Original Research Article

EVALUATION OF RADIONUCLIDE CONCENTRATION IN SUSPENDED PARTICULATE MATTER FROM TWO QUARRY SITES IN NIGERIA

Abstract

Evaluation of Radionuclide contents of suspended particulate matter from two quarry sites in Nigeria, have been carried out using High Volume air sampler and Gama spectrometer Na1(Tl) Detector. The results obtained showed the mean value for ^{226} Ra, ^{232}Th and ^{40}k for Ishiagu samples were 2.76, 1.57 and 5.29 Bq/l respectively while that obtained in Okpella samples were 1.84, 2.21 and 6.2 Bq/l respectively. The values obtained were higher than their control values but lower than the world recommended safe values. The results obtained in Okpella are slightly higher than that obtained in Ishiagu sites. This could be due to different geological constituents and other mining activities going on at Okpella that might have contributed to the slight change. All the radiological health risk parameter estimated were within their safe values. Comparison of results with other similar works shows positive correlation. We therefore conclude that inhalable suspended particulate matter within quarry sites does not constitute radiological health hazard though precaution should be taken to avoid long term accumulation that can lead to stochastic health risk.

***Keywords*:** Radionuclide, suspended particulate matter, Quarry sites, Gamma spectrometer.

1. Introduction

Many contaminates arising from anthropogenic activities into the environment has caused major concern in areas of research over the years. This results to environmental and animal health consequences with respect to exposure level above safe limits. Radiations like those of terrestrial and extra-terrestrial origin are from natural sources. Terrestrial ionization radiations are due to uranium (^{238}U), Thorium (^{232}Th) and potassium (^{40}K) radioactive nuclides seen in rocks, soils and air. Extra-terrestrial radiation is those coming from outer space as cosmic rays [1]. The major human induced ionizing radiation comes from nuclear reactors, industries, x-rays, mining and other activities[2]. According to UNSCSAR [3] a background exposure from normal levels of the naturally occupying radiative materials (NORM) is present in all materials of the environment at effective average dose of 2.4m3ry^{-1} .

Technologically enhanced naturally occurring radioactive material (TENORM) referred to where human activities and others increase the relative concentration of radionuclides [1]. Since radionuclides are not evenly distributed, the knowledge of their distribution in soils, rocks and atmosphere play an important role in radiation protection and measurement. Some of the exposures are fairly constant and uniform for all individual persons everywhere. However, other exposure varies widely over location. Radiation exposure arising from the mining and mineral processing industries results from three principal pathways; external gamma radiation from ores, inhalation of dust containing long-lived alpha-emitting radionuclides and inhalation of short-lived decay products of radon. Mining activities and the processing of natural resources have impacted considerably on human beings and its environment and this operation are associated with environmental degradation, destruction of ecosystem and general pollution of the environment [4].

Quarrying activities have lots of challenges on the environments as all stages emit a lot of dust and radioactive elements [5]. Dust particulate of about 20 – 500 microns found in the atmosphere can be carried away easily by wind. Dust from quarry spread along large areas through wind, rain etc. and are accumulated in and on soils, plants and animals and can adversely affect humans above all when these dusts are from radioactive origin [6]. Air pollutants are major environmental problems affecting both the developed and developing countries. The various impacts produced by quarrying activities are both sizes and location dependent, manifesting itself in specific impacts on air, soil, water, human, flora and funna when the particulate are suspended in air for long period [7]. Particulate matters having significant number of radionuclides in them can be carcinogenic or mutagenic in nature [8]. Slight higher degree of radionuclide composition inhaled to the human lungs will result in lung damage [9].

The main hazardous materials in cement production includes fuel (oil, diesel, LNG), chemicals, additives and engine oils and lubricants. The use of hard-standing and secondary containment is evident for large storage tanks, but not for smaller storage tanks, intermediate bulk containers (IBC), mobile equipment, and oil/lubricant drums. BUA will ensure the provision and/or integrity of hard-standing and secondary containment around hydrocarbon storage vessels (including for such materials at workshops). The integrity of secondary containment will be enforced and regularly monitored, at all locations where hazardous liquids are stored (openings to evacuate rainwater were observed perforating the containment walls of secondary containment bunds, at some locations) [8].

