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Radiological Survey of Oil and Gas Wastes and its Health Risks in Niger Delta Region of Nigeria

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Authors' contributions

This work was carried out in collaboration among all authors. Author GOA designed the study. Author CPO performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors JIE and YECU managed the analyses of the study. Author JIE managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

Aim: The aim of this study is radiometric survey of oil and gas wastes and its health risks in Niger Delta region of Nigeria.
Study Design: This study was purely an experimental work which involves collection of samples and laboratory analysis.
Place and Duration of the Study: This study was carried out at oil and gas company waste stream facilities and waste pipe market within Niger Delta region between June 2018 and May, 2019.
Methodology: Sixteen samples (4 drill cuttings, 4 pipe scales, 2 sludges and 6 produced water) were randomly collected from four waste streams in six locations within the oil and gas production facilities and used pipe market. These samples were taken to the laboratory, prepared following the ISO procedure and packaged in a porcelain bottle, sealed and kept for twenty eight days in order to ensure secular equilibrium between ^{238}U, ^{232}Th and their progenies and counted with high purity Germanium detector (HPGe). The activity concentration of all the radionuclides were used to determine the radiological health risks using mathematical models.

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Results: The lowest and highest specific activity concentrations of ^{226}Ra, ^{238}U , ^{232}Th and ^{40}K, in solid wastes are 5.28±1.08 and 25727.75 $Bqkg^{-1}$, 3.61±0.76 and 23021.73±1041.58 $Bqkg^{-1}$, 2.40±0.56 and 21468.25±1125.57 $Bqkg^{-1}$ and 35.31±2.38 and 1527.73 ±86.60 $Bqkg^{-1}$ respectively. In the liquid waste, the lowest and highest activity concentration of ^{238}U, ^{232}Th and ^{40}K are 0.34±0.15 and 1.11±0.28 Bql^{-1}, 0.63±0.17 and 1.06±0.18 Bql^{-1} and 9.60±0.66 and 14.20 ±1.04 Bql^{-1} respectively. ^{226}Ra was below detectable limit in the liquid waste except in one sample (CZI). Downhole pipe scales recorded the highest activity concentration of all the radionuclides. Surface pipe scale also recorded very high activity concentration of all the radionuclides. The radiological health risk parameters assessed from the activity concentration of these radionuclide were all above the safe reference levels in downhole pipe scales and surface pipe scales while they are within the safe values in other samples. The estimated Exposure rate for both solid and liquid wastes were higher than the recommended reference level of 600 μRh^{-1} and the associated dose rate was also higher than reference level.

Conclusion: The result of this work revealed that downhole and surface pipes from oilfield commonly used in the construction of buildings and domestic overhead tank-stands are associated with high levels of ionizing radiation which may be detrimental to human health and the environment.

Keywords: Norm; radiological survey; sludge; HPGe; effective dose.

1. INTRODUCTION

Naturally Occurring Radioactive Materials (NORMs) are usually found in scales which are formed in pipelines, valves and pumps during oil and gas production. Components such as wellheads, separation vessels, pumps and other processing equipment can become NORM contaminated due to scale deposition on them and the associated sludge formation and other waste media [1]. This can create a potential radiation hazard to workers, general public and the environment if certain controls are not established. Naturally occurring radioactive materials (norm) is a by-product waste of oil production and its presence in pipelines, plant and machinery may cause restriction of operability and potential radiological health hazards [2]. Scale is caused by precipitation due to chemical reaction with the surface of the metal, a change in pressure and temperature and a change in the composition of the solution. Scale deposition occurs when the solution equilibrium of the water is disturbed by pressure and temperature changes, dissolved gases or incompatibility between mixing waters. All waters used in well operations can be a potential sources of scale [3].

Oil and gas equipment from waste stream are usually treated before disposal or released for sale. Waste materials contaminated with enhanced level of NORM requires to be disposed in a controlled manner to ensure it does not provide an unacceptable risk to the environment and the general public [4]. Researches have shown that petroleum industry generates several thousand tons of waste including produced water, scales sludges and contaminated equipment. The amount produced varies depending on the geological location, formation condition, type of production operation and age of the production well [5].

Oil and gas production wastes contain radionuclides found in nature, such as uranium, radium and thorium. They can become hazardous through potential ingestion or by direct exposure to radiation from the materials. Uranium and radium-226 are of particular concern because it decays to the radioactive gas radon. Studies have shown that exposure to high level of Radon can increase the risk of lung cancer. Radiological surveys conducted by EPA [6] showed that some equipment and disposal locations exhibited external radiation levels above 2 mRh^{-1} and soil contaminated with radium -226 above 37 Bqg^{-1}. Scale and some other wastes are low specific activity (LSA) radioactive materials. Because scale contains radium and its short lived decay products, scale and other Norms can (depending on the radium specific activity) generate intensive gamma radiation fields that are a source of personal external gamma radiation exposures. Inhalation of radon gas and other norm in particulates can lead to internal radiation exposures [7]. In order to manage personal radiation exposures, NORM management plan ought to be established in oil and gas industries. This plan is to ensure that personal radiation exposures are maintained not only below the relevant annual radiation exposure limit but as low as reasonably achievable (ALARA) [8].

