

Evaluation of Gross Alpha and Gross Beta Radioactivity in Crude Oil Polluted Soil, Sediment and Water in the Niger Delta Region of Nigeria

Ezekiel O. Agbalagba¹, Stephen U. Egarievwe² , Eunice A. Odesiri-Eruteyan³,
Mebougna L. Drabo⁴

¹Department of Physics, Federal University of Petroleum Resources, Effurun, Nigeria

²Nuclear Engineering and Radiological Science Center, Alabama A&M University, Huntsville, AL, USA

³Department of Environmental Management and Toxicology, Federal University of Petroleum Resources, Effurun, Nigeria

⁴Department of Mechanical Engineering, Alabama A&M University, Huntsville, AL, USA

Email: agbalagba.ezekiel@fupre.edu.ng

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Abstract

The evaluation of gross alpha and beta activities in crude oil contaminated soil, sediment and water samples was conducted in ten oil polluted environment of Delta State using Gas-flow proportional counter. Samples were collected from the oil polluted environment in each oil field and samples were prepared and analyzed following standard procedures. The mean gross alpha and beta activities obtained are 331.4 ± 24.5 Bq kg⁻¹ and $11,335 \pm 112$ Bq kg⁻¹ respectively for soil, 259.2 ± 17.6 Bq kg⁻¹ and 4508 ± 96 Bq kg⁻¹ respectively for sediment, and 1.00 ± 0.09 Bq kg⁻¹ and 20.3 ± 1.7 Bq kg⁻¹ respectively for water. The estimated average values of the total annual effective dose equivalent ($AEDE_{T(\alpha,\beta)}$), the total annual gonadal dose equivalent ($AGDE_{T(\alpha,\beta)}$), and the total excess lifetime cancer risk ($ELCR_{T(\alpha,\beta)}$) are 10.64 mSv y⁻¹, 0.037 μSv y⁻¹ and 0.037 μSv y⁻¹ respectively. The gross alpha and beta activities values obtained in soil and sediment were relatively high compared to values reported in some parts of the country and other regions and countries of the world. The radiological risk parameters examined show that $AEDE_{T(\alpha,\beta)}$ and $AGDE_{T(\alpha,\beta)}$ are above recommended permissible limits while $ELCR_{T(\alpha,\beta)}$ is within the recommended permissible limit. The overall results obtained in this study indicate that the environmental samples have been radiologically impaired due to the crude oil spillage. An appropriate remediation technique was therefore recommended to remediate the polluted soil, sediment, and water to their near original state.

Keywords

Alpha Activity, Beta Activity, Crude Oil, Sediment, Soil, Water

1. Introduction

The radiological contamination of the human environment always results in the elevation of natural background ionizing radiation. The radiological hazards to man and the environment from radioactive contamination depend on the nature of the radioactive contaminants, the degree of contamination and the level of the spread of the contamination [1]. Human activities such as mining, milling and processing of uranium ores, and mineral sands, smelting of metalliferous ores, manufacture of fertilizers, drilling of oil and gas, transportation, processing and burning of fossil fuels have raised the concentration of naturally occurring radioactive materials in the environment [1] [2] [3] [4]. Researchers in recent time have focused on the assessment of gross activities in soil, sediments and water, to ensure that the Reference Dose Level (RDL) of 1.0 mSv y^{-1} for soil and sediments and the committed effective dose of 0.1 mSv y^{-1} for consumption of drinking water is not exceeded in an environment [5] [6]. For water, the RDL of 0.1 mSv is equal to 10% of the dose limit for members of the public as recommended by the International Commission for Radiological Protection (ICRP) [7] and the International Basic Safety Standard by IAEA [8], and is acceptable to most World Health Organization (WHO) member States, European Commission, Food and Agriculture Organization [9] [10].

Gross alpha activity concentration in soil and sediment samples is defined as the total radioactivity of all alpha emitters in the samples. The value of gross activity originating from these alpha emitters in soil and sediment depends on the geological formation of the area, the concentration of mineral component and the nature of human activities in the area [1]. Alpha emitters mixed with ground or surface water percolating through soil and sediment samples have contributed to the increased concentrations of gross alpha in drinking water samples [10]. The presence of gross beta radioactivity in soil and sediment is due to the natural long-lived isotopes ^{40}K , ^{210}Pb and ^{228}Ra [11] [12]. Alpha and beta radiations are less penetrating unlike γ -ray that has the highest penetrating power, but the effects of alpha and beta particles within the body either through inhalation or ingestion are far more detrimental because of their ionizing power [1].

Crude oil, being particulate in nature, when spilled into the environment, can settle on farmlands, farm crops and in the communities' sources of water and can as well be inhaled continuously. Crops grown in such an oil spilled environment could absorb the radioactive content from the crude oil in the soil or through the leaves, while sea foods/aquatic organisms or drinkable water could also be radioactive to a level that could be harmful to man. When these contaminated crops and aquatic animals are eaten by man, radioactive elements get

into the body and could attain a detrimental level depending on the type of radionuclides involved, the rate of consumption and the extent to which the food/water has been contaminated. These have manifested in the organs of man as kidney or lung cancer, cataract, leukemia, and other radioactive induced sicknesses. This, no doubt, is a major environmental challenge in the oil producing communities of the Niger Delta region, especially now that a new set of militant groups has resurfaced in the region, destroying pipes carrying crude oil and gas and causing great oil spillage on land and water. Petroleum, a naturally occurring liquid mineral resource, is deposited beneath the earth surface, and its occurrence is sometimes accompanied with the existence of natural gas. The oil, gas and associated gas are generally contaminated with radionuclide in the earth crust. All these provide the source of radiation of α , β and γ often found in the petroleum matrix [13] [14] [15]. High concentrations of naturally occurring radionuclide materials (NORMs) in water, sediment and soil can be harmful to land and marine animals and plants, and they can upset delicate ecological balances and contaminate food sources [16]. These occur as a result of oil spillage into land, failure in underground crude or gas pipelines, vandalizing of crude oil pipes, and road accident by crude oil transporting trucks or vehicles. Studies have shown that almost all the elements in the periodic table including heavy metals (radioisotopes) are found in the crude oil matrix [14] [16] [17]. Thus, the release of gas through flaring, oil spillage on land and water bodies and its derivative may have serious radiological and hazardous effects on man and direct impact on the soil and water [18].

The removal of radioactivity from soil, sediment and water when present above ambient level is gamine to environmental sustainability and human health protection. Natural processes that result in the removal of crude oil from the natural environment include evaporation, oxidation and biodegradation [19]. Evaporation, oxidation and biodegradation are some known methods that can naturally remediate crude oil polluted water, soil and sediment [20] [21] [22]. These constitute the natural remediation web that can reduce the toxic (radioactive) content of crude oil spill into any ecosystem. Thus, the natural remediation processes with the passage of time on the study area will be examined.

