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Research Article

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Radiological Assessment of Coal Samples from Selected Coal Mines in Nigeria

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Abstract Representative samples of coal from Okaba, Kogi State, Enugu, Enugu State, Garin Maiganga, Plateau State, and Gindi Akwati, Gombe State were collected and analyzed for possible radioactivity using Sodium Iodide Detector (NaI). The results shows that coal samples from Okaba coal, Kogi State has 70.622Bq/kg of K-40, 34.183Bq/kg of Ra-226 and 18.826Bq/kg of Th-232; Enugu, Enugu State has 90.669Bq/kg of K-40, 31.518Bq/kg of Ra-226 and 26.910Bq/kg of Th-232, Garin, Maiganga coal Plateau State has 117.325Bq/kg of K-40, 32.213Bq/kg of Ra-226 and 46.636Bq/kg of Th-232, Gindi, AkwatiGombe State has 25.505Bq/kg of K-40, 29.085Bq/kg of Ra-226 and 14.367Bq/kg of Th-232. The results showed that the radioactivity dosage from the sample were below the World Average Natural Dose which is 370Bq/kg per year. The rank profile of radioactivity in the coal samples are, for K-40; Garin Maigang>Enugu>Okaba>Gindi Akwati, for Ra-226; Okaba>Garin Maiganga>Enugu>Gindi Akwati, for Th-232; Garin Maiganga>Enugu>Okaba>Gindi Akwati. The determined radioactivties and the concentration of the various samples from the selected location are within the permissible limits.

Keywords Radioactivity, Radiological Assessment, Coal

Introduction

Coal, like most material found in nature contains trace quantities of naturally occurring radionuclides NORM. (Uranium and thorium families and potassium-40). Therefore, the combustion of coal results in release of radionuclides and non-combustible mineral matter, containing bottom ash and fly ash, which are eventually release into the environment. Emissions from thermal power stations in gaseous and particulate form contain radioisotopes arising from the uranium and thorium series as well as from Potassium-40 (^{40}K). They are discharged into the environment causing changes in the natural radiation background and radiation exposures to the population. The continual release of these materials to environment may result in a buildup in the air, water and soil of radionuclides, particularly radium-226. Consequently, there will be an increase of the basic radiation rate in the neighborhood area of plants and higher exposure of the local population. Coal burning is, therefore, one of the sources of technologically enhanced natural radionuclides materials (TENORM) [1].

Coal is largely composed of organic matter, but it is the inorganic matter in coal minerals and trace elements that have been cited as possible causes of environmental health, and technological problems. Some trace elements in coal are naturally radioactive. These radioactive elements include uranium (U), thorium (Th), and their numerous decay products, including radium (Ra) and radon (Rn). Although these elements are less chemically toxic than other coal constituents such as arsenic, selenium and mercury. In order to accurately predict the mobility of radioactive elements during the coal fuel-cycle, it is important to determine the concentration, distribution, and form of radioactive elements in coal and fly ash [2]. Assessment of the radiation exposure from coal burning is critically dependent on the concentration of radioactive elements and in the fly ash that remains after combustion [2].

Radon, which is a nuclide of public health concern, has been found to be an ubiquitous air pollutant to which most population are exposed. (^{222}Rn), a progeny of ^{238}U formed from the radioactive decay of radium emits alpha radiation and decays with a half-life of 3.824 days [3]. Radon is present in trace amounts almost everywhere (indoor and outdoor) on the earth, being distributed in the soil, the ground water and in the lower atmosphere. When radon decays to form its progeny (^{218}Po and ^{214}Po), they are electrically charged and can attach themselves to tiny dust particles, water vapor, oxygen, trace gases in indoor air and other solid surfaces. These daughter products remain air borne for a long time. These dust particles (aerosols) can easily be inhaled into the lung and can adhere to the epithelial lining of the lung, thereby irradiating the tissue. Bronchial stem cells and secretion cells in airways are considered to be the main target cells for the induction of lung cancer resulting from radon exposure. The deposited radioactive daughter nuclides decay emitting alpha particles, beta particles and gamma rays respectively. The longer range and lower biological effectiveness of beta particles, gamma rays, have made their dose equivalent to lung tissue very negligible. Alpha radiation being densely ionizing has the potential to damage the DNA of the lung cells. This DNA damage has the potential to initiate chain of events leading to lung cancer. The damage being localized to lung cells, the lung cancer becomes the only potential hazard posed by radon in indoor air. It has been estimated that the radon, largely in homes, constitutes more than 50% of the dose equivalent received by general population from all sources of radiation, both naturally occurring and man-made [4]. It is well known that exposure of population to high concentrations of radon and its daughter nuclides for a long period lead to pathological effects like the respiratory functional changes and the occurrence of lung cancer [5]. Measurement of Radon (^{222}Rn) and its progeny Concentration is fundamental in the environment of thermal power plants, as it is expected to be high due to burning of lot of coal [5]. In this study, the radiological assessment of the coal sample from Okaba, Kogi State, Enugu North, Enugu State, Garin Maiganga, Plateau State, and Gindi Akwati, Gombe State is undertaken to educate the populace around the study area on possible radioactive pollution as a result of domestic and industrial usage.

