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RADIOLOGICAL HAZARD IMPACT OF RESERVOIR FRACKING WASTEWATER: DRINKING WATER IMPLICATION, NIGERIA

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ABSTRACT

Petroleum can be extracted from the earth reservoir through hydraulic fracking drilling technique. It is the technique being used by hydrocarbon operatives in Otu-Jeremi community. The wastewater released into natural water bodies during this process may contain natural occurring radioactive materials as a result of geological formation and radiologically hazardous chemicals used during production. This study area contains the local geological formations (Benin, Agbada and Akata formations) of the Niger Delta. This research thus investigated the radiological hazard impact of reservoir fracking on drinking wells water due to release of wastewater into Otu-Jeremi community waters. Thirty wells water samples were collected using 2-liter plastic containers and were analyzed using 3"x3" Sodium iodide [NaI(TI)] detector. The mean radionuclide concentrations values are 7.68±1.14 Bq/L, 5.70±0.92 Bq/L, and 30.40±1.54 Bq/L respectively. These values exceeded the (WHO, 2008) limit values of 1.0 Bql^{-1}, $0.1Bql^{-1}$ and $10.0Bql^{-1}$ respectively. The radiological hazard risk were computed using scientific mathematical models and their mean values are 18.07±2.65 BqL^{-1}, 0.129 $mSvy^{-1}$, $0.0499mSvy^{-1}$, $0.0803mSvy^{-1}$, 8.364 ηGyh^{-1}, 0.103 mSv/y, 0.410 mSv/y, 0.029 mSv/y and 0.114 mSv/y respectively. The radiological elements computed reveal that AEDE and ELCR values did not exceeds recommended limits. Although the overall results obtained do not indicate potential radiological risks in the community drinking water. However, as a preventive major, it is essential to conduct regular radiological monitoring of public drinking water to ensure future potential risks on the residents.

Keywords: Radiological, Oil and Gas, Hydraulic Fracking, Water, Environment

INTRODUCTION

Hydraulic fracturing is one of the very few unconventional methods employed by operators of oil and gas to extract oil and natural gas from the earth (Kerr, 2010; Arthur *et al.*, 2009). This technique has improved exploration and production capacity due to increase in demand for petroleum byproducts in recent years (Avner *et al.*, 2013; Kargbo *et al.*, 2010; Kerr, 2010). However, during production, crude oil may spill and possible discharge of hazardous waste (produced water) into the environment that may lead to environmental and public health impacts on the residents. Hence various international organizations have set up standard and policies to safeguide the environment from radiological hazards (Howarth *et al.*, 2011; Goldstein *et al.*, 2012; Avwiri and Esi, 2014; Shi *et al.*, 2021). One of the concerns regarding hydraulic fracking technique is its impact on drinking waters (Vidic *et al.*, 2013; Gregory *et al.*, 2011) which occurred as a result of the release of naturally occurring radioactive materials (NORMs) among the other constituent from the drilling process which may increase the level of radiological pollution of the environment (Osborn *et al.*, 2011; Warner *et al.*, 2012; Nte *et al.*, 2013; Brown, 2014). These NORMs such as produce water, sludge scales etc may have potential radiological public health impacts on the environment.

In Nigeria, crude oil exploration and production started in 1956 after its discovery in Oloibiri oil field, Niger Delta and gradually spread across the Niger Delta region. The research area (Otu-Jeremi community) has experienced oil and gas operations which may have led to spill and released of fracking wastewater containing radiological elements and may have impacted on the drinking water of the residents.

Consequently because of these, to the best of our knowledge, there is no documented literature on the radiological hazard impact of reservoir fracking wastewater on drinking water in Otu-Jeremi community in particular and the Niger Delta region in general. Lately, the public health centers in the community have recorded similar health-related issues such as skin cancer, breast cancer, leukaemia, eye cataract, sterility, genetic disorders etc that may be traced to wastewater pollution. Hence, these study to evaluate radiological contamination level, potential health impact and implication of reservoir fracking wastewater in drinking water of the area. Therefore, this research will serve as baseline study of the area. Otu-Jeremi is the headquarter of Ughelli South Local Government Area, Delta State, Nigeria. The inhabitants is about 23,576 with occupational history of farming, fishing trading and government workers (ministry and oil and gas). The area has geographic coordinates of l*atitude 5.433 and longitude 5.867*The area play host to Otorogun gas plant and many oil fields (Agaja, 2012).

