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Assessment of radiological hazard of quarry products from southwest Nigeria

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ABSTRACT

The term "quarry products" comprises different natural rocks with different mineral contents, crushed into various sizes at quarries. The concentrations of the naturally occurring radionuclides ^{40}K, ^{238}U and ^{232}Th in quarry products from southwest Nigeria have been measured using gamma ray spectrometry. The gamma absorbed dose rate (D_R), radium equivalent activity (Ra_{eq}), annual effective dose (A_d), activity concentration index (I), external radiation index (H_{ex}) and internal radiation hazard index (H_{in}) associated with the radionuclides are evaluated in order to assess the radiation hazard of quarry products used as building materials. The results showed that a few of the calculated radiological parameters are higher than permissible limit, hence, may pose a radiological hazard when used as building materials.

All the radiological variables above were subjected to correlation analysis to determine the similarities and correlations among various samples. The data sets consist of 10 measured variables. The principal component Analysis (PCA) yields a two component representation of the acquired data, in which 93.3% of the total variance is explained.

1. Introduction

The natural radioelements uranium, thorium and potassium are lithophile elements widely distributed in crustal rocks being concentrated preferentially in acid igneous rocks compared with intermediate, basic and ultra-basic varieties (Alnour et al., 2012; Moura, Artur, Bonotto, Guedes, & Martinelli, 2011). Potassium is a major element widely distributed in crustal rocks, for instance, calcium rich granites may contain up to 2.5% of potassium (Cox, 1991). Thorium occurs predominantly as a tetravalent cation and as a trace constituent in phosphates, simple and multiple oxides and silicates, as well in the major rock-forming minerals such as monazite, thorianite (ThO_2) and thorite ($ThSiO_4$) among others. While Uranium is found in rocks of different mineral species (like apatite, sphene and zircon) as a secondary/accessory mineral or it can form its own minerals. Uranium distribution in rocks is linked to isomorphous mineral substitution, adsorption or inclusion process (Pertlik, Roger, & Adams, 1974). Biotite ("black mica") contains between 19% and 22% of the total uranium because it may contain inclusions of minerals rich in this element, such as zircon. Heavy minerals

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such as zircon, monazite, apatite, magnetite, ilmenite and riebeckite, contain between 61% and 65% of the uranium in a rock (Brown & Silver, 1955; Gabelman, 1977; Gascoyne, M., 1992; Larsen & Phair, 1954; Moreira-Nordemann, 1977; Speeer, Solberg, & Becker, 1981; Tieh & Ledger, 1981).

The presence of naturally occurring radionuclides in construction materials originating from quarry products offers radiation exposure both inside and outside the building environments. This is mainly due to gamma radiation of ^{40}K and members of the uranium and thorium decay series. The term "quarry products" comprises a wide number of different natural rocks with different mineral contents, crushed into various sizes at quarries. This include different geological materials such as gneiss, granite, diorite, granodiorite and other rocks that after an industrial process are suitable for use as building material and ornamental rocks (Ministry of Energy, British Columbia 2014).

Granodiorite is an intermediate coloured, medium to coarse-grained intrusive rock. It falls between granite and quartz diorite, containing more dark minerals than granite but less than quartz diorite. Granite is a pink or greyish coloured rock and represents the lightest coloured variety of intrusive rock. It is medium to coarse-grained and evenly granular. The grains are mainly white to pink orthoclase feldspar with lesser amounts of white to grey plagioclase feldspar and quartz. Diorite is a medium to dark grey, medium to coarse-grained intrusive rock. Gneiss is a medium to coarse-grained, banded, granular metamorphic rock. Distinct colour bands or streaks are produced by the alternation of layers of light (e.g. quartz and feldspar) and dark-coloured (e.g. biotite and hornblende) minerals. The layers do not split readily and when broken are not smooth. Gneiss may form from diorite, granite, shale, sandstone, schist or other rocks (Ministry of Energy, British Columbia 2014). In view of the attractive appearance of these rocks, there is an increasing trend of the public for their use in flooring and interior decoration of building. The use of building material from quarry products containing enhanced gamma activity of naturally occurring radionuclides can pose a radiological hazard to the occupants of such buildings.

