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Original Research Paper

Different Size Aggregates of Stone Quarry Products and Airborne Particles around a Facility in Akure, Southwestern Nigeria: Radioactivity Concentrations, Radiological Hazard and Dose Assessment

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ABSTRACT

This study sets out to verify the hypothesis that size aggregate of quarry rock particles affects the radiation doses. Samples of freshly and previously crushed finished stone dust of different aggregate size were collected in the factory and airborne dusts were collected around quarry facility during October, 2014 to September, 2015. The activity concentrations of ^{40}K, ^{238}U, and ^{232}Th in samples were determined by gamma ray spectrometry. The average mean activity concentrations in fresh samples for ^{238}U, ^{232}Th and ^{40}K were 35.56±5.67, 42.41±5.66 and 1164.17±16.33 Bq/kg respectively. Meanwhile, those of previous samples were 29.93±6.09, 44.87±5.73 and 1087.94±15.87 Bq/kg for ^{238}U, ^{232}Th and ^{40}K respectively. The radium equivalent values for the stone dust samples in this work are higher than the accepted safe limit of 370 Bq/kg . The committed effective dose via inhalation of dust ranged from 0.450 to 0.471 μSv. It was concluded that prolong inhalation of dust in the area could pose health risk to individual.

INTRODUCTION

In recent time, assessing the consequential risks of dust has been subject of concern to environmentalists due to hazards created by its suspension. Quarry activities introduce fine pulverized materials as well as particulate matters into the atmosphere (ISO 1995). The dust particles dispersed into the atmosphere where they eventually gain access to respiratory and circulatory systems (WHO 1997, Lippmann 1977, Liu et al. 2006) even at far distance from source. From literatures, suspended dust samples in and around mining sites are known to contain elevated levels of heavy metals and radioactive isotopes of natural and technogenic origins (Hamadneh et al. 2015, Ademola et al. 2014, Bollhofer et al. 2006). In a study by Hamadneh et al. (2015) on the radioactivity of seasonal dust in the Middle East country of Jordan, it was observed that large amount of radionuclides are deposited to soil through dust storm. Previous studies (Straden 1979, UNSCEAR 1988, Fasasi et al. 1999) have also shown that primordial radionuclides (^{40}K, ^{238}U, and ^{232}Th series) are widely spread in soil and rocks of the earth's crust and they are the major contributors to external exposures from γ-radiation in the environment. Acute inhalations of radionuclides have been reported in animals to cause somatic and genetic effect that tend to damage critical and/or radiosensitive organs of the body, which ultimately can lead to death (Srinivasa et al. 2015, ATSDR 1999, Ajayi 1999).

In Nigeria, several studies have reported the radioactivity measurements in building materials (Fasae 2011) and mining sites (Shittu et al. 2015, Enyinna & Onwuka 2014, Gadebo et al. 2011). Most of these studies, reported high radiation levels in surface soils where granite rocks and ornamental stones exist. According to Shittu et al. (2015), radium equivalent for the granite rocks are above the internationally accepted value of 370Bq.kg^{-1} in Abuja, Nigeria. However, none of these studies investigated the variation of activity concentrations with different aggregate size of the dust particles. In southwestern part of the country, where this study was carried out, there are vast resources of dimension stones such as granites and other ornamental stones with high quality, thereby making them suitable for quarry and processing (Ajayi & Ajayi 1999, Fasae 2011).

The three reasons have aroused our interest in this work are that: (1) most of these quarry sites are located in the vicinity of villages and towns and the relative high exposure to dust pollution in these areas especially during the

production peak of these factories is enough to justify the assessment of air contamination levels resulting from the quarry activities. (2) the products of this quarry factories are used in construction industries, and (3) despite the above, the appropriate data describing doses incurred from dust inhalation by population living in the vicinity of stone mining sites is unavailable hence assessment of radiological hazard risks and consideration of mitigation measures is impossible.