Figure 1: Map of Otu-Jeremi town

MATERIALS AND METHODS

Sample Collection and Preparation

In Otu-Jeremi town, a total of thirty water samples were collected (in triplicates, this is to ensure dependability of results) from ten hand-dug wells (three per well). Each water sample was given a code 1 to 3 per location and the mean coding per location ranging from HDWW 1 to HDWW 10. During the sampling process, 2-liter plastic containers with approximately 1% air space were used to collect the samples. To minimize contamination from the container itself a deionized water was first used to wash the inner part of the container and allow to dried. However, the well water to be sampled was also used to wash the sample containers thoroughly foe about three to four times before sampling the well water. After sample collection, 10ml of acid per liter (2 M HCL) of water solution was added to the sampled water to avoid assimilation of radioactivity on the walls of the container. Subsequently, the collected samples were forwarded to Obafemi Awolowo University's Energy Research Centre for detailed sample preparation and analysis. Preparations of samples were carried out using one-liter capacity beakers that were thoroughly washed and rinsed with water down sulfuric acid and subsequently carefully desiccated in an oven. The beakers were filled with a predetermined quantity of samples were tightly sealed for an interval of thirty days (30 days before radionuclide concentrations analysis was done via 3"x3" Sodium iodide [NaI(TI)] detector. This procedure was carried out to ensure the preservation of radon and establish an equilibrium state in the samples.

Sample analysis

The detector, thallium-activated 3"x3" Sodium iodide [NaI(TI)] attached to an ORTEC 456 amplifier, used was in a lead shielded house of about thickness of 100mm, and a computer software SAMPO 90 was connected to the detector. In order to ascertain accurate quantitative measurements of the samples, appropriate energy calibration was carried out on the detector using Cs-137 and Co-60 system efficiency standard sources with resolutions energy of 39.5% and 22.2% respectively. These standard sources were purchased from IAEA in Vienna. For background measurement, spectra were accumulated for 29000 seconds at 900 volts, resulting in strong peaks at specific gamma emitting energies: 1460keV for 40K, 609keV for 214Bi, and 911keV for 228Ac. These peaks were used to evaluate the concentration of ^{238}U and ^{232}Th respectively. During the calibration process, the activity levels of the standards were determined to be 25.37KBq and 4.84KBq for Cs-137 and Co-60 respectively. The surroundings background spectra were used to adjust the calculated activity concentrations of the samples. This correction was done in accordance with the methodology described by Avwiri *et al.* (2007) and Agbalagba *et al.* (2012). Subsequently, the sample concentration (C) of the radionuclides were calculated in units of Bql^{-1} (becquerels per liter) after applying decay correction and using the expression equation provided in equation 1.

$$C_S = \frac{N_{ey}}{Q_{ey} \times V_y \times P_y \times R_y} \ (\mathrm{BqL^{-1}}) \qquad (1)$$

C_s = Concentration of sample, N_{ey} = energy net peak area, Q_{ey}= the γ-energy of interest efficiency of the detector, V_y = Sample volume, P_y = Counting period summation, R_y = Emission probability interest of the radionuclide.

Calculation of Radiation Hazard Indices

Table 1: Formulas for Calculating Radiation hazard risk of Community Drinking Water Implication (UNSCEAR, 2000; Diab et al., 2008; Esi and Akpoyibo, 2024)

S/No	Hazard Index	Formulas	
1	Radium Equivalent Index	$Ra_{eq}=H_{Ra}+1.43H_{Th}+0.077H_k$	(2)
2	Representation Level Index (I_{yr})	$I_{yr} = \frac{H_{Ra}}{150} + \frac{H_{Th}}{100} + \frac{H_k}{1500}$	(3)
3	Absorbed Does Rate (D)	$D=0.462H_{Ra}+0.621H_{Th}+0.0417H_k$	(4)
4	External Hazard Index (HI_{ex})	$HI_{ex} = \frac{H_{Ra}}{370} + \frac{H_{Th}}{259} + \frac{H_k}{4810}$	(5)

5	Internal Hazard Index (HI_{in})	$HI_{in} = \frac{H_{Ra}}{185} + \frac{H_{Th}}{259} + \frac{H_k}{4810}$	(6)
6	Annual Effective Dose Rate (Outdoor)	*AEDR (mSvyr^{-1}) = D (nGrh^{-1}) × 8760 hryr-1×0.7 × (10^{3}mSv/10^9) nGy × 0.2*	(7)
7	Annual Effective Dose Rate (Indoor)	*AEDR (mSvyr^{-1}) = D (nGrh^{-1}) × 8760 hryr-1× 0.7 × (10^{3}mSv/10^9) nGy × 0.8*	(8)
8	Excess Lifetime cancer risk (ELCR)	*ELCR = AEDE × LD × RF*	(9)