Materials and Methods

Description of the study locations:

The sampling sites are shown in fig. 1 below.

a) Okaba Coal: Okaba district lies some 16km NE of Ankpa town, headquarters of Ankpa Local Government Area, Kogi State. The study area is located between latitudes $7^0 20^1$- $7^0 43^1$N of the Equator and longitudes $7^0 22^1$ - $7^0 52^1$E of the Prime Meridian. The area is within the tropical hinterland. Annual rainfall totals range between 100-200 cm, spread over 6-8 months.

b) Barakinladi Coal: BarakinLadi is a populated place in Plateau State, Nigeria (Africa) with the region font code of Africa/Middle East. It is located at an elevation of 1,315 meters above sea level and its population amounts to 71,657. Its coordinates are 9°31'60" N and 8°54'0" E in DMS (Degrees Minutes Seconds) or 9.53333 and 8.9 (in decimal degrees).

c) Maiganga Coal: Maiganga Coal deposit is located at Maiganga village in Akko Local Government Area of Gombe state. The Maiganga coal mine is located at 8 km off Gombe-Yola road immediately after Kumo town.

d) Enugu Coal: Enugu state is situated within longitudes $6^0 50^1$E and long.$7^0 52^1$E and latitudes 60001N and lat.$7^0 10^1$N. The entire areal extent is 7,161 sq. kilometers and the estimated population is 4,267,837 people (Census 2006). Presently Enugu State consists of 17 local government areas. It is bordered to the north by Benue state and to the west by Anambra state, to the east by Ebonyi state, and Abia State to the south.

Figure 1: Map of Nigeria showing Sample Locations

Locations of coal mines = Garin Maiganga, Gombe State, = Gindi Akwati, Plateau State

= Okaba, Kogi State = Enugu, Enugu State

Sample collection and preparation

Coal samples were collected by collecting 2-3 samples from each coal mines using a stopped belt sampler and were homogenized into a fine powder using a jaw crusher and a ball mills.

Each of the coal samples collected were dried and crushed to a fine powder with use of pulverizer. Packaging of the samples into radon-impermeable cylinder plastic containers which were selected based on the space allocation of the detector vessel which measures 7.6 cm by 7.6 cm in dimension (geometry) was also carried out. To prevent radon-222 escaping, the packaging in each case was triple sealed.

The sealing process included smearing of the inner rim of each container lid with vaseline jelly, filling the lid assembly gap with candle wax to block the gaps between lid and container, and tight-sealing lid-container with masking adhesive tape. Radon and its short-lived progenies were allowed to reach secular radioactive equilibrium by storing the samples for 30 days prior to gamma spectroscopy measurements.

Evaluation of Radioactivity of Samples

The analysis was carried out using 76x76mm NaI (TI) detector crystal optically coupled to a photomultiplier tube. The assembly has a preamplifier incorporated into it and a 1kilovolt external source. The detector is enclosed in a

6cm lead shield with cadmium and copper sheets. This arrangement is a mined at minimizing the effects of background and scattered radiation.

The data acquition software is Maestro by Camberra Nuclear Products. The samples were measured for a period of 29000 seconds for each samples. The peak area of each energy in the spectrum was used to compute the activity concentrations in each sample by using the following equation:

$$C(Bq.kg^{-1}) = \frac{Cn}{C_{fk}}$$

Where,
C = activity concentration of the radionuclides in the sample given in $BqKg^{-1}$
Cn = count rate (counts per second)
C_{fk}= calibration factor of the detecting system.
Count per second (cps) = Net Count/Live Time

Calibration and Efficiency Determinations

Calibration of the system for energy and efficiency were done with two calibration point sources, Cs-137 and Co-60. These were done with the amplifier gain that gives 72% energy resolution for the 66.16Kev of Cs-137 and counted for 30minutes.

Standards

The standards used to check for the calibration are the International Atomic Energy Agencies (IAEA) gamma Spectrometric reference materials RGK-1 for K-40, RGU-1 for Ra-226 (Bi-214 peak) and RTG-1 for Th-232 (Ti-208)

Background

The background count rate done for 29000 seconds.

