[33]
US	-	-	-	(30-2690)	(35-763)	-	[34]
US	-	-	-	(<0.002-58)	(0.02-59)	-	[35]
Azerbaijan	-	(ND-13.71)	(26.1-194.5)	(ND-101.7)	-	-	[36]
Poland	<30	-	75c	<2	<2	-	[37]
Texas	-	-	-	(0.1 − 5,150)	ND	-	[38]
Brazil	-	-	-	(0.012-6)	<0.05-12	-	[39]
Norway	-	-	-	3.3c	2.8c	-	[40]
Norway	-	-	-	(0.5-16)	(0.5-21)	-	[41]
Syria	-	-	-	51.9c	37.5c	(0.2-3.7)	[42]
Oman	-	-	(1522-1535)	(514-529)	-	-	[43]
Turkey	-	-	-	6c	3.17c	2.83c	[44]

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

IV. Risks Assessment of Human Exposure to NORM

The dose of Radiation risks due to NORMs was assessed following two main techniques, first by measuring the gamma dose rate directly in indoor and outdoor, subtracting the radiation due to cosmic rays. On the other hand, the concentration of the radionuclides was measured and used for gamma radiation assessment [4]. Finally, the activity concentration of naturally occurring radioactive materials was included to assess the radiation risks into several radiological indices, counting the radiation dose assessment and radiation risk assessment.

For dose assessment, the absorbed dose rate in indoor and outdoor was noticed as the principal factor for dose assessment calculated [4], either directly or according to the radionuclides activity concentration. the absorbed dose Relevant to the radon inhalation pathway taken place in some studies as dose assessment index [49].based on the absorbed dose, the equivalent and effective dose were estimated in various studies to describe the amounts of gamma radiation resulting from NORMs decay in the air [50,51], some studies have specified the dose assessment for some tissue and organ in the body such as gonadal by measuring the AGDE, Furthermore, Excess lifetime cancer risk was a helpful index that was assessed in different studies based on the effective annual dose's value.

While in risk assessment, Radium equivalent activity was the widely used index to assess the radiation hazards [52,53], which based on the assumption that 370 $Bq.Kg^{-1}$ of Ra^{226}, 259 $Bq.Kg^{-1}$ of Th^{232} and 4810 $Bq.Kg^{-1}$ of K^{40} produced the same gamma dose rate, their limits of 370 $Bq.Kg^{-1}$ correspond to an effective dose of 1 $mSv.y^{-1}$ [54]. The external hazard index, another index was frequently used which signifies the radiation hazards due to the external

Table 2: Radiation hazard indicators values in different region in the world, the world average and the recommended values.

Radiological indices	Ra_{eq} $(Bq.Kg^{-1})$	D $(nGy.h^{-1})$	H_{in}	H_{ex}	ELCR× 10^{-3} mSv	AEDE $(mSv.y^{-1})$	I_{γ}	AGDE $(nGy.h^{-1})$	AUI	Ref
Iraq	83.95	41.76	0.308	0.214	-	0.204	0.612	-	-	[57]
Nigeria	205.67	186.8	0.67	0.56	3.21	0.92	-	701	1.2	[58]
India	99.35	45.189	0.33	0.268	0.19	0.22	-	316.72	0.713	[59]
India	84.57	41.7	0.051	0.228	0.180	0.051	0.64	0.282	0.496	[60]
India	961.8	414.1	-	2.6	1.7	507.8	3.3	2850	-	[61]
India	68	61.72	0.19	0.18	0.39	0.11	0.91	350.63	0.71	[62]
China	364.9	332.3	1.62	0.99	-	1.63	-	-	-	[63]
India	102.56	86.95	0.2872	0.2777	0.3735	0.1067	0.762	0.3325	0.6651	[64]
Min	68	41.76	0.051	0.18	0.19	0.051	0.612	0.282	0.496	
Max	961.8	414.1	1.62	0.99	3.21	507.8	3.3	701	1.2	
Average	246.35	151.31	0.49	0.66	1.007	66.88	1.24	703.16	0.75	
World average	370	57	1	1	1.16	0.41	-	300	0.07	[4]
Recommended values	370	84	<1	<1	0.29	0.46	0.5	300	-	[4],[65]

Ra_{eq}, AGDR, AEDR, H_{ex}, H_{in}, , AGDE, AUI, ELCR, I_{γ} are radium equivalent activity in $Bq.Kg^{-1}$, absorbed gamma dose rate in $nGy.h^{-1}$, annual effective dose rate in mSv, external hazard index, internal hazard index, annual gonadal dose equivalent in mSv, activity utilization index, excess lifetime cancer risk, activity concentration index and alpha index, respectively

exposure to gamma radiation and the internal hazard index representing an assessment of the internal exposure due to radon and their progenies emissions were also utilized as an index for alpha radiation from radon and their progenies. [55].