Where all parameters retained their usual meaning: Where Ra_{eq} is the radium equivalent index, I_{yr} is the representative level index, D = absorbed dose rate, HIex and HI_{in} are external hazard index and internal hazard index and H_{Ra}, H_{Th} and H_K, are the activity concentrations symbols representing values of ^{238}U, ^{232}Th and ^{40}K respectively. For AEDE the following constant values are recommended for both outdoor and indoor of 0.7 Sv/Gy conversion coefficient and 0.2 and 0.8 for outdoor and indoor factor of occupancy respectively (UNSCEAR, 2000). Where, AEDR is the annual effective dose rate. Also, for ELCR, AEDE is the annual effective dose Equivalent, LD is life duration (55.75 years) (Ramasamy *et al.,* 2009; Avwiri and Esi, 2015) and RF is risk factor (Sv^{-1}), fatal cancer risk. ICRP 60 uses values of 0.05 for the public for stochastic effects (Ramasamy *et al.,* 2009; Mokobia et al, 2020).

RESULTS AND DISCUSSION

The radiological hazard impacts of reservoir fracking wastewater in the studied samples of Delta Central Community (Otu-Jeremi) town are specified in Table 2. The mean radionuclide concentrations values varied between 3.7±0.6 to 11.7±0.4 Bq/L with a mean of 7.68±1.14Bq/L, 3.9 ± 0.6 to 8.2 ± 1.0Bq/L with a mean of 5.70±0.92Bq/L and 24.2 ±1.5 Bq/L to 42.1±0.3 Bq/L with a mean of 30.40±1.54 Bq/L for ^{238}U, ^{232}Th and ^{40}K respectively. The obtained mean values when compared with world limits, exceeded the (WHO, 2008) limit values of 1.0 Bq/L, 0.1 Bq/L and 10.0 Bq/L for ^{238}U and ^{232}Th and ^{40}K correspondingly. The findings are noticeably depicted in figures 1-3. However, ^{40}K results in the water samples is consistently higher than values of ^{238}U and ^{232}Th in all points of sampling and this may be ascribed to the area geographical structure and the content of mineral substances explored by the operators of the oil industry (Agbalagba *et al.,* 2013; Shi *et al.,* 2021). It was observed from the present study which was compared with similar scientific reports as presented in table 4. The obtained results were observed to be greater than the reported values from Algeria; Samsun, Turkey; Anglian, England; Kirkuk, Iraq; Saudi Arabia and Ghana. However, the observed values are in agreement the scientific report in Nigeria. The mean calculated radiological results of radium equivalent (Raeq) ranged from 12.23± 2.9 Bq/L to 25.61± 2.0 Bq/L with mean value of 18.07±2.65 Bq/L. The obtained results of Raeq are lower than the world limit of 370 Bq/L (Agaja and Ajisafe, 2013; Mohammad *et al.,* 2015). The mean representative index (Iyr) ranged from 0.088 mSvy^{-1} to 0.179 mSvy^{-1} with mean of 0.129 mSvy^{-1}.

Table 2: Mean activity concentration of radionuclides in hand dug well water from Otu-Jeremi town

S/N	Coding	^{238}U (Bql^{-1})	^{232}Th (Bql^{-1})	^{40}K (Bql^{-1})
1	HDWW 1	8.5 ± 1.6	5.3 ± 0.8	24.2 ± 1.5
2	HDWW 2	9.6 ± 0.9	4.8 ± 1.2	28.5 ± 1.8
3	HDWW 3	7.8 ± 1.1	6.6 ± 0.7	31.1 ± 2.2
4	HDWW 4	11.7 ± 0.4	8.2 ± 1.0	28.4 ± 2.1
5	HDWW 5	6.9 ± 1.2	7.0 ± 1.3	42.1 ± 0.3
6	HDWW 6	6.4 ± 0.4	5.1 ± 0.6	26.8 ± 1.8
7	HDWW 7	9.2 ± 1.4	5.6 ± 0.7	35.7 ± 3.1
8	HDWW 8	3.7 ± 0.6	4.4 ± 1.1	29.02± 0.9
9	HDWW 9	7.4 ± 1.5	3.9 ± 0.6	32.09 ±0.3
10	HDWW 10	5.6 ± 2.3	6.1 ± 1.2	26.10 ±1.4
	MEAN	**7.68±1.14**	**5.70±0.92**	**30.40±1.54**