Surface disposal of radioactive sludges/scales and produced water may lead to ground and surface water contamination. This will lead to exposure of the rig workers, waste disposal workers and nearby residents/office workers. The extent to which wastes are produced and the need to remove it regularly from the facilities vary strongly between reservoirs, individual wells, installations and production conditions [9].

The Niger Delta region is situated at the apex of the Gulf of Guinea on the west coast of Africa [10] and on the Nigeria's South–South geopolitical zone (Fig. 1). The Niger Delta, which is home to some 31 million people, occupies a total area of about 75,000 km^2 and makes up 7.5% of Nigeria's land mass. The Niger Delta region consists of 9 oil–producing states (Abia, Akwa Ibom, Bayelsa, Cross River, Delta, Edo, Ondo, Imo and Rivers) and 185 local government areas. This region cuts across over 800 oil–producing communities with an extensive network of over 900 producing oil wells and several petroleum production–related facilities [11]. Niger Delta is the third largest wetlands of the world with large fresh water swamp forest rich in bio-diversity [12]. This signifies the vastness in diversity of the ecosystem in need of conservation. The Niger Delta geological complex consists of the Akata, Agbada and Benin subsurface sedimentary formations [13].

International Association of Oil and Gas Producers (IOGP) [14,15] explains the various routes through which radioactive pollution occur during oil and gas production. ^{228}Ra, ^{228}Ra and ^{224}Ra leached from reservoir rocks are transported in produced water that are disposed into water bodies. Also water used in water flood operations and filtrate from completion work over or treating fluids contains norm which deposits in pipes and valves as scales. Used pipe market are found everywhere within Niger Delta and builders use them for construction of tank stands. Hence the need for this study on radiometric survey of oil and gas production wastes and its health implication.

2. MATERIALS AND METHODS

2.1 Sample Collection and Preparation

Sixteen samples were collected from four waste streams in six locations as presented in Table 1. Five hundred grams (500 g) of drill cuttings, surface pipe scales and downhole pipe scales each were collected in a black polyethene bag from different locations. Drill cutting (A) and pipe scales (B) were sampled dry, pulverized and homogenized by grinding it into powdery form. The powdered sample was then sieved using a 2 mm sized mesh screen to obtain a fine texture of the samples [14]. The pulverized samples were then oven dried at 110°C until they attained constant weight. Three hundred (300 g) of each dried sample was sealed in radon impermeable cylindrical plastic container. The samples were then stored for 28 days before gamma activity counting, to enable then reach secular radioactive equilibrium. One liter each of produced water and sludges were also collected in a plastic container. Produced water (C) and Sludge (D) were taken with at least 1% air gap left in the container for thermal expansion. Sample containers were rinsed three times with sampled liquid to minimize contamination from the original content of sample container. To prevent adherence of the radionuclide on container walls drops of nitric acid (HNO_3) were added into the samples. After being evaporated, the residues were then transferred into a thoroughly washed and dried 120 ml cylindrical container and hermetically sealed with a plastic tape to ensure air tightness and kept for 28 days to establish secular equilibrium between ^{238}U, ^{232}Th and their daughter products [16]. The samples were counted using High Purity Germanium (HPGe) detector.

2.2 Gamma Spectrometric Analysis

The Gamma Spectrometer System used for evaluation the activity concentration of the radionuclides in the Laboratory is High Purity Germanium (HPGe) detector which comprises Canberra manufactured P-type detector model GC 8023 (Serial Number: 9744) with a pre-amplifier model 2002CSL (Serial Number: 13000742) connected to Multi Channel Analyser (MCA). The detector characteristics include: 78 mm diameter; 69.8 mm length with Resolution (FWHM) of 2.3 KeV of ^{60}Co at 1.33MeV and Relative efficiency of 80%. Canberra Nuclear Genie-2000 software collects and process gamma spectrum into peak widths estimates, for calculating the peak area and subsequent conversion into radionuclide activity. This Gamma Spectrometer System used for this research work is available at the National Institute of Radiation Protection and Research (Nigerian Nuclear Regulatory Authority), University of Ibadan, Ibadan, Nigeria.