Therefore, this study was set out to evaluate the distribution of ^{238}U, ^{232}Th and ^{40}K and to estimate radiological risk likely to be exposed by inhabitants living in the vicinity through the airborne dust generated from the quarries activities. The objectives of the study include among others: to establish a background data on the radiation profile of this area and to assess the radiological impact of these nuclides on the health of the people living in the surrounding community. It is expected that the data generate will provide the distribution pattern of natural radionuclides activities of ^{238}U, ^{232}Th, and ^{40}K in the airborne dust generated from quarries to the surroundings, and would provide adequate information for environmental policy makers on the levels of radionuclides in the environment.

MATERIALS AND METHODS

Study areas: Akure lies within Longitudes 05°00"E and 05°17"E and Latitudes 07°10"N and 07°20"N in the southwestern part of Nigeria. According to Shitta (2007), the three types of charnockitic rocks in Akure area on the bases of their textural characteristics can be described as: (1) coarse -grained as exemplified by Akure body, (2) massive fine-grained which form along the margins of the granitic bodies as seen in Ijare, Uro and Idemo, (3) the gneissic fine-grained which were recognized within the bodies of gneissic in Ilara and Iju.

The stone crushing facilities used for this study were located along Akure-Owo/Benin road at the outskirt of Akure. The location of the facilities is on a relatively high elevation with values in the range of 350 and 380 meters. Located around the facilities are: communities of people, vegetation and public buildings such as schools and markets.

Sample collection: Samples of freshly and previously crushed finished stone dust of different aggregate size were collected in the factory. Airborne dust samples were collected around the quarrying facilities twice in a week, during different days of the week to obtained representative sample for six months of October, 2014 to September, 2015. Airborne dust samples at the premise and different locations in the perimeters of the plant were collected using active sampling method and sample sites denoted as P1 to P5. The sampling method used the filter holder (AEA Technology filter holder, Casella London Limited), where a known volume of air is drawn through a pre-weighted glass fibre filter paper (25mm diameter, pore size 80μm) by means of an air pump. The filter holder was placed in a static position at approximately 1.6m high (breathing zone). The flow rate of the pump was set at 2L/min and the pump was allowed to run for 8 hours. The dust concentration was determined us-

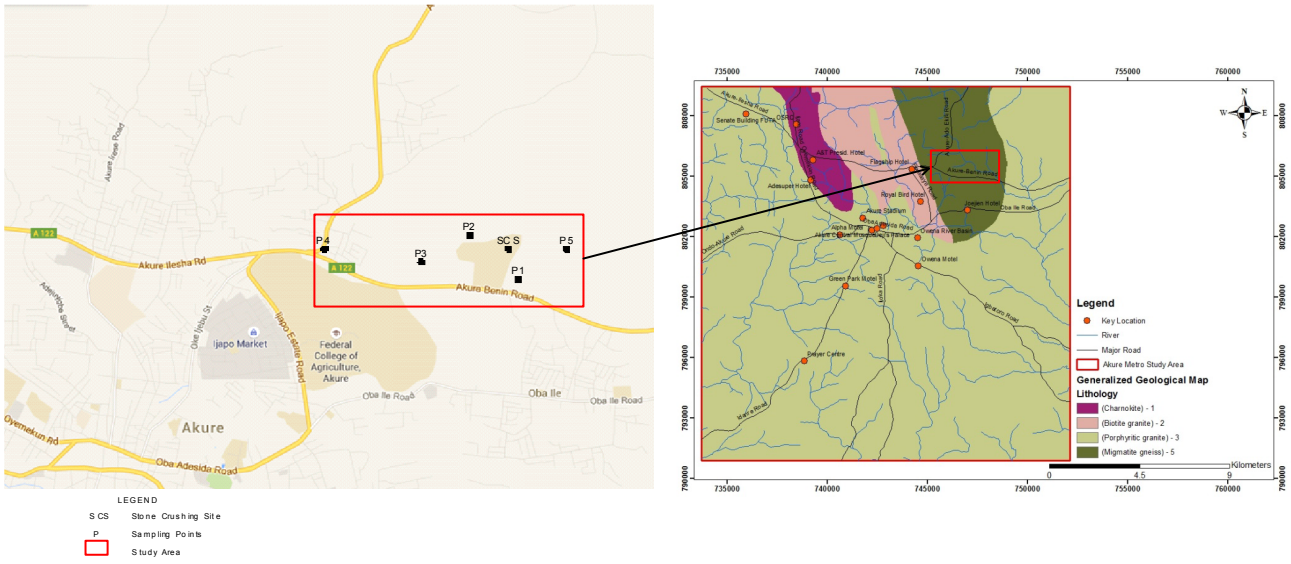

Fig. 1: Sampling sites around the studied facility and generalized geology of the area around Akure metropolis (Source: Nigerian Geological Survey Agency Map, 1966).