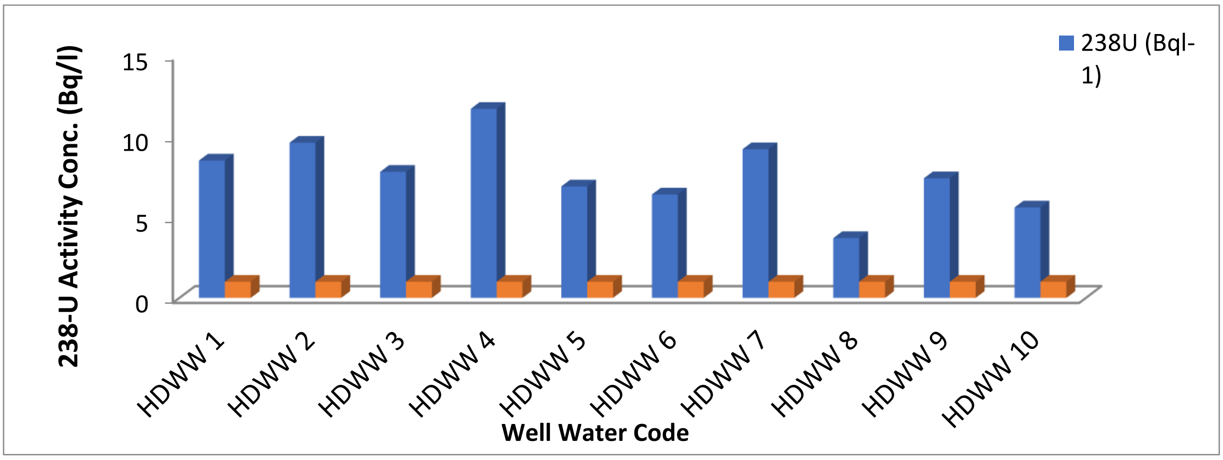

Figure 2: Relationship of ^{238}U obtained values (Bql^{-1}) in well water with WHO world limit

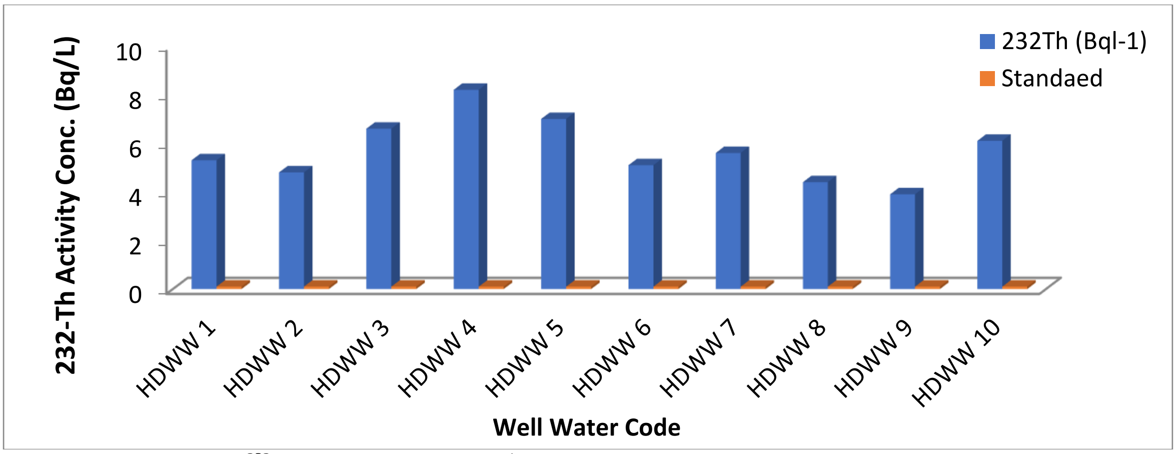

Figure 3: Relationship of ^{232}Th obtained values (Bql^{-1}) in well water with WHO world limit