Table 1. Sampling location and plan

S/N	Location	Sample code	Sample size	Sample
1	Drilling Rig, U (Top hole 2 and	AU_1	500 g	Drill cuttings/Mud
2	bottom hole 1)	AU2	500 g	Drill cuttings/Mud
3	Drill Cuttings Treatment Facility, V	AV_1	500 g	Drill cuttings/Mud
4		AV_2	500 g	Drill cuttings/Mud
5	Used Pipe Market, W	BW	500 g	Surface Pipe Scales
6	Pipe Inspection Facility, X	BX	500 g	Surface Pipe Scales
7	Well Site, Y	BY	500 g	Downhole Pipe Scales
8		CY_0	1 L	Produced Water
9		CY_1	1 L	Produced Water
10		CY_2	1L	Produced Water
11		DY	1L	Sludge
12	Flow Station, Z	BZ	500 g	Surface Pipe Scales
13		CZ_0	1L	Produced Water
14		CZ_1	1L	Produced Water
15		CZ_2	1L	Produced Water
16		DZ	1L	Sludge

Table 2. Radiological health hazard indices

S/N	Parameter	Models	Sources
1	Air Absorbed dose Rate (D_{air})	D_{air} ($nGyh^{-1}$ =$0.462C_{Ra} + 0.621C_{Th} + 0.0417C_K$	[21,22]
2	Annual Effective Dose Rate	AEDE ($mSvy^{-1}$) = D_{air} x DCF x OF X T	[23]
3	Annual Gonad Effective Dose	AGED($mSvy^{-1}$) = $3.09C_{Ra} + 4.18C_{Th} + 0.314C_k$	[23]
4	Excess Lifetime Cancer Risk	ELCR = AEDE ($mSvy^{-1}$) x DL x RF	[24]
5	Radium Equivalent Activity	Raeq = $C_{Ra} + 1.43C_{Th} + 0.077C_K$	[21]
6	Exposure Rate (ER)	ER (μRh^{-1}) = $1.90 A_{Ra} = 2.82A_{Th} + 0.179C_K$	[25,26]
7	Dose rate (D_R)	DR ($mSvy^{-1}$) = 0.0833 ER	[8]

After the in-growth period of 28 days, each NORM waste sample was subjected to a low background gamma ray spectrometer consisting of Broad energy Germanium detector (GC 8023) manufactured by Canberra industries. As reported by the manufacturers, it has a resolution of 0.5 Kev at 5.9 Kev of ^{55}Fe, 0.75 Kev at 122 Kev of 57Co and 2.2 Kev at 1332 Kev of ^{60}Co respectively. To prevent high background counts due to external radioactive sources, with the intention to reduce the counting time and improve the detection limit, the detector is placed in a low level Canberra model 750 lead shield having a thickness of 10 cm. Furthermore, a multichannel Analyzer (MCA) was used to generate energy distributions of the radioactive samples. In order to obtain a statistical good computational net peak area, each sample was counted for 86400s. The background has been evaluated before running the samples and it was counted for 172800 seconds. ^{226}Ra activity concentration was calculated based on the gamma ray transitions of ^{214}Pb, ^{228}Ra (^{232}Th) concentration was calculated based on the gamma ray transitions of ^{208}Ti and ^{228}Ac while ^{40}K was directly determined using 1460.83 Kev (10.7 %) gamma ray transition [17]. Activity concentration of the radionuclides were computed using the equation [18,19].

$$A\ (Bqkg^{-1}/Bql^{-1}) = \frac{CPS\ X\ 1000}{\varepsilon_\gamma\ X\ I_\gamma\ x\ W} \quad (1)$$

Where A is the specific activity, CPS is the net counts per second for each sample investigated, εγ is the detector photo-peak efficiency at respective gamma-ray peak, Iγ is the corresponding gamma-ray intensity, W is the sample mass in g and 1000 is the mass conversion factor from gram (g) to kilogram (kg).

2.3 Radiological Health Hazard Indices

Eight radiological hazards indices were used to assess the health hazards associated to public or occupational exposure within an irradiated environment [20]. They are presented in Table 2.

2.4 Isotropic Activity Ratios

Among the same series the isotopic activity ratios between a parent and daughter or daughter and another daughter gives the indication if there is an equilibrium or not in that series. The equilibrium is achieved if ^{226}Ra /^{214} Pb = 1 and ^{214}/^{214}Bi =1 for ^{238}U series and ^{212}U / ^{212}Bi =1 for ^{232}Th series [27]. The presence of secular equilibrium for all measured samples are tested by calculating the isotopic activity ratio.

3. RESULTS AND DISCUSSION

3.1 Activity Concentration of Radionuclide in the Oil and Gas Waste Samples

The activity concentration of radionuclides in oil and gas wastes samples determined using Hyper Germanium detector are presented in Table 3 a and b for activity concentration of radionuclides in solid wastes. Table 4a and b presents the activity concentration of radionuclides in liquid wastes while Table 5 presents the radiological parameters. Figs. 1, 2, 3 and 4 are comparison of the activity concentration of ^{40}K, ^{238}U, ^{232}Th and ^{226}Ra with their respective reference levels.

3.2 Radiological Parameters

The radiological health hazard indices from oil and gas wastes is presented in Table 5 for solids and Table 6 for liquids. The radiological hazard indices such as radium equivalent activity (Ra_{eq}), absorbed dose rate (D), annual effective dose equivalent (AEDE), annual gonadal effective dose (AGED), excess lifetime cancer risk (ELCR), external hazard index (H_{ex}) and internal hazard index (H_{in}), exposure rate (ER) and dose rate and its association with ER are calculated from the activity of the nuclides ^{226}Ra, ^{232}Th and ^{40}K using their appropriate models from Table 3.