The main objective of this study is to evaluate the gross alpha and beta activity concentration in soil, sediment and water in crude oil spilled environment and its natural remediation effect with passage of time on the gross alpha and beta radioactivity concentration in spilled/environment. This is to ascertain the safety of drinking water, the suitability of soil for farming and sediments for optimal use as building material in the study area. The health implications of living and of drinking water in the environment and working within the study locations will be examined.

2. Experimental Methods

2.1. Description of Study Area

The study was conducted over a period of six years between 2009 and 2014

within Delta State of Nigeria. This is well above the four years of study in previous research. This is to give round for wider range of study time and for comprehensive data acquisition for good inference. Ten crude oil polluted sites were identified, and samples of soil, sediment/sludge and water were each collected from the ten locations (oil producing field). The study area lies within latitude 5°18"N and 5°68"N and longitude 5°33"E and 6°40"E Mid-West of the Niger Delta region of Nigeria. The geology of the study area has been reported elsewhere [23].

2.2. Sample Collection and Preparation Techniques

Twelve samples each were collected for soil, sediment/sludge and water in each of the ten crude oil polluted environment (oil fields). Collection of samples was carried out twice a year (wet and dry season) in all the sampling sites for the period of six years. One sample each of soil, sediment/sludge and water from non-oil polluted site from the same environment was collected as control sample.

The bulk soil samples (stones, vegetation and organic debris removed) were collected each in a black polythene bag at a depth between 0 to 15 cm (topsoil), which represent the soil permeability to crude oil spillage and particle settlement depth. Sediment samples were collected at the bottom of waste pit (sludge) and water beds (sediment) of crude oil polluted water bodies, using a steel hand geological auger. The geological auger was first cleaned with acid, detergent and rinsed with tap water. Samples were collected into new aluminum foil labeled and placed in black polythene bags, while the water content in sediment samples was separated from samples by decantation and filtration. Samples were later kept in a slow-air flow, low temperature (50°C) drying cabinet to help accelerate the drying process without loss of radionuclides from the samples [8]. The dried samples were grinded with mortar and pestle and then allowed to pass through a 100-mesh sieve. The prepared samples were then sent for pelleting into counting planchet geometry using the hydraulic compressor machine. Pelleted samples were then kept in desiccators and stored in the laboratory before counting.

The water samples were collected in oil polluted rivers/streams and drinking water in the host communities, using the grab sampling techniques. Samples were collected into 2-liters plastic containers after rinsing containers thrice with sample water to minimize contamination, and about 1% air space left for thermal expansion [6]. The water samples collected were acidified at point of collection with 20 ml \pm 1 ml of nitric acid per liter to minimize the absorption of radionuclides into the walls of the containers {International Standard Organization (ISO), 9697 & 9698} [24]. The samples were then tightly covered with container lids and kept in the laboratory until analysis. At the laboratory samples were slowly evaporated (avoid boiling) in a furnace temperature at 60°C down to a 50 ml volume. The residues were transferred quantitatively to stainless-steel planchet and dried. Samples were then allowed to equilibrate with ambient tem-

perature and then weighed.

The counting or screening time of samples was 30,000 s for gross alpha and beta activities. Further detailed procedures for the actual counting technique to determine the gross alpha and beta radioactivity in the samples are as reported by Avwiri and Agbalagba [6], which are in accordance with ISO, 9697 & 9698(E) guidelines [24].

2.3. Counting Equipment

The counting system employed in the analysis was the gas-flow proportional counter {Eurisys Measure IN20 low-background multiple (eight) channel counter} at Centre for Energy Research and Training (CERT) Material Laboratory, Ahmadu Bello University Zaria (ABU) Nigeria. Each channel of the counter has a $450 \mu\text{g}\cdot\text{cm}^{-3}$ window thickness and is 60 mm of diameter. The chambers were covered with lead whose thickness can be varied. The detectors were operated within a background radiation environment of $<101 \mu\text{rad h}^{-1}$. The system was connected to a microprocessor loaded with a spreadsheet program (Quattro-Pro) and graphic program (Multiplan). The system was operated at a bias voltage (~ 1100 V with P10 gas: argon-methane of 10%) where only alpha particles were detected, which is commonly referred to as “alpha-only” mode. But when the bias voltage was increased to ~ 1650 V with same gas, the counter responded to both alpha and beta particles simultaneously. Operation at this higher voltage was referred to as “simultaneous” or “alpha+ beta” mode [9].

2.4. Detector Calibration

The alpha standard was ^{239}Pu with half-life of 24,110 years, while the beta standard was ^{90}Sr with half-life of 28 years. These standards were certified by CERCA LEA Laboratories in France with certification numbers CT 001/1285/001920-1927 and CT 1271/00/1778-1783, respectively.

Plateau test was run with the manufacturer’s calibration standards (^{239}Pu and ^{90}Sr) whose activities ranged from 133.29 to 185 51 Bq and 92.31 to 103.68 Bq respectively in all the three operating modes which was run for 1800 s for five cycles. The result of the efficiency calibration indicated an average channel efficiency of 35.5% for the alpha counts in the alpha-only mode, 40.0% for the beta counts in beta-only mode, while the average efficiency in alpha/beta was 22.8%. The alpha-only and beta-only mode of counting was more efficient and therefore was employed in the final counting stage.

The background radioactivity counting procedure for the counting of water, soil and sediment samples followed as reported elsewhere [6] [9] [25]. The results of background activity indicated reproducibility in the channels of the counter. The results also gave an average background radiation activity of 0.17 Bq for alpha in alpha-only mode, and 1.13 Bq for beta in beta-only mode. The results gave an average efficiency value greater than 30%. The guidance level by WHO for drinking water is 0.5 Bq l^{-1} for gross alpha activity and that for gross

beta activity is 1 Bq l^{-1} [26]. These values obtained are good representative of the environment and could therefore be employed in the subsequent measurement of the soil, sediment and water samples.

2.4.1. Counting of Samples

The counting equipment was automated; the procedure involved entering the present time, number of cycles and the counting (operational) voltage. Also, the counter characteristics (channel efficiency and background count rate), volume of sample used, and sample efficiency were entered. The sample efficiency was calculated as:

$$\text{Sample Efficiency} = \frac{M_T}{0.1A} \times 100\% \quad (1)$$

where M_T is the mass of pelleted sample gotten from sample preparation and $0.1A$ (mg) is the expected mass in the planchet.