Particulate matter contains some level of radionuclides studies have shown that inhalation of alpha emitter could lead to internal exposure. Exposure to radiation care cause several health hazards to man and its environment, some of radiation related risks ranging from malignancies and damage to generic materials have been observed from long term epidemiological studies of population exposed to radiation [10]. Radiation dose of different levels, delivered at different

rates to different parts of the body can cause different types of health effects at different times [11]. At very high radiations exposure, death will occur within several months or less. Based on these deleterious effects, this study aimed at analyzing the radionuclide content of inhalable particulate matter from two quarry sites in order to ascertain the radiological risks of the workers and host community dwellers.

2. Material and Methods

2.1 Study Area

This study was carried out in two different states where Quarrying activities are going on: Okpella Edo State and Ishiagu Ebony State. Okpella – is a clean situated along Benin – Abuja Federal high way located at coordinate of 7.272^0N latitude, 6.3465^0E Longitude. It plays host to BUA cement processing plant. Okpella is endowed with natural sedimentary rock based mineral resources, which include limestone, calcium, granite, feldspar, talc, clay, marbles and so on. In view of the abundance of other solid minerals it is home for several granite and marble-making industrial which gives the community a vibrant industrial outlook (Figure 1).

Ishiagu Quarry site is located at latitude is $5^052^1 – 6^000^1$N and longitude $7^030^1 – 7^035^1$. It is part of the geologic complex called Benue trough, a deep linear sediment filled basin which extends to Niger Delta for over 700 km toward the North – Eastern part of Eke, Isikwuato by South and Lokpa and Lekwensi by West [5]. Ishiagu area is generally a dominant low lying to gentle undulating shaly terrain of 85 – 100km above sea level and punctuated by few isolated low hills. The land surface is usually marshy in wet season which prevails from April – October yielding annual rainfall of between 1200 – 2000mm [11]. According to Eburua and Ezeribe [12] Ishiagu has many mineral deposit, making the inhabitants prominently farmers and miners.

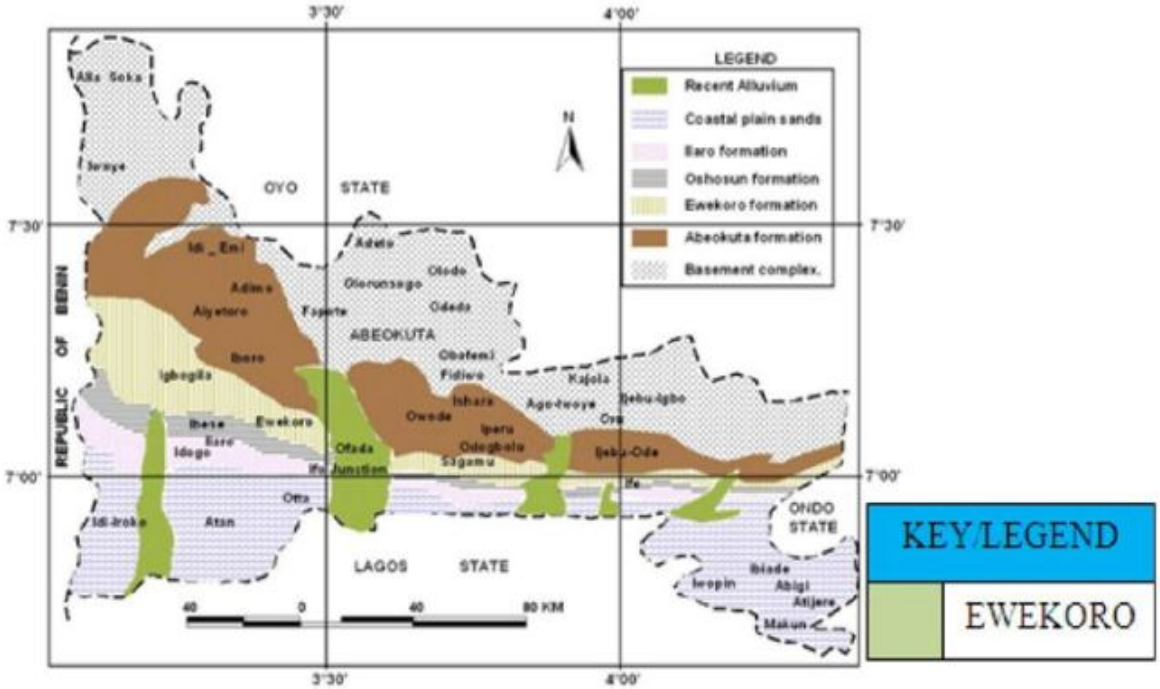

Fig.1: Geological Map of the Study Area (Okpella)

