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Radiological Risks from Natural Radionuclides in Surface Soil of Agbara Industrial Area, Ogun State, Nigeria

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ABSTRACT: Measurement of natural radioactivity in soil samples from the Agbara Industrial area, Nigeria, was measured using gamma-ray spectrometry with NaI (Tl) detector. The concentration of ^{238}U ranged from 10.21±3.50 to 67.41±18.2 Bqkg^{-1}, from 26.43±10.8 to 96.24±18.81 for ^{232}Th and from298.65±60.70 to 840.52±150.25 Bqkg^{-1} for ^{40}K. Their means were, respectively, 28.69±11.00, 45.86±10.25, and 481.22±106.17 Bqkg^{-1}. Annual effective varies from 0.08 to 0.16 mSv y^{-1} with a mean of 0.11 mSv y^{-1}. The mean contamination factor was 0.87 for ^{228}Ra, 1.02 for ^{232}Th and, 1.15 for ^{40}K.The soil is moderately polluted with ^{232}Th and ^{40}K. The Pollution Level Index indicates a drop in soil quality in about 50% of the areas covered. The mean cancer risk (0.4 x 10^{-3}) obtained in this study is above the World Health Organization limit, indicating a high probability for inhabitants to develop lung cancer in the long term when a lifetime is spent in this area under study.

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Natural radionuclide has been present since the existence of the earth. So Man is continuously exposed to natural radioactivity everywhere on the earth's surface. About eighty per cent of the radiation a human received per year is due to natural background gamma radiation and received about 2.4 mSv of natural radiation in a year (IAEA 1996). The level of natural radioactivity of an area depends on its geology, rocks, and soil types (Tzortzis *et al.*, 2004). The sources of natural radiation are ^{40}K, ^{238}U, ^{232}Th, and their progeny found in the ground (Al-Jundi *et al.*, 2003). Urbanization, mining activities, waste disposal systems, and several human activities are contributors to the radioactivity of the environment (UNSCEAR, 2000). Many industries discharge untreated wastes (wastewater, effluent, sludge) into their vicinity, which may contain an elevated concentration of radionuclides due to the technological processes involved in some productions in some of these industries. High levels of NORM are well-known in major industrial areas (Faisal *et al.*, 2014 in Savar industrial area, Bangladesh; Attia *et al.*, (2015) in Port Said Egypt; Zaim *et al.*, (2016) in Turkey; Ugbede and Benson(2018) in Nigeria and Shen *et al.*, 2019 in general industrial waste in Guizhou, China). Radioactivity in soils is gaining considerable interest from researchers both in Nigeria and abroad because it estimates public doses and predicts changes caused by radionuclides in the environment (UNSCEAR, 2000). Hence, the growing concern about the quality of the environment needs a quality impact assessment of radioactivity of the environment to predict radioactive damage done by the industries. The study aimed to measure natural radioactivity in soil samples from the Agbara Industrial area, Nigeria. The results obtained will be used to estimate the radiological hazards and pollution level index.

MATERIALS AND METHODS

Study Area: Agbara industrial area is positioned within longitude 2.82o and 3.09o E, latitude 6.50o and 7.92o N.

Fig 1: Geo-satellite map of Agbara Industrial Area showing the sampling points

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It is situated about 30 km away from Lagos along the Lagos-Badagry expressway in Ogun State, Nigeria. The altitude is about 37 asl (Ojekunle *et al.,* 2018) (Figure 1). Agbara is known for industrial activities. Many industries are domicile in the area ranging from milling, pharmaceuticals and chemicals, processing, and manufacturing industries like (building materials, e.g. tiles, aluminum companies, beverages, pampers, etc.)