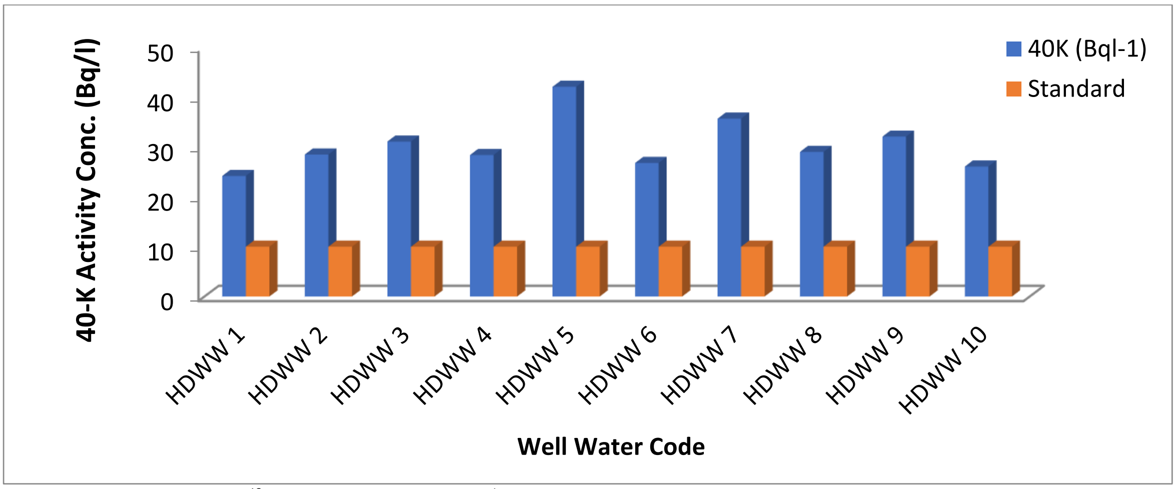

Figure 4: Relationship of ^{40}K obtained values (Bql^{-1}) in well water with WHO world limit

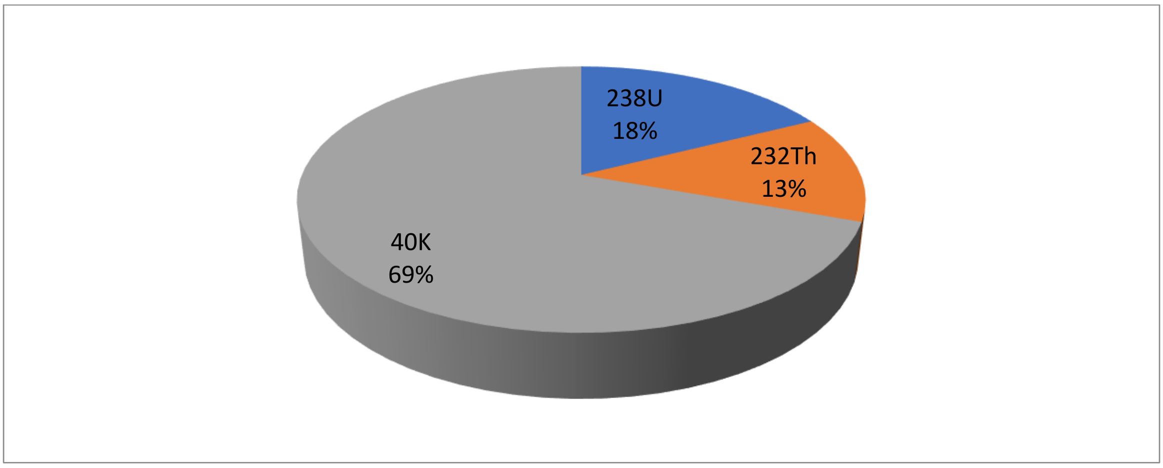

Figure 5: Percentage distribution of Radionuclides in well water

Table 3: Computed Radiological Hazard Values

S/N	Code	Ra_{eq}	Iyr	Hex	Hin	D	AEDE (Outdoor)	AEDE (Indoor)	ELCR×10^{-3} (Outdoor)	ELCR×10^{-3} (Indoor)
1	HDWW 1	17.94± 2.9	0.126	0.0492	0.0714	8.227	0.101	0.404	0.028	0.113
2	HDWW 2	18.66± 2.8	0.131	0.0511	0.0764	8.605	0.106	0.422	0.029	0.118
3	HDWW 3	19.63± 2.3	0.139	0.0540	0.0741	8.999	0.114	0.441	0.032	0.123
4	HDWW 4	25.61± 2.0	0.179	0.0703	0.1008	11.762	0.144	0.577	0.040	0.161
5	HDWW 5	19.10 ±3.1	0.144	0.0554	0.0731	9.290	0.114	0.456	0.032	0.127
6	HDWW 6	15.76± 1.4	0.112	0.0432	0.0599	7.242	0.089	0.355	0.025	0.099
7	HDWW 7	19.96± 2.6	0.141	0.0547	0.0787	9.217	0.113	0.452	0.032	0.127

8	HDWW 8	12.23± 2.9	0.088	0.0336	0.0430	5.652	0.069	0.277	0.019	0.077
9	HDWW 9	15.45± 2.4	0.110	0.0427	0.0617	7.179	0.088	0.352	0.025	0.098
10	HDWW 10	16.33± 4.1	0.116	0.0450	0.0593	7.464	0.915	0.366	0.026	0.102
	MEAN	**18.07±2.65**	**0.129**	**0.0499**	**0.0803**	**8.364**	**0.103**	**0.410**	**0.029**	**0.114**