2.4.2. Gross Alpha Counting

For gross alpha counting, the high voltage was set at 1650 V and samples were counted for 13 cycles of 180secs per cycle. The results were displayed as raw counts; count rate (count/min), activity and standard deviation. The data were acquired for alpha only mode and the alpha count rate R_α as well as alpha activity A_α was calculated using the formula:

$$R_\alpha = \frac{\text{Raw } \alpha \text{ count}}{\text{Count time}} \quad (2)$$

$$A_\alpha = \frac{(R_\alpha \text{ background}) \times (\text{unit coefficient})}{(\text{channel } \alpha \text{ efficiency}) \times (\text{sample efficiency}) \times (\text{sample volume})} \quad (3)$$

where unit coefficient is multiplication coefficient making it possible to obtain the results expressed in the units used by the operator. The alpha activity is expressed as activity concentration C_α in Becquerel per liter or (Bq kg^{-1}) for water and soil/sediment respectively. The activity concentration C_α is calculated using the formula ISO 9696:1992 (E) [24].

$$C_\alpha = \frac{R_b - (R_0 \times a_s \times m)}{R_s - (R_0 \times 1000 \times V)} \quad (4)$$

where R_b is the observed sample count rate (s^{-1}), R_0 is the background count rate (s^{-1}), R_s is the observed standard count rate (s^{-1}), a_s is the specific activity of the alpha standard, V is the volume of the sample soil used.

2.4.3. Gross Beta Counting

The high voltage for gross beta counting was set at 1700 V and samples were counted for 25 cycles of 180 seconds per cycle in beta only mode. The count rate and the activity were calculated using the formula.

$$R_\beta = \frac{\text{Raw } \beta \text{ count}}{\text{Count time}} \quad (5)$$

$$A_{\beta} = \frac{(R_{\beta} \text{ background}) \times (\text{unit coefficient})}{(\text{channel } \beta \text{ coefficient}) \times (\text{sample efficiency}) \times (\text{sample volume})} \quad (6)$$

The gross beta activity is expressed as activity concentration C_{β} in Bq kg⁻¹ and Bq l⁻¹ calculated as ISO 9697:1992(E) [27]

$$C_{\beta} = \frac{R_b - (R_0 \times 14.4 \times m)}{R_s - (R_0 \times 1000 \times V)} \quad (7)$$

where 14.4/1000 represents the specific activity of ⁴⁰K. All other terms have their usual meaning. The standard deviation σ associated with the activity of the samples collected for background was calculated using ISO 9697:1992 (E) [27]

$$\sigma = \sqrt{\frac{R_b}{t_b} + \frac{R_0}{t_0}} \times \frac{14.4 \times m}{1000 \times v \times (R_b - R_0)} \quad (8)$$

3. Results and Discussion

3.1. Gross Alpha and Gross Beta Activity in Soil Sediment

The results of the analysis for gross alpha activity concentration in the soil, sediment and water samples are presented in **Table 1**, while **Table 2** presents the corresponding gross beta activity. The average alpha activity concentration in the studied oil contaminated soil samples ranged from 24.0 ± 1.0 Bq kg⁻¹ in Oweh field to 925.6 ± 45.3 Bq kg⁻¹ in Kokori field with a mean value of 331.4 ± 24.5 Bq kg⁻¹.

The beta activity concentration values ranged from 2330 ± 98 Bq kg⁻¹ in Uzere field to 46,150 ± 217 Bq kg⁻¹ in Olomoro field with a mean value of 11,335 ± 112 Bq kg⁻¹ and a control gross alpha and beta activities of 43.3 ± 13.4 Bq kg⁻¹ and 1660 ± 13.4 Bq kg⁻¹ respectively. The average gross alpha activity concentration in the sediment/sludge samples in the study locations ranged from 32.5 ± 5.6 Bq kg⁻¹ in Oweh oil field to 620.8 ± 45.5 Bq kg⁻¹ in Ughelli East (Eruemukohwarien) field with a mean value of 259.2 ± 17.6 Bq kg⁻¹ and a control gross alpha value of 30.1 ± 9 Bq kg⁻¹. The corresponding sediment/sludge beta activity concentration ranged from 1500 ± 61 Bq kg⁻¹ in Olomoro/Oleh oil field samples to 11,911 ± 169 Bq kg⁻¹ in Agbarha oil field with a mean activity value of 4508 ± 96 Bq kg⁻¹ and a control activity value of 30.1 ± 9.6 Bq kg⁻¹.

The high gross alpha and beta activity concentration observed in soil samples of Kokori, Olomoro and Ughelli West (Ekakpamre) oil spilled locations may be attributed to high radium (radon) concentration and the presence of ²¹⁰Pb and ²²⁸Ra, in the spilled crude oil samples on the soil. It may also be attributed to non-cleanup of the spilled crude oil observed throughout the study period in these sampling sites. This is an indication that the crude oil from these locations will be of high leaded Brent crude from the reported beta activities.

The relatively high alpha activity level in sediment samples in Uzere, Ewreni and Ughelli West study locations may be attributed to the transport phenomena of the affected river waters and the settlement and accumulation of the crude

Table 1. Summary of alpha activity concentration for the different soil, sediment and water samples in the study area.

Sample Location	GPS Coordinate	Soil Sample (Bq kg ⁻¹)		Sediment Sample (Bq kg ⁻¹)		Water Sample (Bq l ⁻¹)	
		Range	Average	Range	Average	Range	Average
Uzere West & East	05°25.57" 06°13.09"	50.2 - 65.7	53.3 ± 9.6	235.1 - 1380.0	620.7 ± 36.9	0.06 - 35.1	7.10 ± 0.60
Olomoro/Oleh	05°28.98" 06°08.37"	85.0 - 1250.8	715.0 ± 27.8	34.6 - 81.1	59.4 ± 8.3	0.70 - 1.70	1.10 ± 0.20
Oweh	05°29.55" 06°07.32"	21.4 - 34.0	124.0 ± 1.0	19.0 - 35.6	32.5 ± 5.6	0.07 - 0.40	0.20 ± 0.04
Evwreni	05°22.42" 06°02.32"	98.3 - 220.5	138.0 ± 21.7	326.2 - 734.1	510.1 ± 9.0	0.02 - 1.20	0.70 ± 0.05
Agbarha	05°32.19" 06°02.33"	44.1 - 100.4	72.6 ± 9.7	13.5 - 58.6	38.0 ± 5.0	0.01 - 0.10	0.07 ± 0.00
Kokori	05°39.17" 06°04.25"	97.2 - 1695.0	925.6 ± 45.3	116.4 - 320.6	250.3 ± 23.0	0.02 - 1.40	0.60 ± 0.02
Afiesere	05°32.86" 06°00.81"	58.8 - 66.4	61.1 ± 8.4	42.6 - 129.4	79.4 ± 8.0	0.02 - 0.10	0.04 ± 0.00
Ughelli East	05°30.97" 06°55.01"	99.1 - 610.1	328.8 ± 27.2	518.6 - 924.2	620.8 ± 45.5	0.02 - 0.05	0.03 ± 0.00
Ughelli West	05°32.29" 06°53.68"	680.9 - 960.7	776.4 ± 61.6	143.9 - 370.4	290.4 ± 19.0	0.01 - 0.50	0.09 ± 0.01
Otorogu	05°25.03" 06°53.09"	47.4 - 390.3	218.9 ± 32.3	21.3 - 34.6	90.3 ± 9.8	0.03 - 0.10	0.05 ± 0.00
Mean		129.5 - 372.1	331.4 ± 24.5	147.1 - 406.9	259.2 ± 17.6	0.10 - 4.00	1.00 ± 0.09
Control		39.4 - 46.0	43.3 ± 13.4	26.2 - 33.7	30.1 ± 9.4	ND - 0.03	0.01 ± 0.00