Quarry products have been extensively used in Nigeria as building materials. In general, granites are widely recognized to exhibit high levels of uranium, thorium and potassium due to the characteristics of the genetic magma and associated tectonic environment. The knowledge of the level of natural radioactivity in building material is of great importance to determine the associated radiological hazards to human health, to develop reference data of radiological parameters in building material and to develop standards for the use of these materials. This work aims to contribute to a better understanding of the radioactivity distribution in quarry product from selected quarry sites in southwest Nigeria, using gamma spectrometry.

2. Materials and methods

2.1. Sample collection and preparation

A total of fifty (50) samples were collected from ten (10) different quarry sites (five samples from each site). These sites spread across five states (Ogun, Oyo, Osun, Ondo and Ekiti) in which major quarry activities take place in Southwest Nigeria. The coordinates of these quarry sites are listed in Table 1. Each sample was sealed in a well labelled polythene bags and taken to gamma laboratory at Centre for Energy Research and Development (CERD), Obafemi Awolowo University, Ile-Ife for analysis preparation. Wet samples were air dried at room temperature to constant weight and all dried samples were crushed and grinded with Rocklab ring mill, after which they were sieved with a 2-mm mesh and weighed using the OHAUS Adventure Pro AV264 digital balance. Two hundred grams (200 g) each of the samples was weighed into cylindrical polyvinylchloride containers sealed and kept for 28 days in order to attain secular equilibrium between the parent and the daughter nuclides present.

2.2. Gamma ray spectroscopic technique

The activity concentration of natural radioactivity in the samples were determined using a 7.62 cm × 7.62 cm NaI (Tl) detector employed with adequate lead shielding which reduced the background by a factor of about 95%. Energy calibration was done using Standard sources, of known gamma-ray energies and activities, prepared by the Isotope Products Laboratories, Burbank California, USA. The calibration provided qualitative and quantitative analysis of the radionuclides present in each sample. Counting was done for 25,200 s for each of the sample, the calibrated reference material and an empty container were used to determine the background. Current decay data for nuclides were obtained from literature (Arzu, Marie-Martine, Edgardo, & Valery, 2010, 2011). The activities of various radionuclides were determined in Bq kg^{-1} using the count spectra obtained from each of the samples. The gamma ray photo peaks corresponding to energy of 1120.3 keV (^{214}Bi), 911.21 keV (^{228}Ac) and 1460.82 keV (^{40}K) were considered to determine the activity of ^{238}U, ^{232}Th and ^{40}K. The detection limits of the NaI (Tl) detector system were calculated as 31.57, 5.73 and 0.26 Bq kg^{-1} for ^{40}K, ^{232}Th and ^{238}U respectively for a counting time of 25,200 s.

3. Result and discussion

3.1. Activity concentrations of ^{238}U, ^{232}Th and ^{40}K

The specific activity concentration of ^{232}Th, ^{238}U and ^{40}K in the samples are presented in Table 2. The results showed that

Table 1 – Sample ID and coordinate.

Sample ID	Coordinates
OGA	N7° 23′ 30″, E 3° 39′ 16″
OGB	N7° 25′ 47″, E 3° 45′ 09″
OGC	N7° 14′ 51″, E 3° 29′ 32″
OGD	N7° 40′ 29″, E 5° 05′ 59″
OGE	N7° 15′ 25″, E 3° 31′ 38″
OGF	N7° 15′ 32″, E 3° 32′ 19″
OGG	N7° 19′ 22″, E 3° 37′ 36″
OGH	N7° 16′ 50″, E 3° 50′ 47″
OGI	N7° 12′ 59″, E 3° 48′ 53″
OGJ	N7° 20′ 46″, E 5° 15′ 00″