Other radiological indices, such as the Alpha index I_{γ}, also known as activity concentration index, which generally contribute to assessing the hazard arising from gamma emission, the alpha index I_{α} which refers to the internal hazard of alpha radiation emitted via radon and their daughters' decay, activity utilization index AUI for simplicity the calculation of dose rate in the air was calculated [56]. Table 3 illustrate radiological hazard indices values in a different area in addition to the world average and the recommended level. Absorbed dose results demonstrate a high level in most of the regions [58],[61]-[62]-[63]-[64], which exceed the recommended levels 84sv, while its world average was below the permissible limits of 57 Sv, external and internal hazard index is within the recommended level, high values of Annual effective dose equivalent AEDE is remarkable in the world average that is relatively equal to the recommended limits, these significant values explain the exceeding value of ELCR in the world which based on the AEDE values for its calculation[58],[61]-[62],[64]. Ra equivalent activity varies from one region to another; due to the difference of radionuclides concentration of K^{40}, Ra^{226}, Th^{232} according to the geological characteristics [11], the radiological indices noticed as the high values in Kerala India [61].

Figure 1 shows the correlation between the Hazard Index, which indicate a positive and robust relationship between the two variables in several studies [57]-[58]-[59]-[60],[62]-[63]-[64], an abnormal distribution of, D, AEDE is remarkable according to the histograms in Figure 2, Figure3, Figure4 respectively in all studies, which confirm the effect of geologic characteristics in the distribution of NORM activity concentration.

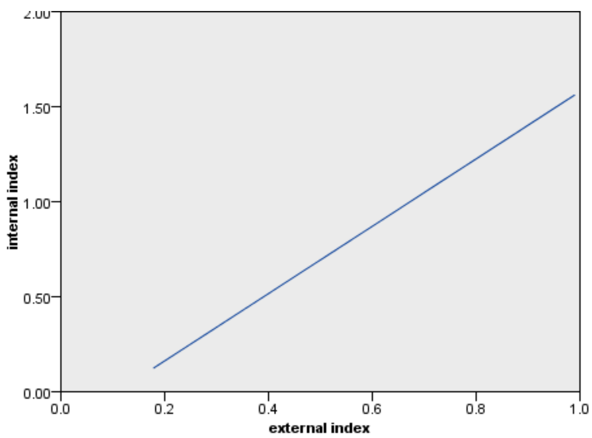

Figure 1: Correlation between Hazard Index (H_{ex} and H_{in}), for the different areas

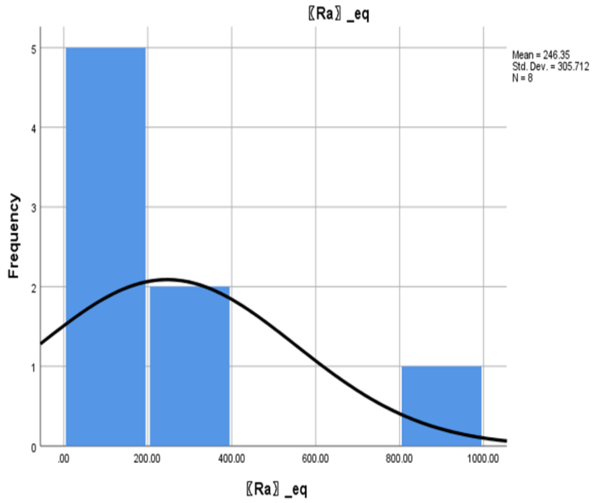

Figure 2: Radium equivalent activity Ra_{eq} distribution for the different areas.

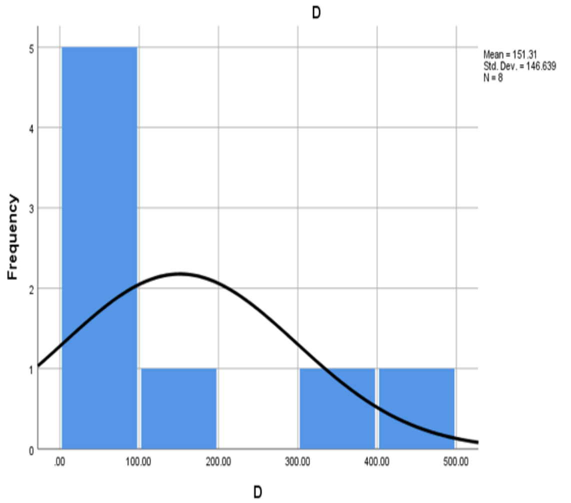

Figure 3: Absorbed Dose D distribution for the different areas

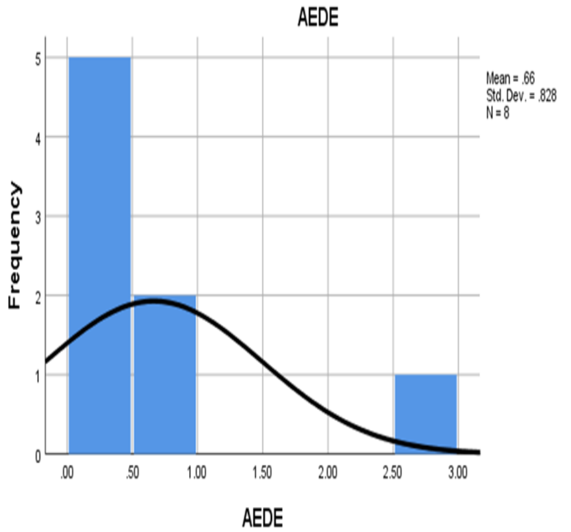

Figure 4: AEDE distribution for the different areas

V. 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-use 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 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 and the radiation hazard indicators are almost above the recommended limits set by 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 and other environmental protection agencies.

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