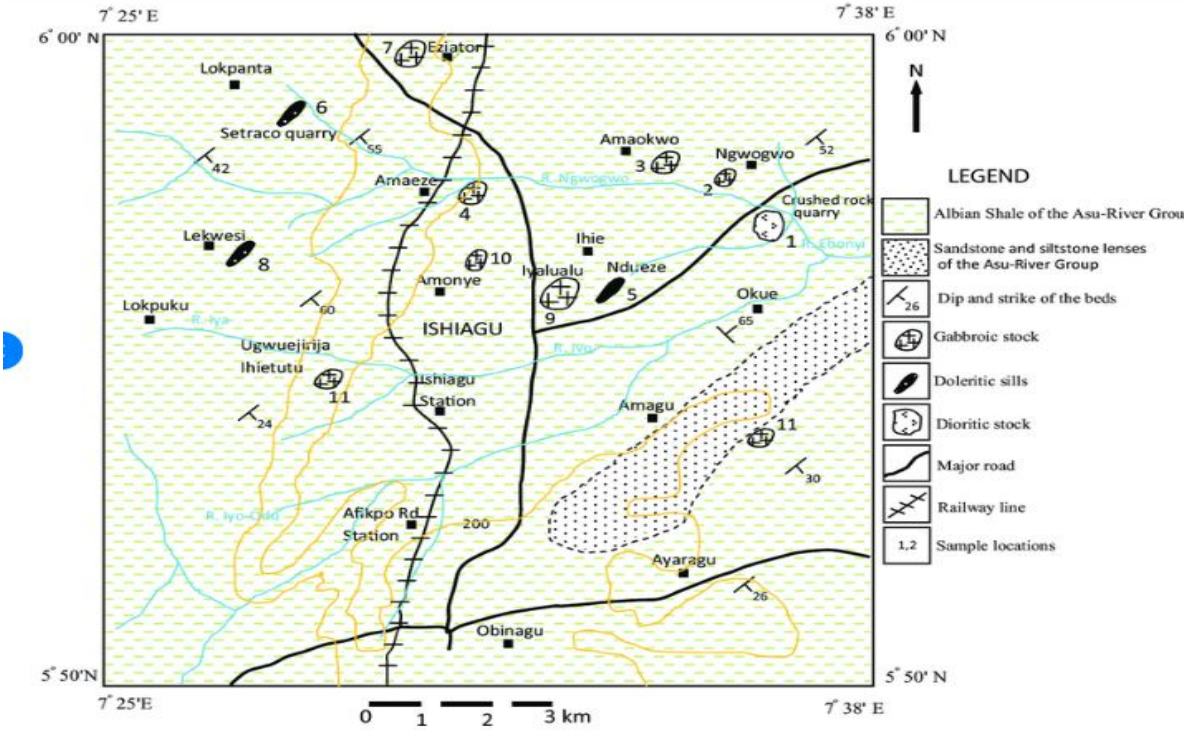

Fig.2: Geological map of Ishiagu Quarry site in Ebonyi State.

2.2 **Sample Collection and Preparation**

The particulate was collected using quarts fibre filter sampling using gravimetric high-volume sampler. Sampling was done once a week and the glass fibre were taken to the laboratory to determine their initial masses; the filters were marked F_1 to F_8 with F_8 as the control in all the locations. These filters were mounted on the gravimetric high volume air sampler which was connected to the generator at a known location and coordinate. Ambient air was drawn through the filter at a constant flow rate of 1.4 m^3/min. suspended particulate matter having dimeter (stroke equivalent diameter) between 0.1 and 100 mm were removed from air stream by filter filtration process on the filter. For each filter used one hour duration was allowed. After which the filter was removed from the sampler and put in a desiccator to avoid moisture and weighed before and after the sampling to obtain the mass of particulate matter (the masses F_2) [13]. The sampling duration for this investigation is one hour, the volume of air sampled was 1.4m^3/min x 60 min = 84 m^3 of air. This is because the shorter the average period the greater the volume of data collected.