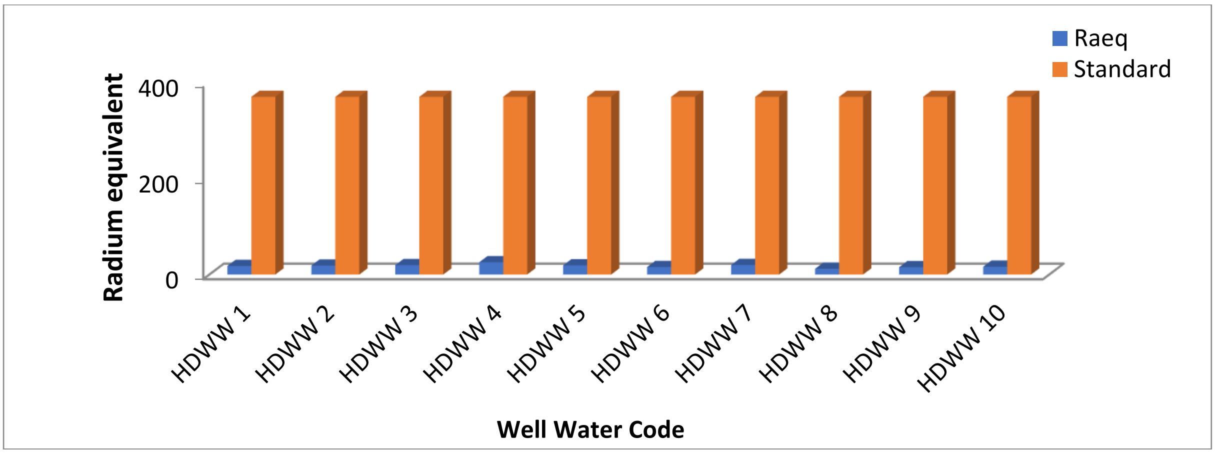

Figure 6: Comparison of the Raeq against UNSCEAR Standard

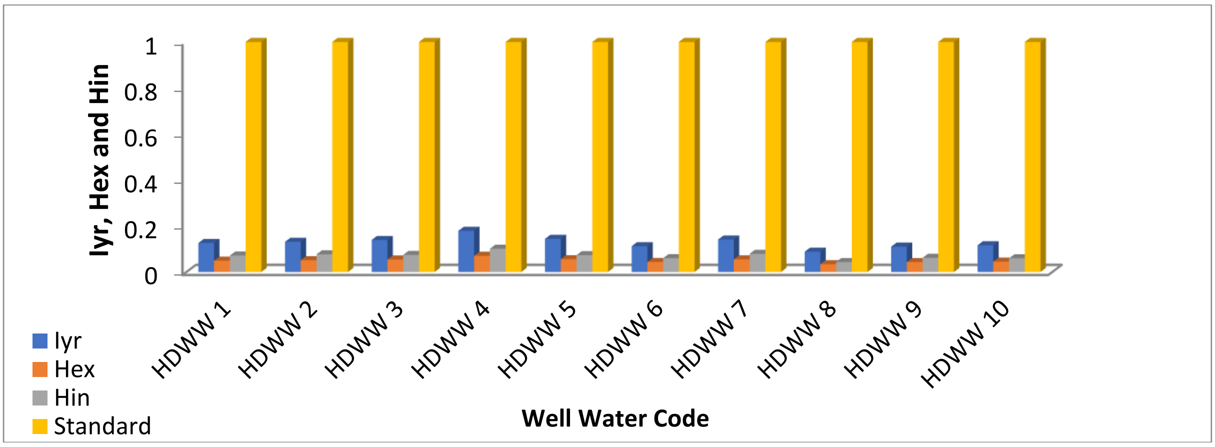

Figure 7: Comparison of the Representative index (Iyr), External hazard index (Hex) and Internal hazard index (Hin), against UNSCEAR Standard