ing equation (1).

$$\text{Dust concentration (g/m}^3) = 100A/vt \quad ...(1)$$

Where, A is the volume of air; v is the flow rate of the pump (2L/min); t is the duration of experiment (465min).

Sample analysis: The activities of the investigated radionuclides in the stone dusts produced from the plants were determined by direct gamma spectrometry, using a high purity germanium (HPGe) detector, with an efficiency of 30% relative to a 33" × 33" NaI (Tl) scintillator and an energy resolution (FWHM) of 1.8 keV for the 1.33 MeV reference transition of ^{60}Co, was utilized for the measurements. The detection of gamma radiation is used to determine the concentration of the ^{40}K, ^{238}U and ^{232}Th.

The polyethylene counting vessels (Marinelli beakers) were sealed gas-tight and stored for 4 weeks to allow radioactive equilibrium of the uranium and thorium series depending on the assumption of secular equilibrium, where the rate of decay of the daughters becomes equal to the rate of decay of the parent. The prepared Marinelli beakers were placed on the detector endcap.

Both the sample and the detector were surrounded by a cylindrical graded-Z shield of 5 cm thickness of lead, 1cm thickness of Iron and 1cm thickness of aluminium to suppress the background radiation.

A spectroscopic amplifier, with an efficient pile-up rejecter, and an 8k ADC (Analog-to-Digital Converter) processed the signal. The MAESTRO-32 multi-channel analyser emulation software was used for data acquisition, storage, display and online analysis of the spectra. Measurements with an empty Marinelli beaker, under identical conditions, were also carried out to determine the ambient background in the laboratory site. The latter was subtracted from the measured spectra to obtain the net radionuclide activities.

The gamma lines; 351.9 keV of ^{214}Pb, 609.3 keV of ^{214}Bi, 1120.3 keV of ^{214}Bi and 1674.5 keV of ^{214}Bi were used for determining ^{238}U. The gamma lines 338.34 keV of ^{228}Ac, 583.0 keV of ^{208}Tl, and 911.1 keV of ^{208}Ac were used for determining ^{232}Th series, while ^{40}K was measured by its gamma line 1460.7 keV. The uncertainty in the calculated efficiency was estimated to be 5 %.

Radiological Hazard Indices: ***Radium equivalent*:** The exposure radiation defined as radium equivalent activity (Ra_{eq}) was calculated according to (Beretka & Mathew 1985) given in equation (2)

$$Ra_{eq} = C_U + 1.43C_{Th} + 0.077C_k \quad ...(2)$$

where C_U, C_{Th} and C_k (^{238}U, ^{232}Th and ^{40}K) are the concentration o f(^{238}U, ^{232}Th and ^{40}K) in Bq.kg^{-1} respectively.

***Internal and external hazard indices (H_{int} and H_{ext})*:** The internal hazard index (H_{int}) gives the internal exposure to carcinogenic radon. It is calculated as given by equation (3) (Beretka & Mathew 1985).

$$H_{int} = \frac{C_U}{4810} + \frac{C_{Th}}{185} + \frac{C_k}{259} \quad ...(3)$$

The external hazard index is an evaluation of the hazard of the natural gamma radiation and calculated by equation (4).

$$H_{ext} = \frac{C_U}{4810} + \frac{C_{Th}}{370} + \frac{C_k}{259} \quad ...(4)$$

Where C_U, C_{Th} and C_k (^{238}U, ^{232}Th and ^{40}K) are activity concentration of (^{238}U, ^{232}Th and ^{40}K) in Bq.kg^{-1} respectively.