The materials used for the digestion of the samples in the filter include Hot plate, 100 ml volumetric flask, 500ml beaker, sample bottle, filter paper, washing balance, distilled water, funnel, conc. H_2SO_4, conc. Per chloric acid and hydrochloric acid.

Each filter sample was inserted in 500ml beaker, 100ml of conc. H_2SO_4, 20ml per chloric acid and 20ml of aqua rigia (3ml of Hcl, 1ml of HNO_3) were added and beaked on hot plate for eight hours. At the end of the digestion, 100ml of each digested sample was transferred into 100ml Marinelli beaker for gamma spectrometry and sealed for 30day to reach secular equilibrium between the parent and daughter nuclide before counting using sodium iodide NaJ(Ti) detector.

2.3 **Gamma ray Spectroscopy**

The gamma-ray counting of the samples are performed on a lower gamma-beam spectrometer comprising of a detector called sodium iodide thallium activated Canberra vertical high purity 2" x 2" sodium iodide coupled with ORTEC 456 digibase amplifier, which amplifiers the incoming signals and integrates them to volts (0 – 10v). The indicator was guided by 1.5m deep lead on every side of the four sides and 1.0m deep lead on every side of the four sides and 1.0 m deep to finish everything. The detector was calibrated for energy and frequency before use and three gamma standard sources , Cs-137, Am-241 and Co-60 were utilized f in order to minimize the effects of background and scattered radiation [14]. The energy calibration was done by obtaining the relationship between peak position in the spectrum and the corresponding gamma ray. The height of each pulse output from a photomultiplier tube which was viewed on the display output and the channel corresponding to it is directly proportional to the initial gamma energy producing the pulse. The calibration was done using gamma emitter sources of known energies,

these are Cs-137 and Co-60 that emits gamma rays with energies of 662 Kev, 1332 KeV and 1173 KeV and Am-241 which is an alpha emitter but also emits some gamma rays with energies 26.3KeV and 59.6 KeV. The gamma emitter sources were exposed to the NaI (TI) detector and gamma spectrum was acquired. These were done with the amplifier gain that gives 72% energy resolution for the 662 KeV of Cs-137 and counted for 30 minutes. The net area corresponding to the photopeak in the energy spectrum was computed by subtracting count from the background source from the total area of the photopeak's. the identification of individual radionuclides was performed using their gamma ray energies and the quantitative analysis of radionuclides were performed using gamma ray spectrum analysis software, Genie 2000.

3. Radiological Health Risk Parameters

The radiological parameters calculated in this work are as seen below:

Radiation Absorbed Dose is a measure of the energy deposited in a medium by ionizing radiation. It is equal to the energy deposited per unit mass of medium, and so has the unit J/kg or gray (Gy) where $1Gy = 1Jkg^{-1}$. The absorbed dose rates (D) due to gamma radiations in the air at 1m above the ground surface for the uniform distribution of the naturally occurring radionuclides ^{238}U, ^{232}Th, and ^{40}K is calculated using

$$D(nayh^{-1}) = 0.462A_{Ra} + 0.621A_{Th} + 0.0417A_k \quad (1)$$

Where A_{Ra}, A_{Th}, and A_k are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}k respectively [3].