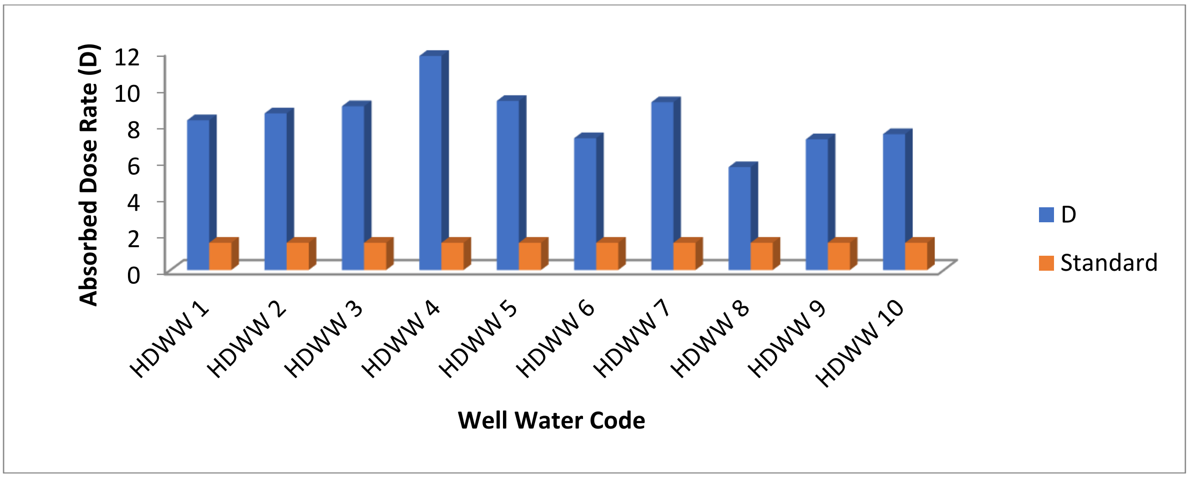

Figure 8: Comparison of the Absorbed Dose Rate(D) against UNSCEAR Standard

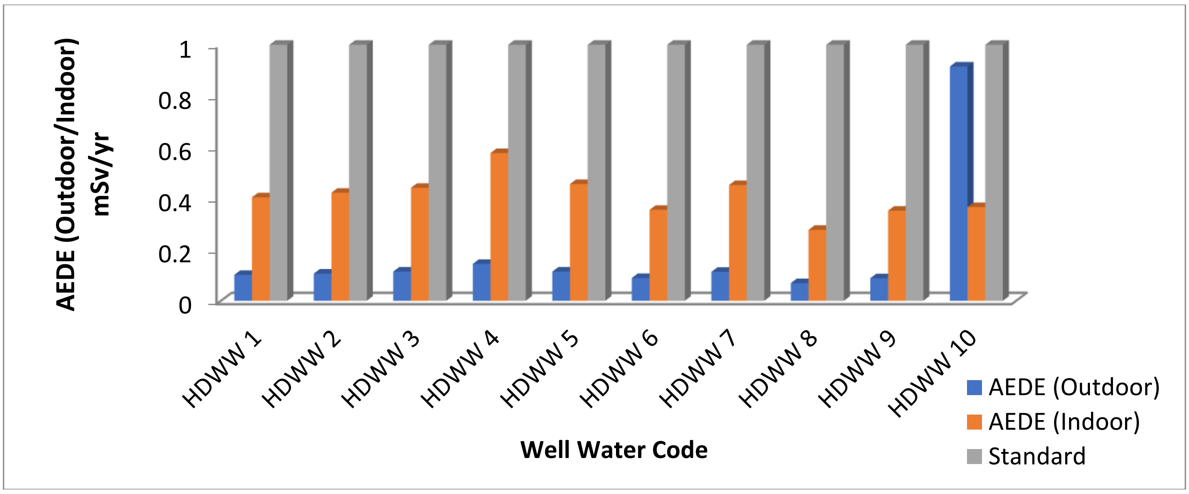

Figure 9: Comparison of the AEDE Outdoor and Indoor against UNSCEAR Standard

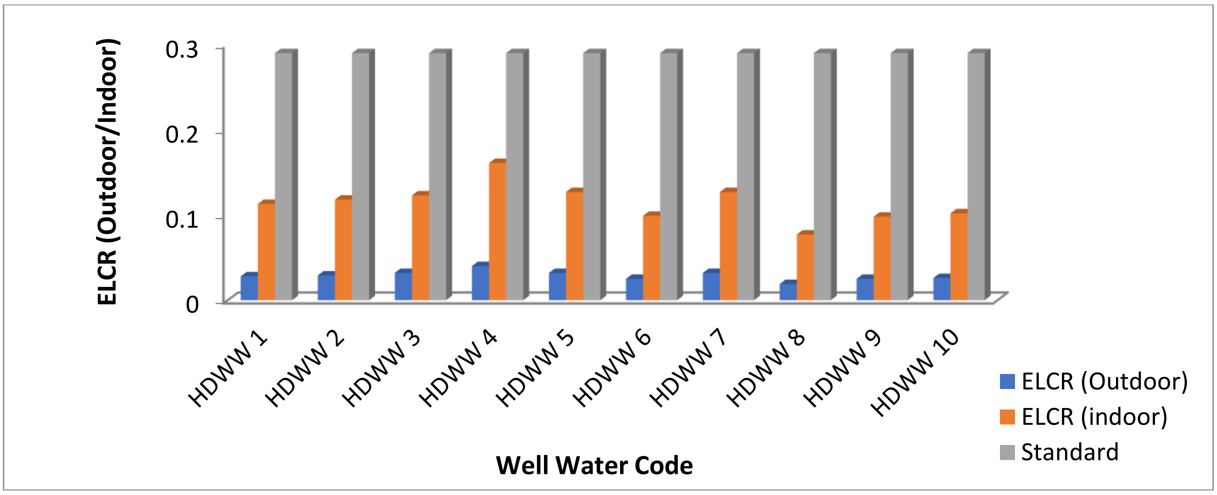

Figure 10: Comparison of the ELCR Outdoor and Indoor against ICRP Standard

Table 4: Similarity of studied values of radionuclide concentrations with reports from other countries