3.3 Discussion

The lowest and highest activity concentration of ^{226}Ra in drill cuttings, scales, sludge and produced water were 5.28±1.08 and 27.49±3.56 Bqkg^{-1}, 19.53±3.13 and 25727.75±1328.44 Bqkg^{-1}, BDL and BDL and 2.28 ±0.44 Bql^{-1} respectively. The lowest and highest activity concentration of ^{238}U in drill cuttings, scales, sludge and produced water were 3.61±0.76 and 20.19±2.27Bqkg^{-1}, 19.07±2.04 and 23021.73 ±12.3 Bqkg^{-1}, 0.34±0.15 and 1041.58 Bql^{-1} and 0.99±0.26 and 1.74±0.37 Bql^{-1} respectively. The minimum and maximum activity concentration of ^{232}Th in drill cuttings, scales, sludge and produced water were 2.40±0.56 and 12.84±1.85 Bqkg^{-1}, 8.02±1.29 and 21468.25±13.06 Bqkg^{-1}, 0.63±0.17 and 1.06±0.19 Bql^{-1} and 0.65±0.22 and 0.90±0.19 Bql^{-1} respectively while the minimum and maximum activity concentration of ^{40}K in drill cuttings, scales, sludge and produced water were 35.31±2.38 and 116.94±6.73 Bqkg^{-1}, 35.95 ±3.97 and 1527.73±86.6 Bqkg^{-1}, 8.31±0.63 and 13.84±1.00 Bql^{-1} and 9.60±0.66 and 14.20±1.04 Bql^{-1} respectively [28].

The result showed that higher activity concentration values of all the radionuclide were obtained in solid samples (Scales and drill cuttings) than the liquid samples (sludge and produced water). This can be attributed to the fact that salts of the radionuclides readily precipitates from solution below certain temperature and pressure regimes as the well effluent flows to the surface, with such depositions in scales having higher concentration of radionuclides than the solution left.

The top-hole drill cutting (AU_2) shows less presence of radionuclides when compared with down-hole drill cutting ($AU_{1)}$. The least significant among the solid samples are the pipe scales from the Pipe Inspection facility BX, followed by the one for the used Pipe Market BW. The pipes scale, BY and BZ were taken downhole in the well and up-hole within the surface equipment respectively. These results are significantly higher when compared to values between 4.9 to 37.7 Bqkg^{-1} for ^{238}U, 17.5 to 76.7 Bqkg^{-1} for ^{232}Th and 16.3 to 319.0 Bqkg^{-1} for ^{40}K reported by Arogunjo [29]. The results of this work compared well with those obtained from other countries as contained in literatures which was presented in Table 7. The mean activity concentration of ^{226}Ra, ^{238}U, ^{232}Th and ^{40}K obtained in scales and sludge are lower than the mean value of 3822, 87, 1215, 870 and 868 Bqkg^{-1} respectively obtained in Iraq by Abdul Ridha Hussain Subber et al. [28]. Also the activity concentration of the ^{226}Ra obtained from pipe scales 25, 727.75±1,328 Bq/kg from this work was found to be slightly lower than the value obtained in previous work carried out by Parmaksiz et al., [30], with value of 35, 1227.5±19.83 Bqkg-1.

^{226}Ra, ^{238}U, ^{232}Th and ^{40}K activity values obtained in downhole pipe scales were higher than the values obtained in other oil and gas wastes. Radionuclide that dissolved in the brine are separated and radiation levels may vary from

Table 3a. Activity concentration of radionuclides determined from solid waste samples

S/N	Sample code	Activity concentration of solid waste samples (Bq/kg)						^{226}Ra /^{214}Pb	^{214}Pb / ^{214}Bi
		^{214}Bi	^{214}Pb	^{212}Pb	^{208}Tl	^{228}Ac	^{212}Bi		
1	AU1	12.88±2.25	20.19±0.99	17.01±1.51	15.86±2.24	14.79±2.94	16.56±2.54	1.36	1.56
2	AU2	2.90±0.56	2.65±0.64	3.87±0.47	3.79±0.60	4.36±1.75	BDL	1.99	0.91
3	AV1	16.52±1.35	15.16±1.18	16.37±1.18	13.22±1.05	12.99±2.77	BDL	1.66	0.92
4	AV2	11.75±1.80	18.58±1.01	12.82±1.59	15.69±0.81	14.21±2.33	BDL	1.44	1.58
5	BY	18115.9±966.6	25221±829.6	13716.4±834.11	11010.3±473.92	23110.03±1578.81	47518.1±1422.3	1.02	1.39
6	BW	19.69±1.47	21.35±1.28	16.20±1.95	17.69±1.07	18.64±3.56	BDL	1.21	1.08
7	BZ	214.23±20.74	313.21±11.17	270.51±15.44	185.13±11.27	281.07±24.68	262.65±25.34	0.88	1.46
8	BX	6.68±1.06	8.10±0.67	16.19±1.93	11.56±2.13	12.35±2.37	BDL	2.41	1.21

Table 3b. Activity concentration of radionuclides determined from solid waste samples cont.