Annual Effective Dose Equivalent (AEDE)

This is a measure of the risk of long- term effects of ionizing radiation on the human body, organs and tissues, accounting for their sensitivity. It is evaluated from the absorbed dose rate (D) using the conversion factor of 0.7sv/Gy and the outdoor occupancy factor 0.2 [15]. The annual effective dose equivalent ($mSvy^{-1}$) was calculated using the formula below [3].

$$\text{Annual effective dose rate } (mSvy^{-1}) = D(nGyh^{-1}) \text{ X } 8760hyr^{-1} \text{ x } 0.7 \text{ x } 10^3 \ (mSv/10^9) \text{ nGy c } 0.2$$

$$AEDE = D \text{ x } 1.2264 \text{ x } 10^{-3} \quad (2)$$

Excess Life Cancer Risk

is the probability of developing cancer over life time at a given radiation exposure level. It is presented as a value representing the number of extra cancers expected in a given number of people on exposure to a carcinogen at a dose. It is calculated as [5].

$$ELCR = AEDE \text{ x } DL \text{ x } RF \quad (3)$$

Where AEDE is Annual effective dose equivalent, DL is average duration of life (estimated to be 70 years) and RF is the risk factor (Svy^{-1}), i.e. final cancer risk per sievert for stochastic effect ICRP uses RF as 0.05 Svy^{-1} for the public exposure [16].

Radium Equivalent Activity (Raeq)

equivalent is used to assess the hazard associated with materials hot contain ^{226}Ra, ^{232}Th and ^{40}k in Bqll and is mathematically defined by Mujahid et al.,[17]

$$Raeq\ (Bqlkg) = A_{Ra} + 1.43\ A_{Th}\ r0.077Ak \qquad (4)$$

Where A_{Ra}, A_{Th} and A_k are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}k respectively.

The External hazard Index (Hex)/ Internal hazard index (Hin)

is a criterion use for the evaluation of external exposure to gamma radiation in the outdoor air. This widely used hazard index was calculated using [18]

$$H_{ex} = = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \qquad (5)$$

Where A_{Ra}, A_{Th} and A_k are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}k respectively.

Internal hazard index

Radon and its short-lived products are radiologically hazardous when inhaled. Internal exposure to radon and its daughter products is very hazardous and can lead to respiratory diseases like asthma and cancer. The internal hazard index (Hin) can be used to determine the internal exposure of living cell to radon and its products[5]. Exposure to radon and its daughter products is quantified by the internal hazard index (H_{in}), which is given equation 6 [18]

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \qquad (7)$$

The values of the indices (H_{in} and H_{ex}) must be less than unity for the reduction hazard to be negligible.

Gamma Index (I_γ)

It is a form of radiation hazard index proposed by the European Commission [19]. It is usually calculated from the activity concentration of ^{226}Ra, ^{232}Th and ^{40}k in the samples collected.

$$I_\gamma = = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \qquad (11)$$

Materials with Iy >1 should be avoided in building construction as such will deliver an effective dose rate higher than $1msvyr^{-1}$ to occupants of such building [17].

Annual Gonadal Equivalent Dose (AGED)

The gonads (organ in which eggs or sperms are produced especially the ovary and testis), the active bone marrow and the bone surface cells are considered as the organs of interest. An increase in Annual Gonadal Equivalent Dose (AGED) has been known to affect the bone marrow, causal destruction of the red blood cells that are then replaced by white blood cells. The AGED (μsvy^{-1}) for the residents of the study area due to specific activities of ^{226}Ra, ^{232}Th and ^{40}k was calculated using the equation given by [19] as :

$$AGED(mSvy^{-1}) = 3.09\,A_{Ra} + 4.18A_{Th} + 0.314A_k \quad (12)$$

Activity Utilization Index (Au)

is a parametric model that enables us to determine the dose ratio in air of Radionuclides (k, Ra and Th) from samples

$$Au = = \frac{A_{Ra}}{500} F_{Ra} + \frac{A_{Th}}{50} F_{Th} + \frac{A_K}{5000} F_K \quad (13)$$

F_{Ra}, F_{Th} and F_k are the fractional conurbations to the total does rate in air due to gamma radiation from the actual concentration as of these radionuclides [20]. The value of F_{Ra}, F_{Th} and F_k are given as 0.462, 0.604 and 0.041 for uranium, thorium and potassium respectively [20] less than 2 corresponds to an annual effective dose of L0.3Amsv/y which is safe for the environment.

4. Results

The results of specific activity concentration of ^{226} Ra, ^{232}Th and ^{40}K in particulate matter samples and their radiological risk parameters from Ishiagy in Eboyi State and Okpella Edo state Quarrying sites are presented in Table 1 to 4 respectively.