Country	Natural Activity concentration of radionuclides			Reference
	^{238}U	^{232}Th	^{40}K	
Algeria	63.56mB/L	9.83mBq/L	6.80mBq/L	Kebir, 2022
Turkey (Samsun)	419 mBq/L	142mBq/L	806mBq/L	Aydan et al, 2015
England (Anglian)	1.2±0.38Bq/L	4.88±2.20 Bq/L	3.56±6.45 Bq/L	Beresford et al, 2007
Iraq (Kirkuk)	121.95 mBq/L	81.52 mBq/L	1091.37 mBq/L	Najeba&Murtadha, 202
Saudi Arabia	0.32 Bq/L	0.12 Bq/L	10.96 Bq/L	Al-Zahrani, 2016
Ghana (Bomaa)	0.38 ±0.02 Bq/L	0.41 ±0.02 Bq/L	4.24 ±0.32 Bq/L	Darko et al, 2015
Nigeria (Delta State)	6.04±2.48 Bq/L	5.18±2.14 Bq/L	48.78±13.67 Bq/L	Avwiri et al, 2013
Nigeria (Out-Jeremi)	7.68±1.14 Bq/L	5.70±0.92 Bq/L	30.40±1.54 Bq/L	Present Study

The mean results of HIex and HIin ranged from 0.0336mSvy^{-1} (0.0430mSvy^{-1}) to 0.0703mSvy^{-1} (0.1008mSvy^{-1}) with mean 0.0499mSvy^{-1} (0.0803mSvy^{-1}) respectively. It was observed from the computed values that HIex and HIin are lower than unity (Agaja and Ajisafe, 2013; Tchokossa *et al.,* 2011) which is the recommended international limit. The mean absorbed dose rate (D) of computed radiological results ranged from 5.652ηGyh^{-1} to 11.762ηGyh^{-1} with mean value of 8.364 ηGyh^{-1}. These results are in agreement with scientific reports of drinking water from other similar study area (Tchokossa *et al.,* 2011; Ajayi and Achuka, 2009; Aguko *et al.,* 2020; Nwankwo, 2013; Fasunwon *et al.,* 2010). The mean results of AEDE outdoor and indoor ranged from 0.06 mSvy^{-1} to 0.144mSvy^{-1} with mean of 0.103mSvy^{-1} and 0.277mSvy^{-1} to 0.577mSvy^{-1} with mean of 0.410mSvy^{-1} respectively. It was observed that the AEDE outdoor and indoor values are below the world limit of 1.0 mSvy^{-1}. These results are also in agreement with reported scientific literature (Agbalagba *et al.,* 2012; Avwiri *et al.,* 2007; Guogang and Giancarlo, 2007). The mean ELCR outdoor and indoor have a minimum value of 0.019 mSvy^{-1} and maximum of 0.040 mSvy^{-1} with mean of 0.029 mSvy^{-1} and 0.077 mSvy^{-1} to 0.161 mSvy^{-1} with mean of 0.114 mSvy^{-1} respectively. It is evident that the obtained values are lower than 0.29 x 10^{-3} world limit (ICRP, 2003; Ramasamy *et al.,* 2009). These may be attributed to discharge of fracking wastewater into the environment area and geology structure of the area. These results do not indicate an immediate radiological risk to the drinking water of Otu-Jeremi, but may have long term associated radiologically potential risks if not manage properly.

CONCLUSION

The objective of the study is to investigate radiological safety and quality of drinking water being consumed by residents of Otu-Jeremi town with regards to the release of reservoir fracking wastewater from oil exploration in the locality. A thallium-activated 3"x3" Sodium iodide [NaI(TI)] detector was used to analyze the concentrations of radionuclides in water samples and consequently, the radiological hazard impact was computed from the obtained results using mathematical models. The results indicate that the community's drinking water is safe for consumption since the computed radiological values do not pose any risk. However, necessary cautions such as constant quarterly radiation monitoring, introducing strict radiation regulations and guidelines for handling hazardous waste by both government agencies and oil and gas stakeholders and appropriate treatment of the reservoir fracking wastewater before release into the environment. Also, it is important creating public awareness about the potential radiological hazards associated with fracking wastewater if found in drinking water.

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