Dose Parameters: ***Dose rate in dust*:** The absorbed dose rate in dust samples was calculated according to equation (5) (UNSCEAR 2008)

$$D\ (nGyh^{-1}) = 0.461C_U + 0.623C_{Th} + 0.0414C_K \quad ...(5)$$

Where, D is absorbed gamma dose rates, C_U, C_{Th}, C_K represent the activity concentrations of ^{238}U, ^{232}Th and ^{40}K in Bq.kg^{-1} respectively.

Calculation of committed effective dose equivalent: The committed effective dose from inhalation of contaminated dust was calculated using the formula: (ICRP) (1994)

$$D_{inh} = t_{exp} \times V \times \Sigma (g_{inh,r} \times C_r) \quad ...(6)$$

Where t_{exp} is duration of exposure (assumed to be 100 h/a), V is the breathing rate (1.2 m^3/h).

$g_{inh,r}$ is the inhalation dose coefficient (Sv/Bq) for radionuclide r and Cr is the ambient air activity concentration (Bq/ m^3) for radionuclide r.

Statistical Analysis: In order to test the significance of the interaction between the mean values of activity concentrations obtained at different locations, analysis of variance (ANOVA) statistical test was used. The significant probability of the test is controlled at specific level ($p<0.05$). The least partial regression between ambient dust concentration and dose rate was carried out using the analysis tools of SPSS statistical package.

RESULTS AND DISCUSSION

Activity of Radioisotopes in the finished products: Blasting and crushing of stones generate particles of different size aggregates which are dispersed into the atmosphere. It has been hypothesized that aggregates size distribution is likely to have effects on radiation (Gadebo et al. 2011). To test this hypothesis, samples of different aggregate sizes of fin-

Table 1: Mean activity concentrations (in Bq/kg) and radiological indices of the radionuclides in different aggregates of finished stone products and dust samples around the plants.

Sample ID	Aggregate size	^{238}UBq/kg	^{232}ThBq/kg	^{40}KBq/kg	Hext	Hint
Freshly crushed sample	<1/2 inch	30.19 ±3.02	39.04 ±4.25	1166.8 ±13.66	0.474	0.556
	1/2 inch	38.33 ±6.09	40.07 ±6.98	1128.62 ±17.10	0.492	0.596
	3/4 inch	38.15 ±4.21	48.11 ±5.99	1197.1 ±15.83	0.537	0.640
Average Mean		35.56 ±5.67	42.40 ±5.66	1164.17±16.33	0.501	0.598
Previous Sample	<1/2 inch	28.31 ±4.44	44.67 ±6.15	1043.7 ±16.90	0.511	0.612
	1/2 inch	30.63 ±6.21	44.89 ±5.51	1109.8 ±14.83	0.517	0.617
	3/4 inch	30.85 ±5.80	45.06 ±5.07	1110.32 ±13.86	0.509	0.610
Average Mean		29.93 ±6.09	44.873 ±5.73	1087.94 ±15.87	0.512	0.613

Table 2: Mean activity concentrations (in Bq/kg) and Radiological indices of the radionuclides in dust samples around the plants.

Sample ID	Coordinate from the facility	Distance	^{238}UBq/kg	^{232}ThBq/kg	^{40}KBq/kg	Hext	Hint
P1	O7^0 16.600"N005^0 14.282"E	50m	16.82±4.09	19.17±5.11	921.44±13.84	0.311	0.356
P2	O7^0 16.594"N005^0 14.601"E	60m	16.02±3.08	19.04±5.11	901.08±14.64	0.304	0.347
P3	O7^0 16.676"N005^0 13.680"E	80m	15.00±2.76	17.02±4.42	913.04±16.17	0.296	0.336
P4	O7^0 16.857"N005^0 13.327"E	1.2km	14.8±3.65	14.64±5.42	913.98±15.42	0.286	0.326
P5	O7^0 16.894"N005^0 13.226"E	1.0km	15.99±2.99	16.43±5.19	913.26±17.36	0.296	0.339
Mean			15.72±3.16	17.26±5.91	912.56±15.87	0.298	0.341

Table 3: Comparison of activity concentrations in this study with other studies in Nigeria and worldwide.