Table 1: Specific Activity Concentration of ^{226}Ra, ^{232}Th and ^{40}K in particulate samples from Ishiagu Quarry Sites

Sample Code	^{226}Ra (Bq/l)	^{232}Th (Bq/l)	^{40}K (Bq/l)	Raeq(Bq/l)
Ishi 1	1.91 ± 0.32	1.40 ± 0.15	1.89 ± 0.08	4.06
Ishi 2	6.41 ± 0.78	0.94 ± 0.21	8.16 ± 0.20	8.38
Ishi 3	0.89 ± 0.06	0.63 ± 0.05	1.65 ± 0.07	1.92
Ishi 4	2.22 ±0.36	1.86 ±0.30	7.71 ± 0.19	5.47
Ishi 5	2.91 ± 0.51	2.22 ± 0.40	5.82 ± 0.15	6.53
Ishi 6	5.08 ± 0.72	4.08 ± 0.46	10.87 ± 0.24	11.75
Ishi 7	1.87 ± 0.23	0.84 ± 0.22	4.31 ± 0.15	3.4
Ishi 8	0.76 ± 0.05	0.56 ± 0.06	1.82 ± 0.07	1.7
Mean	2.76 ± 0.38	1.57 ± 0.23	5.29 ± 0.14	5.41
Min	0.76 ± 0.05	0.56 ±0.06	1.65 ± 0.07	1.7
Max	6.41 ± 0.78	4.08 ± 0.46	10.87 ± 0.24	11.75

Table 2: Radiological health Risks estimated in samples from Ishiagu

Sample Area	DR ($nGyh^{-1}$)	AEDE ($mSvy^{-1}$)	AGDE (μsvy^{-1})	AUI	H_{ex}	H_{in}	I_γ	ELCR
Ishi 1	1.81	0.002	12.35	0.02	0.01	0.02	0.05	0.01
Ishi 2	3.87	0.005	26.3	0.03	0.02	0.04	0.14	0.02
Ishi 3	0.86	0.001	5.9	0.02	0.01	0.01	0.03	0.00
Ishi 4	4.13	0.005	17.06	0.04	0.01	0.02	0.07	0.02
Ishi 5	2.93	0.004	20.1	0.05	0.02	0.03	0.08	0.01
Ishi 6	5.26	0.006	36.16	0.10	0.03	0.05	0.15	0.02
Ishi 7	1.55	0.002	10.64	0.03	0.01	0.01	0.05	0.01
Ishi 8	0.77	0.001	5.26	0.01	0	0.01	0.07	00.0
Mean	2.52	0.003	16.72	0.04	0.01	0.02	0.02	0.01
Min	0.77	0.001	5.26	0.01	0.01	0.01	0.02	0.00
Max	5.26	0.006	36.16	0.1	0.03	0.05	0.15	0.02

Table 3: Specific Activity Concentration of ^{226}Ra, ^{232}Th and ^{40}K in particulate samples from Okpella Quarry Sites

Sample Code	^{226}Rq (Bq/l)	^{238Th} (Bq/l)	^{40}K (Bq/l)	Raeq
Okp 1	1.45 ± 0.39	1.68 ± 0.29	6.76 ± 0.18	4.37
Okp 2	2.18 ± 0.48	2.42 ± 0.34	8.06 ± 0.20	6.26
Okp 3	0.53 ± 0.23	0.81 ± 0.21	8.77.65 ± 0.21	2.41
Okp 4	1.25 ±0.31	1.91 ±0.24	7.09 ± 0.19	4.53
Okp 5	1.72 ± 0.12	2.04 ± 0.39	3.20 ± 0.13	4.83
Okp 6	4.61 ± 0.76	4.97 ± 0.43	11.47 ± 0.26	12.6
Okp 7	1.86 ± 0.41	2.44 ± 0.48	3.47 ± 0.10	5.62
Okp 8	1.11 ± 0.78	1.39 ± 0.66	1.13 ± 0.11	3.18
Mean	1.84 ± 0.44	2.21 ± 0.38	6.24 ± 0.20	5.48
Min	0.53 ± 0.23	0.81 ±0.21	1.13 ± 0.25	2.41
Max	4.61 ± 0.76	4.97 ± 0.43	11.47 ± 0.264	12.6
MDA	0.04(Rq)	0.08(Th)	0.44(k)	