*Collection of samples and preparation for analysis***:** Sixty (60) samples; four (4) each from twenty locations in Agbara industrial areas were randomly collected. The samples were collected 15-20 cm deep at each location. The samples were collected 15-20 cm deep at each location. All the samples were individually parked into a polythene bag, labeled, and spread on a tray for 3days to dry at room temperature. They were processed using a standard procedure that is irrelevant materials such as stones, roots, gravel, etc, were removed, and the samples were well mixed afterward. The samples were crushed into powder and sieved through a 2 mm sieve. A 200 g of the sieved samples were placed into the plastic container and sealed for a month for secular equilibrium before measurement (Sathyapriva *et al., 2017*). Table 1shows the sampling locations and their coordinates.

Table 1: sampling location and their coordinates in Agbara area

Sampling Point Sample ID	Location	Latitude N^o	Longitudes E^o
S1	Ketu Adie-Owe (Momo Agent area)	06^{o}35` 12.13``	003^{o}04`55.2``
S2	Big Cola plant	06^o 30` 22.32``	003^{o}04` 30.0``
S3	Servico area (Phase 3)	06^o 30` 43.92``	003^o 04` 18.84``
S4	Drury Industrial area	06^o 30` 37.08``	003^{o}04`19.99``
S5	Opic Estate (Petedo area)	06^o 30` 50.94``	003^o 04`19.45``
S6	Opic (NPF Station Area)	06^o 31` 8.7``	003^{o}05`12.7``
S7	Access Bank Area	06^o 30` 2.2``	003^o 05`36.64``
S8	Overcomer N/P School area	06^o 30` 23.27``	003^{o}05`50.86``
S9	Nestle Factory area	06^o 30` 6.84``	00305`16.84``
S10	Corona Sec Sch. Area	06^o 30` 37.44``	003^{o}05`17.41``
S11	Reckitt Company area	06^o 30` 19.87``	003^{o}05` 30.3``
S12	Procter and Gamble plant area	06^o 32` 34..80``	003^{o}04`37.7``
S13	Omoshola Phase 2 (Ologbo Eremi	06^o 32` 0.42``	00.3^o 02`1.86``
S14	Beta Glass area	06^o 30` 11.02``	003^o 05`39.16``
S15	Print Color Africa Area	06^o 32` 51.02``	003^{o}04` 26.65``
S16	Crown City Resorts and Hotel area	06^o 32` 37.64``	003^{o}04`24.64``
S17	Access Closa Agent Area	06^o 32` 49.42``	003^o 04`27.59``
S18	Firstmonie Agent area	06^o 32` 45.02``	003^{o}04` 34.19``
S19	Beloxxi Group Industries area	07^o 54` 56.39``	002^o 49`37,2``
S20	Primera food	06^o 32` 21.84``	003^o 02`16.87``

Activity determination analysis: A Sodium Iodide NaI (Tl) detector was used to measure radionuclides in the samples. A scintillation detector and a Canberra multi-channel analyzer were set up for the counting. A 7.6 x 7.6 cmcm2 NaI (Tl) manufactured by Bicron Electronics Ltd, USA with model no. 8020 was used and connected to the multi-channel analyzer with a coaxial cable. The detector is shielded by a cylindrical lead of 5 cm thick at the bottom and at the top to shield against background radiation. The calibration for efficiency detection was performed with a standard reference gamma source supplied by Rocketdyne Laboratories, Canoga Park, CA, USA. The resolution of the detector assembly is ~ 8% at 0.662 MeV of 137Cs. The energy calibration was performed with gamma sources from the IAEA, Vienna. The energy-channel calibration obtained was fit linearly, and the equation was stored in the memory of the analyzer to measure the activities of the radionuclides. The count of an empty container was taken as background count and was removed from the gross count to get the net count. The counting time was 10 h. The concentration of radionuclides in the samples was determined using1.764 MeV gamma-rays from 214Bi for226Ra, gamma-ray energy of 2.614 MeV from 208Tl for the activity of 232Th, and gamma-ray of 1.460 MeV from 40K for the40K activity. A software (Genie 2K), spectrum acquisition and analysis software manufactured by Canberra Industries Inc. USA) was used to analyze the activity of radionuclides.