S/N	Activity concentration of solid waste samples (Bq/kg)					Ra_{eq}
	Sample code	^{226}Ra	^{238}U	^{232}Th	^{40}K	
1	AU1	27.49±3.56	20.19±2.27	12.84±1.85	110.35±6.32	54.35
2	AU2	5.28±1.08	3.61±0.76	2.40±0.56	35.31±2.38	11.43
3	AV1	25.21±2.35	18.96±1.63	8.52±1.00	111.14±6.67	45.95
4	AV2	26.89±3.31	19.07±2.04	8.54±0.95	116.94±6.73	48.11
5	BY	25727.75±1328.44	23021.73±1041.58	21468.25±1125.57	1527.73±86.60	56544.9
6	BW	26.00±4.07	22.35±2.27	10.51±1.32	130.22±7.75	51.06
7	BZ	277.47±24.42	268.30±18.78	199.87±15.35	35.95±3.97	566.05
8	BX	19.53±3.13	11.44±1.62	8.02±1.29	74.01±4.74	36.70
	UNSCEAR (2000)	35.00	35.00	30.00	400.00	

Table 4a. Activity concentration of radionuclides determined from liquid waste samples

S/N	Sample	Activity Concentration of Liquid Waste Samples (Bq/l)						^{226}Ra /^{214}Pb	^{214}Pb / ^{214}Bi
		^{214}Bi	^{214}Pb	^{212}Pb	^{208}Tl	^{228}Ac	^{212}Bi		
1	DZ	1.33±0.43	1.57±0.43	1.24±0.27	0.98±0.39	0.92±0.20	BDL	0	1.18
2	DY	0.59±0.12	0.43±0.34	1.14±0.26	1.63±0.41	2.22±0.28	BDL	0	0.72
3	CY_1	1.75±0.44	1.57±0.40	0.86±0.26	2.74±0.35	1.71±0.28	BDL	0	0.89
4	CY_2	1.70±0.35	1.27±0.44	1.16±0.23	1.09±0.37	1.02±0.52	BDL	0	0.74
5	CZ1	1.46±0.18	1.49±0.50	0.866±0.13	1.82±0.38	1.11±0.25	BDL	1.53	1.02
6	CZ2	1.61±0.44	1.51±0.45	1.27±0.31	1.51±0.34	1.711±0.29	BDL	0	0.93
7	CZ0	BDL	BDL	BDL	BDL	BDL	BDL	0	0
8	CY0	BDL	BDL	BDL	BDL	BDL	BDL		0

Table 4b. Activity concentration of radionuclides determined from liquid waste samples cont.

S/N	Activity Concentration of Liquid Waste Samples (Bq/l)					Ra_{eq}
	Location	^{226}Ra	^{238}U	^{232}Th	^{40}K	
1	DZ	ND	0.97±0.29	0.63±0.17	8.31±0.68	2.51
2	DY	ND	0.34±0.15	1.00±0.19	11.74±0.94	2.67
3	CY1	ND	1.11±0.28	1.06±0.18	13.84±1.00	3.69
4	CY2	ND	0.99±0.26	0.65±0.22	11.86±0.89	2.84
5	CZ1	2.28±0.44	1.74±0.37	0.76±0.15	14.20±1.04	3.92
6	CZ2	ND	1.04±0.30	0.90±0.19	9.60±0.66	3.06
	UNSCEAR	10.00	10.00	1.00		

Table 5. Radiological hazard indices from the oil and gas solid waste samples

S/N	Location	D_{air} ($nGyh^{-1}$	AEDE $mSvy^{-1}$	AGED $mSvy^{-1}$	ELCR X 10^{-3}	H_{ex}	H_{in}	ER μRh^{-1}	DR $mSvyr^{-1}$
1	AU1	25.28	0.039	173.27	0.135	0.146	0.221	108.19	1.68
2	AU2	5.40	0.008	37.43	0.029	0.031	0.045	23.12	0.30
3	AV1	21.57	0.033	148.41	0.116	0.125	0.192	91.82	1.58
4	AV2	22.60	0.035	155.51	0.121	0.130	0.203	96.11	1.59
5	BY	25281.7	38.76	169715.7	135.65	152.74	222.2	109696.7	1917.71
6	BW	23.96	0.037	165.16	0.129	0.138	0.208	102.35	1.86
7	BZ	253.81	0.389	1704.13	1.362	1.529	2.279	1097.26	22.35
8	BX	17.0895	0.026	117.11	0.092	0.099	0.152	72.97	0.95
	Standard	57	1	300	0.029	1	1	600	