Table 4: Radiological health Risks estimated in samples from Okpella

Sampling Area	DR ($nGyh^{-1}$)	AEDE ($mSvy^{-1}$)	AGDE (μSvy^{-1})	AUI	H_{ex}	H_{in}	I_γ	ELCR (x 10-3)
Okp 1	1.96	0.002	13.63	0.03	0.07	0.02	0.03	0.01
Okp 2	2.80	0.003	19.38	0.05	0.01	0.03	0.04	0.01
Okp 3	1.10	0.001	7.78	0.02	0.01	0.01	0.02	0.00
Okp 4	4.13	0.002	14.07	0.04	0.07	0.02	0.03	0.01

Okp 5	2.93	0.003	14.85	0.04	0.01	0.02	0.03	0.01
Okp 6	5.61	0.007	38.62	0.1	0.03	0.05	0.09	0.02
Okp 7	2.48	0.003	17.04	0.05	0.02	0.02	0.04	0.01
Okp 8	1.40	0.002	9.59	0.03	0.01	0.01	0.02	0.01
Mean	2.44	0.003	16.87	0.05	0.02	0.02	0.04	0.01
Min	1.10	0.001	7.78	0.01	0.01	0.01	0.02	0.01
Max	5.61	0.007	36.62	0.05	0.03	0.05	0.09	0.02

5. Discussion

Evaluation of activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in suspended particulate matter collected from two quarry sites have been evaluated using the appropriate instrument. The minimum activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in inhalable particulate matter from Ishiagu quarry site are 0.76±0.05, 0.56±0.06 and 1.65±0.10 Bq/l while the maximum activity concentration values are 6.41 ±0.78, 4.08±0.46 and 10.87±0.24 Bq/l respectively. The minimum activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in inhalable particulate matter from Okpella Bua cement quarry site are 0.53±0.23, 0.81±0.21 and 1.13±0.25 Bq/l while the maximum activity concentration values are 4.61 ±0.76, 4.97±0.43 and 11.47±0.26 Bq/l respectively. The result compared well with results of other similar works [21, 22].

The result of the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the samples from Ishiagu quarry site are comparable with that from Okpella Bua cement quarry site with slight variation. All the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K obtained are higher than the activity concentration obtained in the control samples. This implies that the quarry mining activities slightly impacted on the concentration of radionuclides in those sites, the activities of ^{226}Ra, ^{232}Th and ^{40}K obtained are also within the permissible safe limit.

All the radiation risk parameters estimated were lower than the World safe values of 55 $nGyh^{-1}$, $0.07mSvy^{-1}$, 0.29 x 10^{-3} and 300 $mSvy^{-1}$ for absorbed dose, annual effective dose, excess lifetime cancer risks and annual gonadal dose respectively in all locations studied. Also, all the estimated radiation hazard indices A_{uI}, H_{ex}, H_{in}, 1_{yr}, ELCR (x10-3) were lower than the world means of ≤ 1 for the three locations. These results are similar to the ones obtained in similar work [23]. The results of this study shows that there will be no immediate radiation risk due to inhalable particulate matter within the quarry sites to the workers and the inhabitants of these communities, though long-term effect might be envisaged.

Conclusion

The evaluation of radionuclide content of suspended particulate matter from some quarry sites have been carried out. The result showed that the particulate matter in the quarries sites has low

radionuclide content since the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the suspended particulate matter were lower than the recommended safe values.

The estimated radiological risk parameters of all the sites were lower than their safe values. Hazard indices estimated were lower than 1 which is the recommended safe value. Therefore, quarrying activities in the two areas Okpella and Ishiagu mining sites have not impacted radiologically the air of the communities hosting the companies. Hence, constant radiological screening of the suspended particulate matter SPM is therefore recommended to keep the possible radiation hazard as low as reasonably achievable (ALARA). The result can as well serve as baseline for further radiological studies for the two states.

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