Determination of radiological hazards: Absorbed dose: The absorbed dose rate (Do) in the air at the height of 1 m above the ground was estimated from the concentrations of ^{238}U, ^{232}Th, and ^{40}K as described in equation 1 given by UNSCEAR, 2000.

$$Do = 0.428C_{Ra} + 0.632C_{Th} + 0.044C_K \quad (1)$$

Where C_{Ra}, C_{Th}, and C_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K.

Effective Dose (ED): The annual effective dose (ED) was calculated using equation 2. This equation converts the absorbed dose rate to effective dose using 0.7 $SvGy^{-1}$ and considers the outdoor occupancy factor, i.e. the average time spent outdoors by people. On average, inhabitants spent 7-8hours a day outdoor, meaning that about 30% of the 365 days in a year (Cevik *et al.,* 2008).

$$ED(\mu Sv) = D_0\,(nGy\,h^{-1}) \times 8760(h\,y^{-1}) \times 0.7(SvG\,y^{-1}) \times 10^{-6} \times 0.3 \quad (2)$$

Where D_0 is the absorbed dose ($nGyh^{-1}$); 8760 is the hours in a year; 0.3 is the outdoor occupancy factor; 0.7 $SvGy^{-1}$ is the conversion factor from D_0 to ED and 10^{-6}converts nano into milli.

Radium Equivalent (Ra_{eq}): This is a quantity that is commonly used to identify the uniformity of radiation exposure i.e. the activity concentration of a radionuclide equivalent to 370 Bq kg^{-1} of ^{226}Ra.The quantity Ra_{eq} was calculated using equation 3 UNSCEAR, (2000)

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (3)$$

Where Ra is the radium equivalent activity; C_{Ra}, $C_{Th,}$ and C_K denote ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

External Hazard Index (H_{ex}): Parts of the area under study are used to cultivate crops, especially vegetables, and to fill in the construction of houses. The soil samples may contain an elevated concentration of natural radionuclides, which may add to the external gamma dose rates the inhabitants receive. To reduce dose the inhabitants from the usage of soil in dwelling construction, H_{ex} was calculated from equation 4(El-Taher, (2010)

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (4)$$

Where Hex is the external hazard index, C_{Ra}, $C_{Th,}$ and C_K denote the usual meaning.

Internal Hazard Index (H_{in}): Radon and its short-lived products are products of the uranium series and are hazardous to the respiratory system. Internal exposure to radon and its short-lived products from the soil termed internal hazard index (H_{in}) was calculated using equation 5 given by (UNSCEAR, 2000)

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (5)$$

Through the inhalations of dust particles, the inhabitants are exposed to radioactive materials from the dust. Through inhalation of contaminated dust, the inhabitants are liable to develop lung cancer. Therefore, the probability of developing lung cancer (Cancer Risk) (CR) was estimated per million of the inhabitants, using equation 6given by ICRP, (2007).

$$ELCR = ED \times L_E \times C_F \quad (6)$$

Where CR is the Cancer Risk, ED, the effective dose, L_E is life expectancy (70 years) and C_F is the risk factor given as 0.05 Sv^{-1} (ICRP, 1991).