Table 6. Radiological hazard indices from the oil and gas liquid waste samples

S/N	Location	D ($nGyh^{-1}$)	AEDE ($mSvy^{-1}$)	AGED (μSvy^{-1})	ELCR x10^{-3}	Hex	Hin	ER μRh^{-1}	DR $mSvy^{-1}$
1	DZ	1.185	0.001	8.232	0.005	0.002	0.005	3.26	0.27
2	DY	1.266	0.002	8.909	0.005	0.03	0.005	4.92	0.41
3	CY1	1.749	0.002	12.215	0.008	0.003	0.007	5.47	0.46
4	CY2	1.358	0.002	9.517	0.006	0.002	0.005	3.96	0.33
5	CZ1	1.867	0.002	13.008	0.008	0.011	0.020	46.92	3.91
6	CZ2	1.438	0.002	9.982	0.006	0.003	0.006	4.26	0.35
	Mean	1.477	0.002	10.310	0.006	0.004	0.008	5.14	0.43
	standard	57	1	300	0.29	1	1		

Table 7. Activity concentration s of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K in scales and sludge collected from different studies [28]

Country	Material	^{238}U kBq/kg	^{226}Ra kBq/kg	^{228}Ra kBq/kg	^{232}Th kBq/kg	^{40}K kBq/kg
Brazil	scales	-	19.1-323.0	-	-	-
Brazil	sludge	-	0.36-367.0	4.0 – 23.5	-	-
Algeria	scale	-	1.0-950	-	-	-
Algeria	sludge	-	0.1 -0.393	-	-	
Tunisia	scale	-	4.3 -658	-	-	
Norway	sludge	-	0.3-32.3	0.3-33.5	-	
UK	Scale	-	0.1-270	0.2-180	-	
UK	Sludge	-	5.0-50.0	1 -170	-	
Oman	sludge	-	1.1-5.67	0.47-0.92	0.11-0.61	
EUA	Sludge	-	15000			
EUA	Scale	-	26000			
USA	Scale	-	10^4-10^6			
USA	Sludge	-	Up to 10^6			
Iraq	Sludge/scale	0.0.202	0.125-96.5	0.05-9.81	0.01-11.45	0.04-56.3
Nigeria (This study)	Drill cuttings	0.004-0.02	0.005-0.03	-	0.002-0.013	0.04-0.12
	-Scale	0.02 -23.02	0.02-25.73	-	0.01-21.5	0.04- 1.53
	-sludge	0 -1.04	BDL	-	0.63-1.06	8.31-13.84
	- produce water	0.99-1.74	BDL – 2.28	-	0.65-0.90	9.6 -14.2

**All the activity concentrations of all the radionuclide in produced water is in Bql^{-1}*

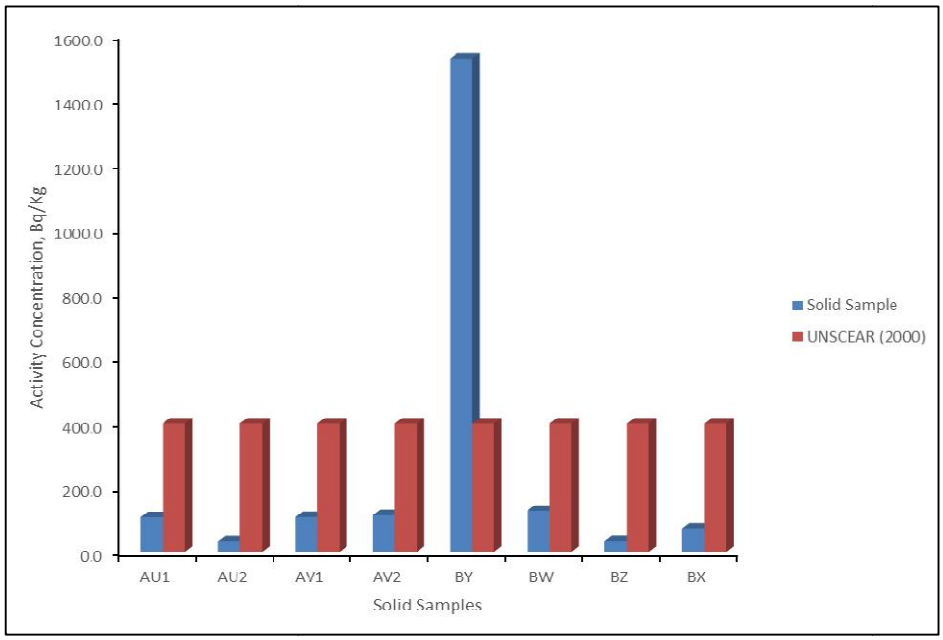

Fig. 1. ^{40}K activity concentration in solid oil and gas waste samples

background soil level of more than 4.0 Bqkg^{-1}. This leads to concentration of radionuclide salt precipitates deposited as scales at certain temperature and pressure zones along the downhole pipe. Scales are normally found on the inside of piping and tubing. American Petroleum Institute, found that the highest concentration of radionuclides are in the scale in well head piping and in production piping near the well head. Although the concentration of NORM is lower in sludges than in scales, sludges are more soluble and therefore more readily released to the environment. As a result, they pose a higher risk of exposure [31].