ND = Not Detected, with detection limit of 0.5 Bq kg⁻¹ soil and sediment and 0.001 Bq l⁻¹ for water.

particles in these rivers water beds and bottom pits. It was observed that all the sediment sample sites with high alpha activities are unperturbed sites while the sites with relative human activities are low in alpha activities. The elevated beta activity level sites of: Uzere, Agharha and Ughelli West (Ekakpamre) were crude oil waste pits sludge. The results obtained in both wet and dry seasons for the six years of the study showed no significant variation in gross alpha and gross beta activity concentration with time.

A comparison of the average activity concentration values obtained in the soil and sediment samples with those obtained at the control, revealed that the crude oil spillage into the soil has elevated the gross alpha activity levels of the soil in the order of 7.7 times magnitude while beta activity is 6.8 times magnitude higher than the value of the control site. In the sediment samples, the alpha activity levels is 8.6 times magnitude and beta activity is 9.6 times magnitude higher than the control samples result for sediment. The result shows that the crude oil spilled environment (soil) has been polluted radiologically. These mean alpha activities values obtained in soil and sediment in this study compare favorably with the alpha activity value of 530 ± 20 Bq kg⁻¹ obtained in similar crude oil polluted environment of Imirigin oil field in Bayelsa State and the

Table 2. Summary of beta activity concentration for the different soil, sediment and water samples in the study area.

Sample Location	GPS Coordinate	Soil Sample (Bq kg ⁻¹)		Sediment Sample (Bq kg ⁻¹)		Water Sample (Bq l ⁻¹)	
		Range	Average	Range	Average	Range	Average
Uzere West & East	05°25.57" 06°13.09"	1960 - 3990	2330 ± 98	2301 - 5230	3480 ± 70	11.9 - 151.2	67.1 ± 4.6
Olomoro/Oleh	05°28.98" 06°08.37"	2060 - 94,910	46,150 ± 217	752 - 2581	1500 ± 61	1.1 - 15.8	5.9 ± 0.3
Oweh	05°29.55" 06°07.32"	500 - 14,340	6124 ± 77	2292 - 5012	3310 ± 102	1.6 - 16.2	6.1 ± 0.4
Evwreni	05°22.42" 06°02.32"	7960 - 15,220	8138 ± 81	3642 - 7316	5840 ± 111	4.1 - 54.7	36.7 ± 3.5
Agbarha	05°32.19" 06°02.33"	1520 - 6430	3101 ± 67	9132 - 15,666	11,911 ± 169	0.9 - 3.0	1.9 ± 0.3
Kokori	05°39.17" 06°04.25"	11360 - 59,540	31,925 ± 35.3	6114 - 10,260	7450 ± 99	7.4 - 135.9	58.7 ± 5.6
Afiesere	05°32.86" 06°00.81"	2170 - 3120	2468 ± 64	2461 - 4529	3840 ± 79	1.0 - 5.1	3.2 ± 0.1
Ughelli East	05°30.97" 06°55.01"	2920 - 3992	3128 ± 27	1674 - 3249	2431 ± 64	2.3 - 11.1	5.8 ± 0.1
Ughelli West	05°32.29" 06°53.68"	12,890 - 62,930	3776 ± 61	7843 - 12,374	10,810 ± 122	1.2 - 31.0	9.4 ± 1.0
Otorogu	05°25.03" 06°53.09"	5410 - 8180	6213 ± 76	2221 - 6136	4240 ± 80	0.7 - 13.2	7.8 ± 0.8
Mean		4875 - 27,265	11,335 ± 112	3843 - 6910	4508 ± 96	3.2 - 43.7	20.3 ± 1.7
Control		1430 - 1810	1660 ± 13.4	396 - 489	465 ± 43	ND - 0.10	0.60 ± 0.01

ND = Not Detected, with detection limit of 0.5 Bq kg⁻¹ soil and sediment and 0.001 Bq l⁻¹ for water.

152.11 ± 61.67 - 322 ± 121.67 Bq kg⁻¹ reported in selected oil polluted fields in Rivers State of Nigeria [28]. But the mean beta activity obtained here is far higher than the reported beta activities of 2929 ± 170 Bq kg⁻¹ in Bayelsa State and the 311.15 ± 83.3 - 615.5 ± 178.83 Bq kg⁻¹ in Rivers State [28] [29]. This high activity concentration can be attributed to the crude oil matrix which contains all the heavy elements in the periodic table including (radioactive elements).

The obtained mean values of gross alpha and gross beta activities in soil and sediments in this research work are well above the reported alpha (48.5 ± 15.8 - 64.0 ± 10.0 Bq kg⁻¹) and beta (411.5 ± 11.5 - 2710.0 ± 150.0 Bq kg⁻¹) in surface soil around a steel processing facility [1], which indicates that elements that make up steel materials are less radioactive compared to crude oil elements. The obtained values in this research work are within the mean gross alpha activity of 522 ± 192 Bq kg⁻¹, but they are higher than the mean gross beta activity value of 681 ± 146 Bq kg⁻¹ reported in the soil from Kavadarci in Republic of Macedonia [30], which shows that environmental conditions in addition to geological formations contribute to the gross activities of an area. Since studies have shown that almost all the elements in the periodic table including heavy metals (radioi-

sotopes) are found in the crude oil matrix [10] [16] [17], the general elevation of the gross alpha and beta activity concentration recorded in all the oil fields compared to activity values obtained in control locations is due to the radioactivity content in the crude oil that has spilled into and impacted the polluted soil in the study locations. This agrees with the reported radiological impacts of crude oil in soil sample Bayelsa and Rivers [28] [29].