RESULTS AND DISCUSSION

The concentrations of ^{238}U, ^{232}Th, and ^{40}K are presented in Table 2. The minimum concentration of ^{238}U (10.21±3.50) was obtained in S6, while the maximum was obtained from S3 (67.41±18.2). The mean of ^{226}Ra was 28.69±11.00 $Bqkg^{-1}$. Although the mean value is below the UNSCEAR recommendation, about 35% of the samples have radium concentration above the world average (UNSCEAR, 2008).Also, about 55% of the samples have concentrations above the world average of 45 Bqkg-1 (UNSCEAR, 2008) for ^{232}Th. The minimum concentration was from S1 (26.43±10.8), while the maximum was from S7 (96.24±18.81) with a mean of 45.86±10.25 $Bqkg^{-1}$. The maximum concentration of ^{40}K was obtained in S7 (840.52±150.25 $Bqkg^{-1}$), while the minimum was from S14 (298.65±60.70). About 65% of the samples have concentrations above the world average. The mean concentration of^{40}K was 481.22±106.17 $Bqkg^{-1}$. The radionuclide concentrations are in the order ^{238}U <^{232}Th <^{40}K, which implies that the soil samples analyzed are rich in 40K. Although the results of this study are not sufficiently higher than UNSCEAR recommendations, it should be noted that overexposure to the high concentration of these radionuclides may cause some health issues like lung cancer and other related health challenges (ATSDR, 2014).The results of this study were compared with similar researches locally and internationally in Table 3. The mean of ^{226}Ra obtained in this study (28.69 $Bqkg^{-1}$) is slightly higher than what was obtained in similar researches in Bangladesh, India, and Greece (Faisal *et al*., 2014, Senthikumar and Narayanaswamy, 2016, Ioannides *et al.,* (1997) but lower than results from Pakistan, Malaysia and Algeria (Tufail *et al.,* 2016; UNSCEAR, 2000; Boukhenfouf *et al.,* 2011). A higher result of ^{232}Th than what was obtained in this study was recorded in some studies from Pakistan,

Malaysia, Algeria, and Greece as shown in Table 2. The concentration of ^{40}K was highest in all the sites and the comparison of the results with other studies confirmed this. The mean of this study is lower than what was obtained by a study in India (Narayanaswamy, 2016) and Pakistan (Tufail et al., 2016) but higher than the result of Senthikumar and Narayanaswamy, (2016) in India and Ioannides et al, (1997) in Greece. In Nigeria, the result obtained by Gbadamosi *et al*, 2018 in Agbara industrial area revealed a higher concentration of ^{226}Ra than the present study, while lower concentrations of ^{232}Th and ^{40}K were recorded in their study. Also, these results are lower than results obtained in Port-Harcourt by (Avwiri and Olatunbosun, 2014). The ^{226}Ra result obtained in this study is lower than the result obtained in Lagos (Oladapo *et al.,* 2012) but higher results were obtained for ^{232}Th and ^{40}K in the present study

Table 2: The concentrations of ^{238}U, ^{232}Th, and ^{40}K in the soil samples

Sample ID	^{226}Ra	^{232}Th	^{40}K
S1	33.61±6.10	49.22±12.01	600.12±60.45
S2	14.50±5.81	34.67±10.8	650.80±75.31
S3	47.81±10.2	65.61±17.2	680.50±76.10
S4	20.45±7.21	41.20±13.22	582.40±65.30
S5	28.60±8.50	46.21±10.62	448.50±55.31
S6	12.61±7.80	48.16±15.00	620.11±81.15
S7	46.22±8.81	66.91±12,21	480.28±55.42
S8	35.66±8.11	52.78±15.21	391.60±95.01
S9	15.00±6.53	36.28±10.41	490.66±60.31
S10	18.65±9.24	40.81±9.81	380.48±59.00
S11	31.10±8.62	61.65±16.70	524.30±56.32
S12	34.56±12.90	45.45±13.51	430.42±75.31
S13	44.65±14.87	36.22±11.51	330.39±99.61
S14	22.13±8.92	58.29±11.85	580.70±55.62
S15	38.62±7.88	40.43±8.20	460.10±72.80
S16	35.32±9.46	43.51±8.98	436.67±90.34
S17	30.41±7.92	38.62±6.52	320.82±50.31
S18	15.19±6.80	40.10±11.21	404.48±66.22
S19	20.12±8.10	36.20±9.80	398.61±78.21
S20	28.63±6.28	34.81±10.2	412.51±64.24
Mean	28.69±11.00	45.86±10.25	481.22±106.17