Country	^{238}UBq/kg	^{232}ThBq/kg	^{40}KBq/kg	Reference
Akure, Nigeria	25.43±6.1	27.45±4.2	1015.96±10.3	This study
Obajana, Nigeria	331.25±132.52	0±31.00	2189.75±219.5	Ajayi et al. 2012
Abuja, Nigeria	74.74±5.67	199±43.30	1021±7.14	Shittu et al. 2014
El-Minya, Egypt	25.3±3.0 – 68.0±0.9	37.0±0.4 – 88.0±2.0	48.0±11.0 – 820. 0±13.0	Ibrahim et al. 2014
Germany	ND	70±6.5	1465.4±106.8	Ahmed et al. 2006
World wide	33	45	412	UNSEAR 2010

ND = Not determined

Table 4: Table showing the dose indices of the radionuclides in the dust samples.

Site ID	Ambient dust concentration(g/m^3)			Dose rate (Gyh^{-1})	Committed Effective Dose(μSv)
	^{238}U	^{232}Th	^{40}K		
SCS	9.93±3.80	4.87±2.13	8.93±3.19	92.34	0.472
P1	2.093±1.05	7.066±3.19	3.332±2.35	57.77	0.073
P2	2.011±1.36	6.331±2.72	3.339±2.10	56.48	0.065
P3	1.934±1.14	7.802±3.66	3.322±2.80	55.28	0.079
P4	2.012±1.02	6.574±2.09	3.342±2.43	53.79	0.068
P5	0.645±0.11	2.438±0.91	1.978±0.99	55.39	0.025
Mean	1.739±2.78	6.042±2.32	3.062±2.63	55.74	0.062

ished products by the factories were collected and the activity concentrations were determined. The results of mean activity measurements in different aggregate size of finished products are presented in Table 1. Although, high activity concentrations of radioisotopes have been observed in building materials and attributed to geological rocks formation (Fasae 2011). It can be seen from the results that activity concentrations in fresh samples for ^{238}U, ^{232}Th and ^{40}K ranged

from 30.19± 3.02 to 38.15± 4.21 Bq.kg^{-1}, 39.04 ±4.25 to 48.11± 5.99Bq.kg^{-1}, 1128.62±17.10 to 1197.10±15.53 Bq.kg^{-1} respectively, with average mean activity of 35.56±5.67, 42.41±5.66 and 1164.17±16.33 Bq.kg^{-1} respectively. Meanwhile, the activity concentrations in previous samples for ^{238}U, ^{232}Th and ^{40}K ranged from 28.31±4.44 to 30.85±5.80 Bq..kg-1, 44.67±6.15 to 45.06±5.07 Bq.kg-1, 1043.70±16.90 to 1110.32±13.86 Bq.kg-1 respectively, with average mean activity of 29.93±6.09, 44.87±5.73 and 1087.94±15.87 Bq.kg^{-1} respectively. This is highest values in both products for ^{238}U, ^{232}Th and ^{40}K were observed for ½ inch and ¾ aggregates. The result showed that finest (<1/2inch) quarry dust has the least activity. Generally, the mean activities of radioisotopes in the freshly crushed products are higher than the worldwide values of 33, 45 and 412Bq.kg^{-1} for ^{238}U, ^{232}Th and ^{40}K respectively, (UNSEAR 2010). However, the values of mean activity concentrations in previous finished products are lower than the worldwide values. The results of this study confirm that the aggregate sizes affect the activity concentrations of radioisotopes and that freshly quarry rocks possess high activity than those from previously quarry rocks.

Activity of Radionuclides in the atmospheric dust samples: The values of mean average activity concentrations (Bq.kg^{-1}) for ^{238}U, ^{232}Th and ^{40}K dust samples in five sites (P1 -P5) in the vicinity of the facility as measured by gamma-ray are presented in Table 2. The activity concentrations of the radionuclides species varied spatially in dust samples, although the variation is not significant ($p>0.05$). In all the sampled sites, the mean activity concentrations are in order ^{238}U < ^{232}Th < ^{40}K. This observed order agrees well with earlier report by Ademola et al. (2014) in soil samples of southwestern Nigeria gold mine site of Itagun. The activity concentration values in the investigated dust samples ranged from 14.8± to 16.82± Bq.kg^{-1}, with mean value of 15.72 ±3.16 Bq Bq.kg^{-1} for ^{238}U. The highest value (16.82 Bq.kg^{-1}) was detected at P1, a distance of 50 meters away from the emission source while the lowest (14.8 Bq.kg^{-1}) was observed at P4, a distance of 1.2km from the source; ^{232}Th has the mean value of 17.26 ±5.91 Bq.kg-1 with the highest value (19.17 Bq.kg^{-1}) at P1 and the lowest (14.64) at P4 ; ^{40}K ranged between 901.08 and 921.44 Bq.kg^{-1} with mean value of 912.56±15.87 Bq.kg^{-1}.