Figure 1 and Figure 2 show the comparison of the alpha and beta activity concentration of soil and sediment in the different locations investigated. A comparison of the alpha activity concentration in soil and sediments in the various locations indicate a clear variation from location to location as shown in Figure 1, which indicates that additional factors may also be responsible for the

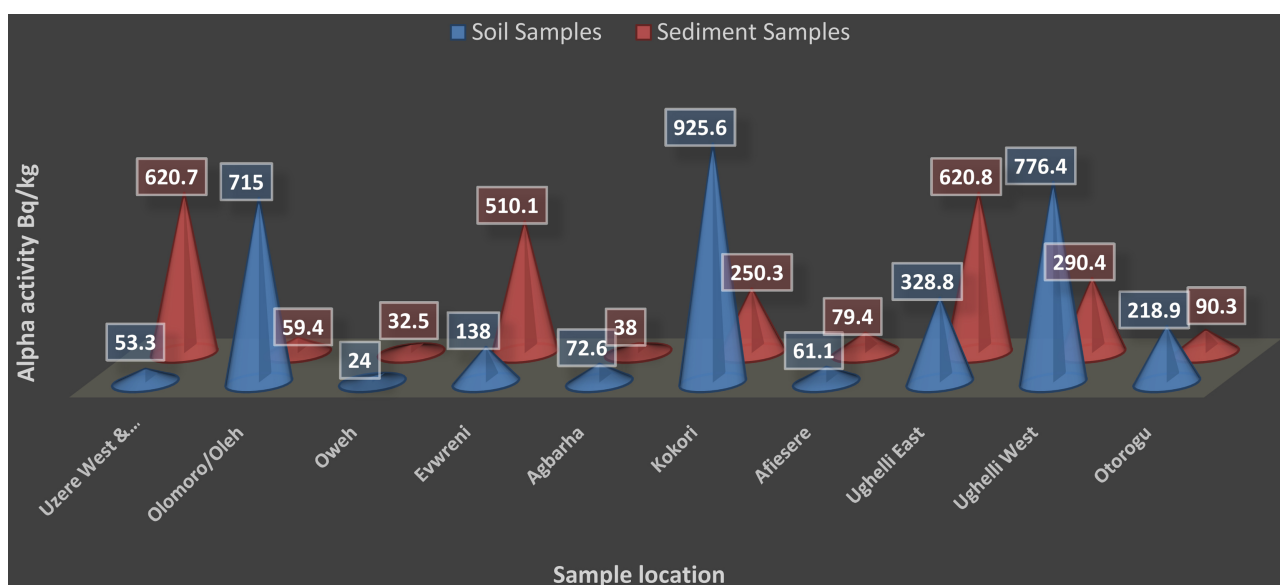


Figure 1. Gross alpha activity in soil and sediment samples.

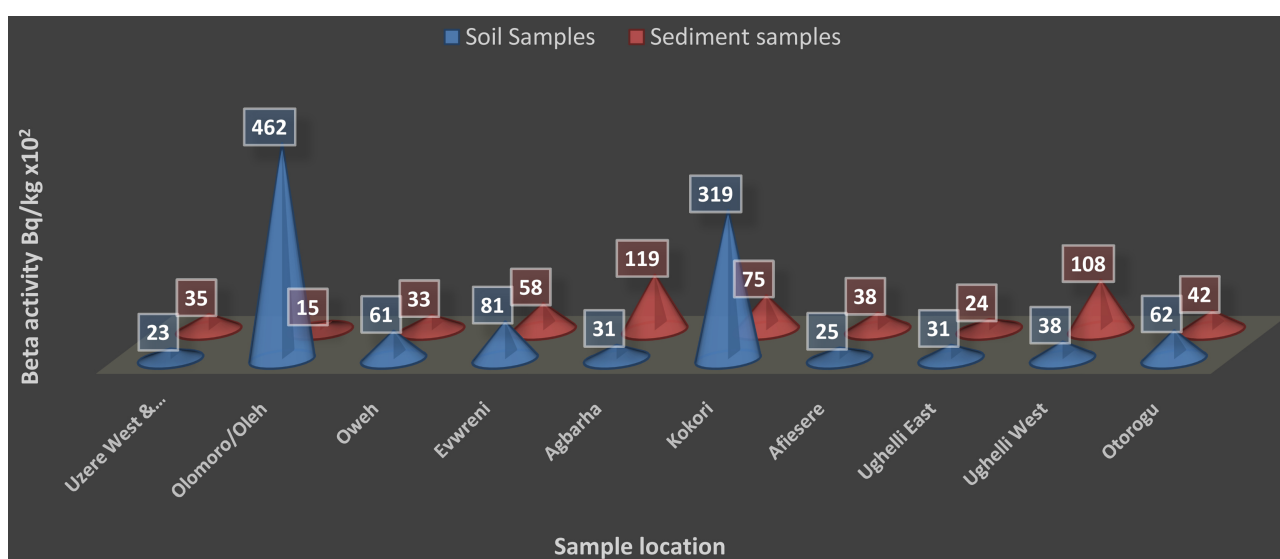


Figure 2. Gross beta activity in soil and sediment samples.

elevation of alpha activity levels in these crude-oil-spilled environments. Similarly, the same trend is also observed in the comparison of gross beta activity in soil and sediment as shown in **Figure 2**. This pattern of variation however confirmed that there is a radiological pollution of the soil and sediment samples through the crude oil spillage on the environment. **Figure 3** shows the comparison of gross alpha and beta activity in water samples.

There is a weak correlation of the alpha and beta activities in the soil samples as shown in **Figure 4(a)** with a regression value of $R^2 = 0.473$, while a poor correlation was recorded between alpha and beta activities in sediment samples as shown in **Figure 4(b)**, with a regression value of $R^2 = 0.0249$. The correlation of gross alpha and beta activity in water sample is also poor, as shown in **Figure 4(c)**. The poor to weak correlations observed can be attributed to the different