Table 3: Comparison of result with other similar work in the literature

S/N	^{226}Ra	^{232}Th	^{40}K	Country/Region	References
1	23.81	42.24	733.19	Bangladesh	Faisal et al, 2014
2	22.80	39.9	171.22	India	Senthikumar and Narayanaswamy, 2016
3	30	56	602	Pakistan	Tufail et al, 2016
4	66	82	310	Malaysia	UNSCEAR, 2000
5	53.2	50.03	311	Algeria	Boukhenfouf and Boucenna et al., 2011
6	16	55	305	Greece	Ioannides et al., 1997
7	42.95	26.84	111.05	Nigeria (Agbara)	Gbadamosi et al., 2018
8	3.0	3.30	122.10	Nigeria (Sango-Ota)	Ademola et al., 2014
9	41.96	62.61	643	Nigeria(Port-Harcourt)	Avwiri and Olatunbosun, 2014
10	69.19	14.49	409.44	Lagos	Oladapo et al., 2012

Table 4 depicts absorbed dose, annual effective dose, radium equivalent dose, external hazard index, internal hazard index, gamma index, and excess lifetime cancer risk results. Absorbed dose rate (D) ranged from 48.28 $nGyh^{-1}$to 90.55 nGy with a mean of 61.51 $nGyh^{-1}$(Column 2, Table 4). This value is higher than the recommended value of 59 nGy h^{-1}(UNSCEAR, 2000). The annual effective dose as presented in Column 3, Table 4 ranged from 0.08 to 0.16 $mSvy^{-1}$ with an average of 0.11mSv y^{-1}. The mean values were above the recommended value of 0.07 $mSvy^{-1}$in all the sites (UNCSEAR, (2000). The mean value of Ra_{eq} (131.32 $Bqkg^{-1}$) is less than the recommended value of 370 Bq kg^{-1}(UNSCEAR, 1982) and is presented, in column 4.The external hazard index, internal hazard index, and gamma index are presented in columns 5, 6, and 7 in Table 4. The mean values obtained were 0.35, 0.43, and 0.49, respectively. These values are lower than unity, indicating that the samples are non-hazardous to be used to construct dwellings. The cancer risk (CR) for the habitants of the area through inhalation of contaminated dust was estimated and presented in column 8 of Table 4. The mean CR in the study area was 0.4×10^{-3} which, is higher the range recommended by USEPA (1.0×10^{-6} - 1.0×10^{-4}) (USEPA, 2003). This higher value indicates high probability of developing cancer in the long term when a lifetime is spent in this area.

Table 4: The absorbed Dose (D), Effective Dose (ED) and External (Hex) and Internal Hazard Index (H_{in}), Radium Equivalent Dose (Ra_{eq}), Gamma Index (I_γ) and Excess Lifetime Cancer Risk (ELCR) of all topsoil samples

Sample ID	Absorbed dose (D) ($nGyh^{-1}$)	Effective Dose(ED) ($mSvy^{-1}$)	Ra_{eq} ($Bqkg^{-1}$)	H_{ex}	H_{in}	I_γ	ELCR x 10^{-3}
S1	70.82	0.13	150.20	0.40	0.50	0.56	0.46
S2	55.78	0.10	114.19	0.31	0.35	0.44	0.36
S3	90.55	0.16	194.03	0.52	0.65	0.71	0.58
S4	59.44	0.11	124.21	0.33	0.39	0.47	0.38
S5	60.28	0.11	129.21	0.35	0.43	0.48	0.39
S6	62.05	0.11	129.23	0.35	0.38	0.49	0.40
S7	82.07	0.15	178.88	0.48	0.61	0.65	0.53
S8	64.95	0.12	141.29	0.38	0.48	0.51	0.42
S9	50.11	0.09	104.66	0.28	0.32	0.39	0.32
S10	49.75	0.09	106.31	0.29	0.34	0.39	0.32
S11	74.23	0.14	159.63	0.43	0.51	0.59	0.48
S12	61.58	0.11	132.69	0.36	0.45	0.49	0.40
S13	55.84	0.10	121.88	0.33	0.45	0.44	0.36
S14	70.73	0.13	150.19	0.41	0.47	0.56	0.46
S15	61.46	0.11	131.86	0.36	0.46	0.48	0.40
S16	60.97	0.11	131.16	0.35	0.45	0.48	0.39
S17	50.84	0.09	110.34	0.29	0.38	0.40	0.33
S18	48.86	0.08	103.68	0.28	0.32	0.38	0.31
S19	48.28	0.09	102.58	0.28	0.33	0.38	0.31
S20	51.65	0.09	110.17	0.30	0.37	0.41	0.33
Mean	**61.5**	**0.11**	**131.32**	**0.35**	**0.43**	**0.49**	**0.40**