From the results, it can be observed that activity concentrations are higher at sites (P1) than others sites. This has been attributed to proximity of these sites to the facilities and the high density dust generated from the stone processing. Generally, it can be observed that the activity concentrations of radioisotopes in dust samples are significantly ($p<0.05$) lower than the values in finished products. This may be explained as effects of materials processing via mechanical, chemical or thermal treatments which can disrupt radioactive equilibrium and that of residence time on the activity, as the dust must have spent some times before eventually suspended (Yu et al. 1992).

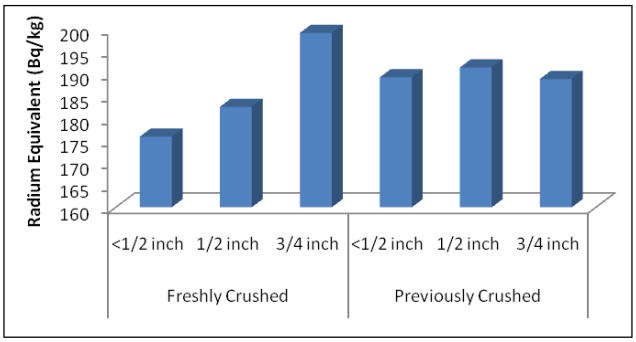

Fig. 2: Plot of radium eqivalent against different size aggregrates.

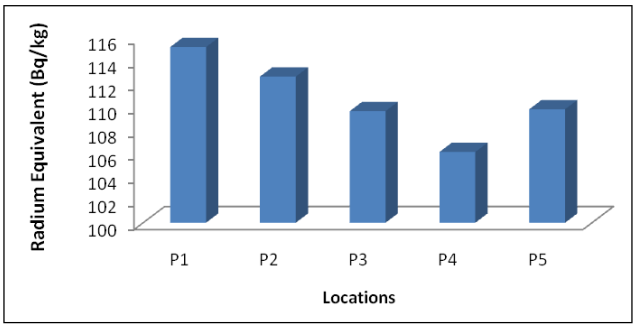

Fig. 3: Plot of radium eqivalent at various locations of the sampling sites.

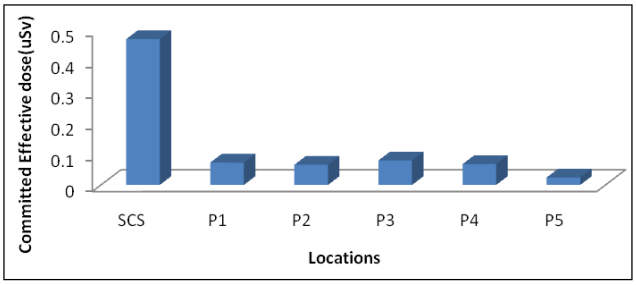

Fig. 4: Plot of committes effective dose at the various studied sampling sites.

A comparison of the average activity concentrations of radioisotopes in the present study with those of granite rocks in Abuja, North central, Nigeria and other studies worldwide (Table 3). The average concentrations of 74.74± 5.67, 199 ±43.30, 1021 ±7.14 for ^{238}U, ^{232}Th and ^{40}K respectively in Abuja are higher than our study. However, the values are comparable to ranges of values of 25.3± 3.0-68.0± 0.9 for ^{238}U, 37.0 ±0.4-88.0± 2.0 for ^{232}Th, and 48.0 ±11.0-820. 0±

13.0 for ^{40}K obtained for quarry raw material in El-Minya, Egypt (Ibrahim et al. 2014). This may be attributed to differences in nature of geological formation in these areas. It is worthy to note that the mean activity concentrations in the studied sites were lower than the world mean activity concentrations of 33 and 45 Bq.kg^{-1} for ^{238}U and ^{232}Th, respectively except for ^{40}K with the value of 412 Bq.kg^{-1} (UNSEAR 2010).