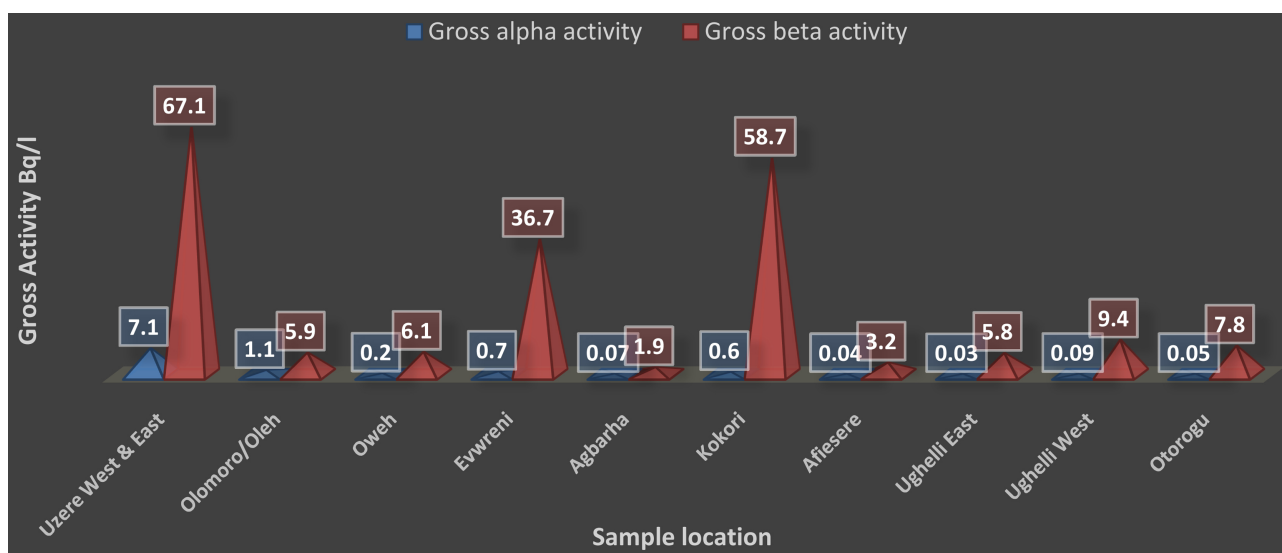


Figure 3. Comparison of gross alpha and beta activity in water samples.

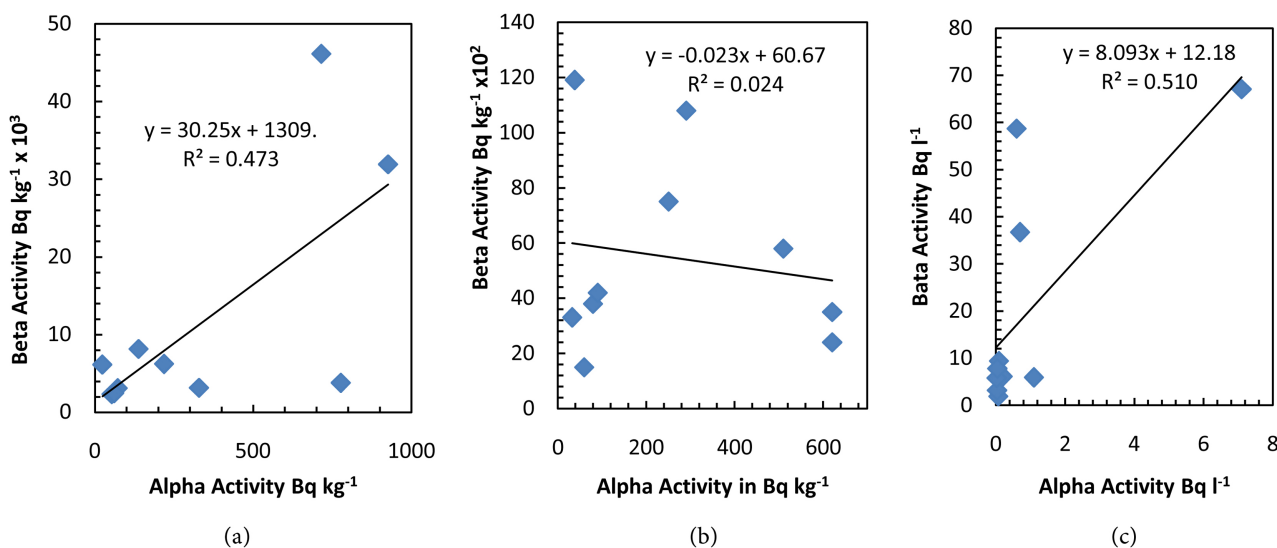


Figure 4. Correlation of gross alpha and beta activity in (a) soil samples, (b) sediment samples, and (c) water samples.

sources of the alpha radiation emitters (more of ^{226}Ra) and beta (^{210}Pb and ^{228}Ra) radiation emitters [11] [12] [31] [32] [33].

3.2. Gross Alpha and Gross Beta Activity in Water Samples

The results of the gross alpha and gross beta activities in the water samples analyzed are presented in columns 5 of **Table 1** and **Table 2** respectively. Average alpha activity concentration in the oil spilled river water samples ranged from $0.03 \pm 0.00 \text{ Bq l}^{-1}$ in Ughelli East (Eruemukohwarien) field location to $7.10 \pm 0.60 \text{ Bq l}^{-1}$ in Uzere oil field with a mean value of $1.00 \pm 0.09 \text{ Bq l}^{-1}$ and a control value of $0.01 \pm 0.00 \text{ Bq l}^{-1}$ while the average beta activity concentration ranged from $1.90 \pm 0.30 \text{ Bq l}^{-1}$ in Agbarha river water sample, to $67.10 \pm 4.60 \text{ Bq l}^{-1}$ in Uzere water sample with a mean value of $20.30 \pm 1.7 \text{ Bq l}^{-1}$ and a control beta activity value of $0.60 \pm 0.01 \text{ Bq l}^{-1}$.

The alpha activity concentrations in Uzere, Olomoro, Oweh, Ewreni and Kokori river water samples were above the practical screening level of 0.1 Bq l^{-1} . The spark (very high) value recorded in Uzere water may be attributed to rich uranium underlying the rock in the crude oil reservoir which may have contaminated the crude during the exploitation processes. The drilling mud used in the drilling of the oil well where the spillage originated from may also have contributed to the high value recorded since the well was recently drilled and coupled rig well. The mean alpha activity concentration of $1.00 \pm 0.09 \text{ Bq l}^{-1}$ recorded in this study is well above the practical screening level of 0.1 Bq l^{-1} recommended by WHO [34]. This is an indication of radiological contaminated water bodies. The mean alpha activity obtained in this research work is higher than the $0.0064 \pm 0.0001 \text{ Bq l}^{-1}$, $0.016 \pm 0.0001 \text{ Bq l}^{-1}$, $0.0129 \pm 0.0001 \text{ Bq l}^{-1}$, $0.0182 \pm 0.0001 \text{ Bq l}^{-1}$ reported in Ovwian, Aladja, DSC Town and Warri respectively [1]. The alpha and beta activity values reported in oil producing domains in Abia State, Nigeria (alpha = $237.78 - 267.00 \text{ Bq m}^{-3}$) (beta = $1323.67 - 1539.67 \text{ Bq m}^{-3}$) where coal and other solid minerals are mined [35] and for ground water reported in Ado-Ekiti where gross alpha and beta values are $0.589 \pm 0.360 \text{ Bq l}^{-1}$ and $0.236 \pm 0.190 \text{ Bq l}^{-1}$ respectively [36], were lower than the reported values in these polluted water samples under investigation. Thus, coal and granites which are solid minerals found in these areas have radionuclide concentration lower than that found in crude oil. However, the values are within the range of values reported earlier in the region by [15] and the alpha average activity of 0.6 Bq l^{-1} reported in Venezuela population well water [37]. But the average alpha activity value of 6.35 Bq l^{-1} reported in Kaduna well water by [25] is higher than the mean value obtained in this study. The higher values in Kaduna well water can be attributed to the flow of water from the fertilizer blending plant into the Kaduna River as reported by the researcher. A comparison of the results obtained in this study with the values reported in drinkable water in different countries of the world, show clearly that they are generally high, compared to the values of tap water in Iran [38], bottled water in Spain [39] and in Mexico

[40], and ground water in Western Anatolia, Turkey [41], Rio Gande do Norte, Brazil [42] and Tekirdag, Turkey [43]. There exists a slight strong correlation of the alpha and beta activities in the water samples as shown in **Figure 4(c)** with a regression value of $R^2 = 0.5107$.