The contamination factor CF of each radionuclide in the soil samples was calculated and the pollution load index was estimated to ascertain the levels of pollution due to radionuclides. The contamination factor (CF) was calculated using equation 7 as described by Hakanson (1980):

$$CF = \frac{C_i n}{C_B} \qquad (7)$$

Where $C_i n$ is the concentration of i_{th} radionuclide in the soil, and C_B is the world average concentration of each radionuclide. CB was taken as 420 $Bqkg^{-1}$, 45 $Bqkg^{-1}$ and 33$Bqkg^{-1}$ for ^{40}K, ^{232}Th and ^{226}Ra, respectively (UNSCEAR, 2008).The result obtained is presented in Table 4. The mean CF was 0.87 for ^{228}Ra, 1.02 for ^{232}Th, and 1.15 for ^{40}K. From the result, the soil is fairly polluted with ^{232}Th and ^{40}K. The pollution load index (PLI) was calculated using equation 8 described by Thomilson *et al*., (1980).

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \ldots\ldots\ldots \times CF_n)^{1/n} \quad (8)$$

Where *n* is the number of radionuclides analyzed and *CF* is the contamination factor of each radionuclide.

PLI < 1 means no contamination; if PLI < 1, means that only baseline levels of pollutants are present and PLI >1 means worsening of the quality of the soil sample (Thomilson *et al.*, 1980).

The result obtained for PLI is presented in Table 5. The PLI of S1, S3, S7, S8, S11, S12, S14, S15, and S16 is higher than one, meaning that there is a drop in soil quality from the but safe in the other sites.

Table 5: Contamination factor (CF) and pollution level index (PLI) of the soil samples

Sample ID	CF (^{226}Ra)	CF ($_{228}Ra$)	CF (40K)	PLI
S1	1.02	1.09	1.43	1.17
S2	0.44	0.77	1.55	0.82
S3	1.45	1.46	1.62	1.51
S4	0.62	0.92	1.39	0.93
S5	0.87	1.03	1.07	0.98
S6	0.38	1.07	1.48	0.9
S7	1.40	1.49	1.14	1.35
S8	1.08	1.17	0.93	1.1
S9	0.45	0.81	1.17	0.76
S10	0.56	0.91	0.91	0.78
S11	0.94	1.37	1.25	1.17
S12	1.05	1.01	1.02	1.05
S13	1.35	0.80	0.79	0.95
S14	0.67	1.30	1.38	1.1
S15	1.17	0.90	1.10	1.05
S16	1.07	0.97	1.04	1.04
S17	0.92	0.86	0.76	0.85
S18	0.46	0.89	0.96	0.73
S19	0.61	0.80	0.95	0.92
S20	0.87	0.77	0.98	0.87
Mean	**0.87**	**1.02**	**1.15**	**1.06**

*Conclusion***:** Measurement of NORMS in the soil around the Agbara Industrial area, Nigeria, was carried out. The mean concentrations of 232Th and 40K were above the world average. Cancer risk is high in the area, indicating a high probability of developing cancer in the long term when a lifetime is spent in this area. The pollution level index indicated a drop in the quality of soil in the study.

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