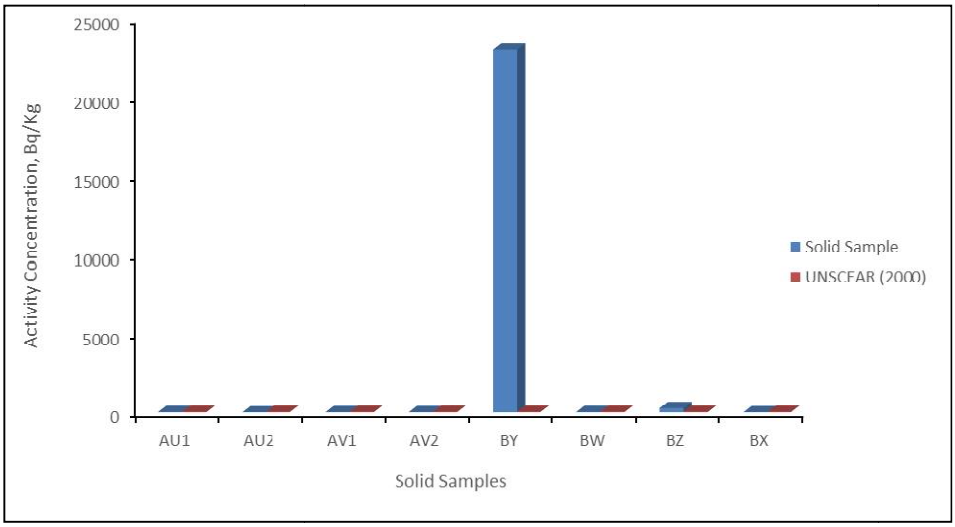

Fig. 2. ^{238}U activity concentration in solid oil and gas waste samples

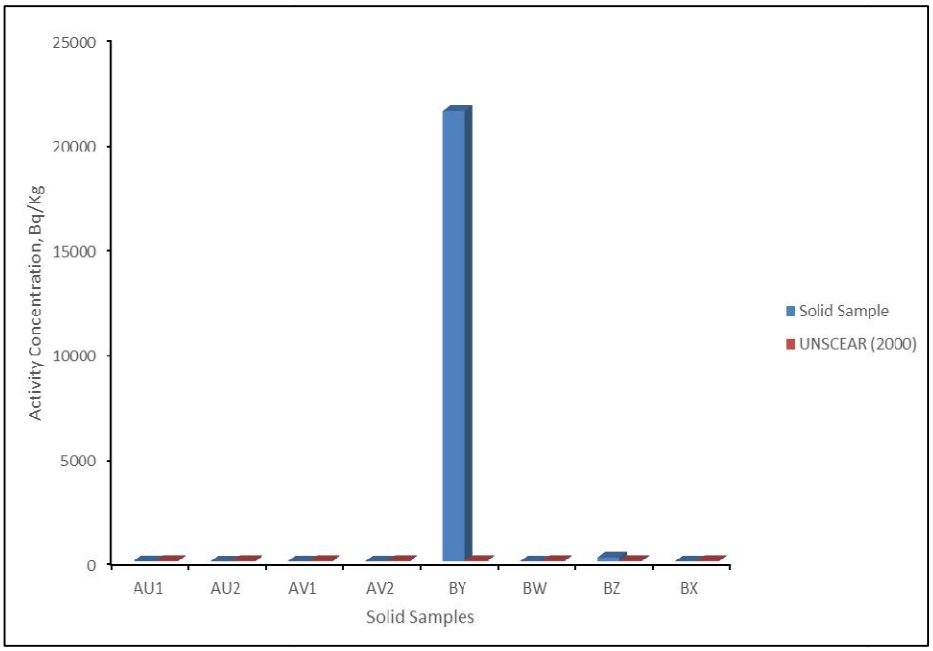

Fig. 3. ^{232}Th activity concentration in solid oil and gas waste samples

It is evident that all the radionuclides found in downhole scales exceeded their recommended values as seen in the figures while they are below the permissible level in other samples.

The study showed that the activity concentration of ^{40}K is lower than reported value of 48.78±13.67 Bq/l and ^{226}Ra which compares favorably with the reported value of 6.04±2.48 Bq/l from gamma spectroscopy analysis of produced water from selected flow stations in Delta state by Avwiri et al., [32]. The result showed that produced water from the Flow Station is radiologically contaminated. Their discharged into water bodies and waste pits could lead to contaminations of aquifer used for drinking within the locality. Agbalagba et al. [33], while the estimation of the dose rate to man in surface and ground water from western Niger Delta argued that even though there may be no immediate effect from intake of the contaminated water, there exist a high probability of long term cancerous effect to the impacted communities.