3.3. Estimation of Radiological Risk Parameters

The following radiological risk parameters were further used to quantify the health impacts associated with the exposure to environmental radiation in water from the gross alpha and gross beta activities.

3.3.1. Annual Effective Dose Equivalent (AEDE)

The annual alpha and beta effective dose equivalent due to intake of water was determined by averaging the individual annual committed effective doses contributed by the major alpha and beta emitters in the ^{238}U and ^{232}Th naturally occurring radionuclides series respectively [1] [44]. The Annual Effective Dose Equivalent (*AEDE*) is the quantity of ionizing radiation a person may receive in a year according to protection guidelines. The formula for computation of *AEDE* for gross alpha or gross beta radiation received is given as [44] [45]

$$AEDE_{(\alpha,\beta)} = A_{(\alpha,\beta)} \times W_C \times DCF_{(\alpha,\beta)} \quad (9)$$

The sum of *AEDE* for gross alpha and gross beta radiation is given as

$$AEDE_{T(\alpha,\beta)} (\text{mSv} \cdot \text{y}^{-1}) = \sum_i^{(\alpha,\beta)} A_{(\alpha,\beta)} \times W_C \times DCF_{(\alpha,\beta)} \quad (10)$$

where $A_{(\alpha,\beta)}$ is the average activity concentrations of alpha or beta in Bq l^{-1} , W_C is the water consumed by an adult in a year (It is assumed that standard water consumption for a normal adult is approximately 2 liters a day which approximates to 730 liters in a year) and $DCF_{(\alpha,\beta)}$ is the ingestion dose conversion factor for individual natural radionuclides for an adult extracted from UNSCEAR report [44]. In line with the procedure by Damla *et al.*, (2006) [32] and Ogundare and Adekoya (2015) [1], it is considered that about 50% or more of the annual dose from the intake of water corresponds to radium (gross alpha radium) [1]. This was adopted in this work, since the machine used could not determine the component radionuclides. The major contributors to the gross beta activities are ^{210}Pb and ^{228}Ra [33]. For calculations, the dose conversion factors of 2.80×10^{-4} mSv Bq^{-1} for ^{226}Ra (gross alpha) and 6.90×10^{-4} mSv Bq^{-1} for both ^{210}Pb and ^{228}Ra (gross beta) published by the WHO [46] were used.

Table 3 presents the summary of the radiological risk parameters of annual alpha and beta effective dose equivalent (*AEDE*) for water samples. The total $AEDE_{T(\alpha,\beta)}$ value ranged from 0.99 mSv y^{-1} in Agbarha sampled field to 36.70 mSv y^{-1} in Uzere field with a mean value of 10.64 mSv y^{-1} and a control value of 0.30 mSv y^{-1} . These reported values are far above those reported by [1] in Aladja, Ovwian and Warri all in Delta State. The mean value obtained in this study is 100 times greater than the recommended reference dose level (RDL) of the committed effective dose equivalent of 0.1 mSv y^{-1} for drinking water. The control value

Table 3. Summary of radiological risk parameters for mean gross alpha and beta activities in water samples (errors ignored).

Sample Location	GPS Coordinate	Gross Alpha Bq l ⁻¹	Gross Beta Bq l ⁻¹	AEDE mSv y ⁻¹	AEDE mSv y ⁻¹	AEDE mSv y ⁻¹	AGDE mSv y ⁻¹	ELCR mSv y ⁻¹ × 10 ⁻³
Uzere West & East	05°25.57" 06°13.09"	7.10 ± 0.60	67.10 ± 4.60	2.90	33.80	36.70	169.73	0.130
Olomoro/Oleh	05°28.98" 06°08.37"	1.10 ± 0.20	5.90 ± 0.30	0.45	2.97	3.42	14.96	0.012
Oweh	05°29.55" 06°07.32"	0.20 ± 0.04	6.10 ± 0.40	0.08	3.07	3.15	15.37	0.010
Evwreni	05°22.42" 06°02.32"	0.70 ± 0.05	36.70 ± 3.50	0.29	18.49	18.78	92.52	0.066
Agbarha	05°32.19" 06°02.33"	0.07 ± 0.00	1.90 ± 0.30	0.03	0.96	0.99	4.81	0.004
Kokori	05°39.17" 06°04.25"	0.60 ± 0.02	58.70 ± 5.60	0.25	29.57	29.82	147.91	0.104
Afiesere	05°32.86" 06°00.81"	0.04 ± 0.00	3.20 ± 0.10	0.02	1.61	1.63	8.06	0.006
Ughelli East	05°30.97" 06°55.01"	0.03 ± 0.00	5.80 ± 0.10	0.01	2.92	3.92	14.61	0.014
Ughelli West	05°32.29" 06°53.68"	0.09 ± 0.01	9.40 ± 1.00	0.04	4.73	4.77	23.66	0.017
Otorogu	05°25.03" 06°53.09"	0.05 ± 0.00	7.80 ± 0.80	0.02	3.93	3.95	19.66	0.014
Mean		1.00 ± 0.09	20.30 ± 1.70	0.41	10.23	10.64	51.25	0.037
Control		0.01 ± 0.00	0.60 ± 0.01	2.10 × 10⁻³	0.30	0.30	1.50	5.25 × 10⁻³
WHO Recommendation		0.1	1.0		0.1	0.1	0.30	0.29 × 10⁻³ mSv y⁻¹

obtained is thrice the RDL, thus both the polluted water and the control water from the environment are not suitable for drinking radiologically. Thus, the surface water of the oil producing areas where these oil spillages occurred needed to be treated before drinking. **Table 4** and **Table 5** present the comparison of the average values obtained in the three different samples of water and soil/sediment with previously reported values in literature. The results of the yearly monitoring of the gross alpha and beta activities in the sampled locations show a gradual yearly reduction in gross activity with the fourth year witnessing the greatest reduction or decline in activity concentration. This reduction/decay process fits in fairly into the exponential decay curve for both gross alpha analyses. However, gross beta exponential curve was more of rapid decay curve compared to alpha activity. This can be attributed to the shorter half-life of beta emitter radionuclides in the samples compared to those of alpha emitter.