Radiation parameters: It has been reported that fine grain aggregates easily allow for escape of radiation. To verify this assumption, we hypothesized that if this would occur the radium equivalent of fine grains would decrease with increase in distance from the emission source. The radiation hazard indices in different aggregate size and those of dust samples are presented in Figs. 2 and 3. It can be observed that the radium equivalent calculated for different aggregates of finished samples ranged 175.86 to 199.12 Bq.kg-1. The lowest radium equivalent was calculated for fine aggregate in both freshly and previously quarry finished products. This observation may be attributed to high radiation associated with granitic and silicic igneous rocks present in the study facilities area (Brimhal & Adams 1982).

Furthermore, the values of Req ranged from 138.112 to 147.184 Bq.kg^{-1} in dust samples, with mean value of 137.823 Bq.kg^{-1} in the studied area. The highest value was observed around the facility premises and the lowest at sites far from the plant. This may be attributed to escape of radiation from the grain as they were transported by wind from the source. It goes further to confirm that fine grain aggregates allow the easy escape of radiation than coarse aggregates, which are easily settled or suspended around the facility or emission sources. The mean value from the dust in this study is greater than the safe limit of 370 Bq.kg^{-1} (UNSCEAR 2000).

The external hazard index is an evaluation of the hazard of the natural gamma radiation exposed by public. The results of calculated external hazard index (H_{ext}) values for dust samples are also presented in Table 2. The mean external hazard index ranged from 0.386-0.396 with a value of 0.736 at the premises of the facilities. This was closely followed by the market with the value 0.672. The results showed that radiation effects can be felt within few meters of radius away from the quarry sites. The H_{ext} values for the finished products and dust samples were less than unity. This is an indication that none of the radioisotopes in this study is considered the major source of external radiation. However, the values of H_{int} are greater than 1 which is an indication that prolong stay around the facility may pose a health hazard.

Dose Assessments: Inhalation is the main route of intake therefore characterization of intake in term of aerosol size and absorption is important in dose assessment. Aerosol size influences deposition in the HTRM and the transfer of unabsorbed particle to the gonad track. One of the objectives of this study is to assess the dose incurred via inhalation of dust by population in the area. To achieve this, we collected airborne dust samples at the premise and different locations in the perimeters of the plant. The dose rate and committed effective dose via inhalation for the member of public per year was calculated as described by IAEA (1996) and the results are presented in Table 4. From Table 4, the absorbed dose rate values for this study ranged from 68.581nGyh^{-1} to75.89 nGyh^{-1}. The highest value was found at P3 and lowest at P1. The mean values of 73.305 nGyh^{-1} in this study is higher than the occupational limit of 20nGyh^{-1} (ICRP 1994). The values from this study were extraneously higher than 10.6 nGyh^{-1} obtained in *in situ* measurements in the mining area of Jos Plateau. (Ademola 2008). This may be attributed to difference in the nature of geological formation in these areas.

The obtained values of committed effective dose via inhalation of dust over the working half year of this study within the plant (SCS1 and 2) ranged from 0.450 to 0.471 μSv as presented in Fig. 2. The values of committed effective dose are less than 1μSv and therefore insignificant. However, the values at sampling sites within the plant are closer to unity, this may signifies that prolong stay around the facility may pose a health hazard. Meanwhile, the values around the plant (P1-P5) ranged between 0.0249 and 0.079mSvy^{-1}

CONCLUSION

The study confirmed that the distribution of radionuclides in dust particles in the studied area is influenced by aggregate sizes and the nature of parent geological materials. From environmental point of view, results indicated that the dust arisen from these quarries have human impacts because the radionuclide levels are above the standard absorption rate. There are some potential risk of such industry having an effect on the environment, which requires attention, mitigations, and management to protect the existing human health.

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