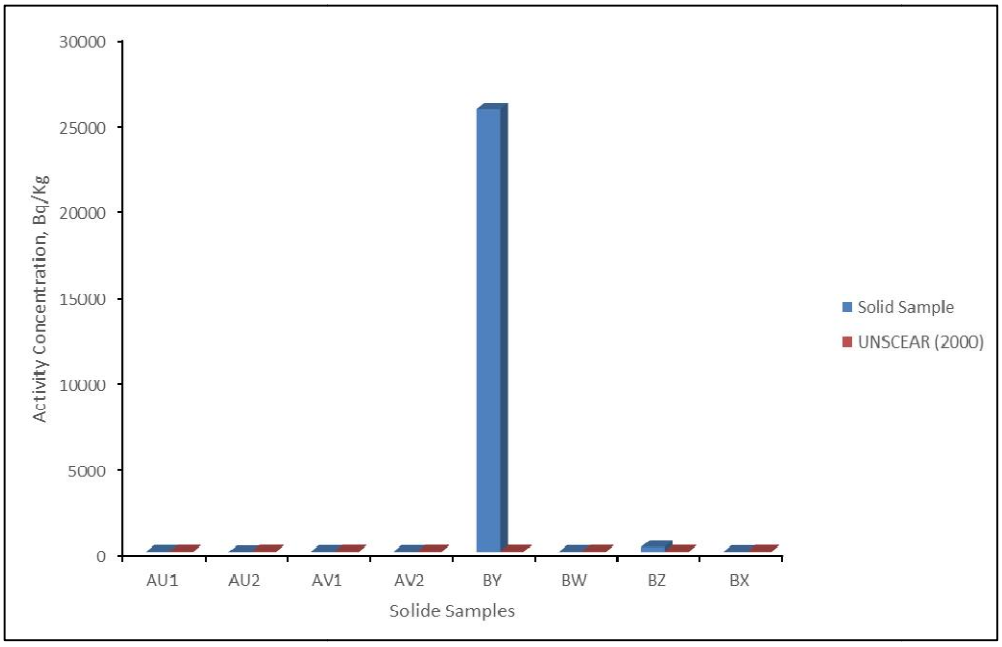

Fig. 4. ^{226}Ra activity concentration in solid oil and gas waste samples

The radiological health hazard indicators for oil and gas liquid wastes samples mostly were lower than reference values. The computed radiological health hazard indices due to the exposure from the analyzed oil and gas solid waste samples as presented in Table 7. The result showed unusually very high values of all indices for downhole pipe samples when compared with their reference values. Typically, the AEDE due to the downhole pipe scale computed was 38.76 mSvy^{-1}, significantly above the universally recommended dose of 1 mSvy^{-1} for non-industrial workers and the public. The mean values of all the radiological health risk parameters (AGDE, AEDE, ELCR,) are higher than ICRP (300 mSvy^{-1}, 1.0 mSvy^{-1} and 0.29 x $10^{-3,}$ values. H_{in} recorded mean value of 28.19 which is much higher than the reference level of 1. H_{in} is the cause of harmful effects to the lungs due to the internal contact of α- particles of a higher ionization power to the sensitive tissues of the lungs and other parts of the respiratory system [23].

The exposure rate (ER) and dose rate (DR) estimated in the solid and liquid waste samples were presented in Table 7. ER ranged from 23.12 to 109696.7 μRh^{-1} with mean value of 13911.06 μRh^{-1} and DR ranged from 0.30 to 1917.71 mSvy^{-1} with mean value of 243.50 mSvy^{-1}. The mean value of ER was higher than the reference level of 600 μRh^{-1} [25] and also DR mean value higher than the reference value of 50 mSvy^{-1}. All the mean values of ER and DR in liquid samples are lower than their reference values. The isotropic activity ratios between parent and a daughter give the indication that there was an equilibrium between the two before the counting was done. The result showed that ^{226}Ra/^{214}Pb = 1, ^{214}Pb/^{214}Bi = 1 for ^{238}U series and ^{212}Pb/^{212}Bi =1 for ^{232}Th series except in scales from downhole which was greater than 1 as presented in Table 2.

4. CONCLUSION

The oil and gas sludge, scales (up hole and downhole), drill cuttings and produced water were successfully analyzed for radioactive elements using hyper germanium detector. The result clearly show that the radioactivity levels in both solid and liquid wastes exceeded the limits set by EPA but within the range found in nearby countries. Nevertheless the health burden due to natural background radiation from oil and gas production wastes on the marketers of the pipes, workers and inhabitants of the areas is high and hence carries significant health hazards.

The test for equilibrium state of the parents' nuclide with the daughters through the use of isotropic ratios showed positive results since all the results was approximately equal to unity except for samples from downhole pipes which

had value greater than unity. The estimated radiation exposure rate (ER) was also higher than the recommended value of 600 μRh^{-1}. The result showed symmetrical distribution of all the studied radionuclides. The result of this work revealed that downhole and surface pipes from oilfield commonly used in the construction of buildings and domestic overhead tank-stands are associated with high levels of ionizing radiation that may be unsafe to people and the environment.

We conclude therefore that if no remedial steps are implemented, there will be long-term risks to health of the workers in the field, waste pipe sellers and inhabitants of the area. Unfortunately, there are no standard methods for the clearance or disposal of such waste and more effort are required to set up such methods.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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