The gradual reduction in the gross alpha and beta activities may be attributed to natural (bio) remediation due to growing plants and microorganisms' activities that have taken place over time in the environment under investigation.

Table 4. Comparison of gross alpha and beta activity concentration in water in study area with other reported values within Nigeria and other countries in the world.

S/N	α -Activity Bq l ⁻¹	β -Activity Bq l ⁻¹	City/Country of the Study	References
1	10.30	0.79	Delta State, Nigeria	Avwiri and Agbalagba [6]
2	0.782	0.816	Bendimalu River, Turkey	Selçuk <i>et al.</i> [47]
3	0.0118	Not available	Owian-Aladja, Nigeria	Ogundare and Adekoye [1]
4	0.0089	0.271	Kastamonu, Turkey	Kam and Bozkurt [48]
5	0.267	1.539	Abia State, Nigeria	Enyinna and Avwiri [35]
6	0.589	0.236	Ado- Ekiti, Nigeria	Fasae [36]
7	0.60	Not available	Venezuela	Sojo-Bohus <i>et al.</i> [37]
8	6.35	Not available	Kaduna, Nigeria	Onoja <i>et al.</i> [25]
9	0.35	Not available	Sokoto, Nigeria	Sa'idu <i>et al.</i> [49]
10	0.057	3.535	Kebbi, Nigeria	Baba-Kutigi <i>et al.</i> [50]
11	1.00	20.30	Niger Delta, Nigeria	Present study

Table 5. Comparison of Gross Alpha and Beta Activity concentration in soil/sediment in study area with other reported values within Nigeria and other countries in the world.

S/N	α -Activity Bq l ⁻¹	β -Activity Bq l ⁻¹	City/Country of the Study	References
1	4.277	11.773	Turkey	Selçuk <i>et al.</i> [47]
2	530	2929	Bayelsa State, Nigeria	Meindinyo and Agbalagba [29]
3	152.11	311.0	Rivers State, Nigeria	Anekwe <i>et al.</i> [28]
4	64.0	411.5	Owian-Aladja, Nigeria	Ogundare and Adekoye [1]
5	522	681	Kavadarci, Macedonia	Dimovska, <i>et al.</i> [30]
6	331.4	11,335	Niger Delta, Nigeria	Present study (soil)
7	259.2	4508	Niger Delta, Nigeria	Present study (sediment)

3.3.2. Annual Gonadal Dose Equivalent (AGDE)

Annual Gonadal Dose Equivalent (*AGDE*) measures the dose of gross alpha and gross beta received by the bone marrow and the bone surface cells as a result of exposure to radiation [51]. The computation of *AGDE* for gross alpha or gross beta is given by the formula [45].

$$AGDE_{(\alpha,\beta)} = \frac{AEDE}{RWF \times TWF} \quad (11)$$

The sum of *AGDE* for gross alpha and gross beta radiation is given as

$$AGDE_{T(\alpha,\beta)} = \sum_i^{(\alpha,\beta)} \frac{AEDE}{RWF \times TWF} \quad (12)$$

where, *RWF* is the radiation weighting factor and *TWF* is the tissue weighting factor; *RWF* for α activity is 20, and for β activity it is given as 1. For gonads, *TWF* is 0.20 for α -activity and β -activity.

Table 3 shows the obtained values of total annual gonadal dose equivalent

$AGDE_{(\alpha,\beta)}$ calculated from the $AGDE_{\alpha}$ radiation and $AGDE_{\beta}$ radiation. The $AGDE_{(\alpha,\beta)}$ values ranged from 8.06 mSv y^{-1} to 169.73 mSv y^{-1} with a mean value of 51.25 mSv y^{-1} and a control value of 1.50 mSv y^{-1} . The estimated values in this study are higher than the reported values in drinking water around steel processing facility and the $AGDE_{(\alpha,\beta)}$ values of the control, which also exceeded the world allowable limit of 0.3 mSv y^{-1} [1] [52].

3.3.3. Excess Lifetime Cancer Risk (ELCR)

Excess Lifetime Cancer Risk (*ELCR*) is the probability of developing cancer over a lifetime at a given exposure level [45]. In this work, 70 years was considered as the average duration of life for humans [53] [54]. *ELCR* for gross alpha or gross beta was calculated using the formula (Mangset *et al.* 2014) [45].

$$ELCR_{(\alpha,\beta)} = AEDE \times DL \times RF \quad (13)$$

$$ELCR_{T(\alpha,\beta)} = \sum_i^{(\alpha,\beta)} [AEDE \times DL \times RF] \quad (14)$$

where *DL* is the average life span of man (estimated to be 70 years), and *RF* is Risk Factor (Sv^{-1}), which is fatal cancer risk per Sievert. For stochastic effects, the International Commission on Radiological Protection (ICRP) recommended *RF* as 0.05 Sv^{-1} equivalent to 5.0×10^{-5} (mSv^{-1}) for the public [55].

The results of the computed total Excess Lifetime Cancer Risk ($ELCR_{T(\alpha,\beta)}$) from gross alpha and gross beta activities are presented in **Table 3**. The $ELCR_{T(\alpha,\beta)}$ values ranged from 0.006 $\mu Sv y^{-1}$ to 0.130 $\mu Sv y^{-1}$ with a mean value of 0.037 $\mu Sv y^{-1}$ and a control value of 5.25×10^{-3} $\mu Sv y^{-1}$. These $ELCR_{T(\alpha,\beta)}$ values obtained are below the 0.29 mSv y^{-1} recommended value [54]. This indicates that the chance of contracting cancer from the reported contamination is low despite the degree of crude oil pollution of the water bodies. The overall results of the gross alpha and beta activity concentration in water of the study area show that the water bodies have been impaired radiologically by the crude oil spillage, which contain radioactive content in crude oil [17].

4. Conclusions

An analytical approach to the assessment of gross alpha and beta activities in soil, sediment and water samples from oil spilled environment in Delta State oil bearing areas has been carried out. The results obtained show a significant elevation of the gross alpha and beta activities due to the crude oil spillage into the soil and sediment environments and the crude contamination of both surface and subsurface water body. The elevation of the gross alpha and beta activity concentration observed in the oil fields is attributed to the radioactivity content in the crude oil that have spilled into and impacted the polluted soil and water in the study locations. A clear evidence of radiological contamination of the study areas was established with some field locations more impacted than others. The average gross activities in water obtained exceeded the practical screening levels recommended by World Health Organization (WHO). The estimation of the ra-

biological risk parameters indicates that the mean annual effect equivalent dose and annual gonadal dose equivalent were far higher than the international recommended limit and higher than values reported in some parts of the world, but the excess lifetime cancers risk factor mean value for water is within the recommendation permissible limits.

It is recommended that clean-up be done on all oil polluted environments and proper treatment of water sources be carried out before use, while further research study on specific radioactivity should be conducted.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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