Table 1: Spectral energy windows used in the Analysis

Isotope	Gamma Energy (Kev)	Energy Window (Kev)
Ra-226	1764.0	1620-1820
Th-232	2614.5	2480-2820
K-40	1460.0	1380-1550

Table 2: Energy Calibration for qualitative Spectral Analysis

Isotope	Calibration factors		Conversion Factors	Detection Limits	
	10^{-3} (cps/ppm)	10^{-4} (cps/ppm)	($BqKg^{-1}$)	ppm	Bq/kg
^{40}K	0.026	6.431	0.032	454.54	14.54
^{226}Ra	10.500	8.632	12.200	0.32	3.84
^{232}Th	3.612	8.768	4.120	2.27	9.08

Principle of Sodium Iodide Detector

NaI detectors is use for identifying specific radionuclides by measuring the energy of each individual gamma that enters the crystal ,this process is called gamma spectroscopy, or can also be called multi-channel analysis (and the instrument set up for this purpose is called a gamma spectroscopy device, or a multi-channel analyzer (MCA). The basic principle behind gamma spectroscopy is that every gamma-emitting radionuclide emits a gamma ray (or a few gammas) with very specific energies like a finger print and if we can identify the gamma energies precisely enough then we can identify the radionuclide(s) present. For example, cesium-137 (Cs-137) gives off a gamma with an energy of 662 thousand electron volts (keV). In an analyzed a gamma ray spectrum and a peak with energy of 662 keV indicates that Cs-137 is present. Along the same lines, seeing twin gamma peaks at about 1.1 and 1.3 million electron volts (MeV) tells us that we've found cobalt-60 (Co-60).

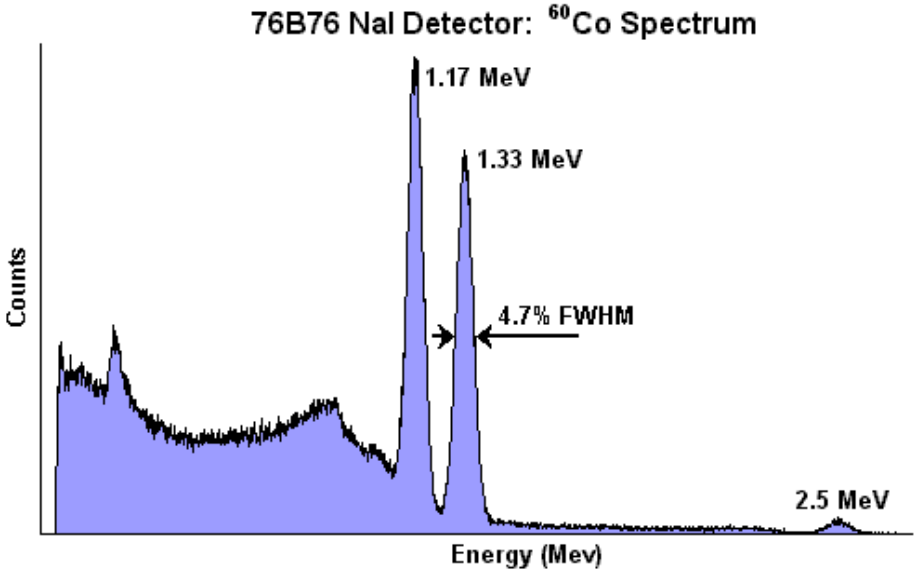

Figure 2: gamma ray spectrum and energy of Cobalt-6

The problem is in figuring out how much energy is in each gamma photon, luckily we can do this with a scintillation detector. When a gamma ray interacts with the NaI crystal it deposits energy, this energy is what causes the photons to be given off. Not only that, but a predictable number of photons are emitted depending on the energy deposited in a sodium iodide crystal, depositing 1 MeV of energy will cause about 42,000 scintillation photons to be emitted. We know the number of photons that it takes to eject a single electron from the photocathode, and we know the amount of amplification (and the size of the electrical pulse) for each electron ejected. So if we can measure the size of the output pulse then we know how much energy was deposited in the crystal and we can know what radionuclide emitted the gamma that we just detected.

Results and Discussion

Table 3: Results of the radioactivity in the coal samples (cps & Bq/kg)

S. No.	Sample ID Coal	K-40 (CPS)	Error ± (CPS)	K-40 (Bq/Kg)	Error ± (Bq/Kg)	Ra-226 (CPS)	Error ± (CPS)	Ra-226 (Bq/Kg)	Error ± (Bq/Kg)	Th-232 (CPS)	Error ± (CPS)	Th-232 (Bq/Kg)	Error ± (Bq/Kg)
1.	Okaba	0.04541	0.00359	70.62208	5.583204	0.0295	0.00336	34.18308	3.893395	0.01651	0.0023	18.82554	2.622577
2.	Enugu	0.0583	0.00449	90.66874	6.982893	0.0272	0.00296	31.51796	3.429896	0.0236	0.0014	26.90992	1.596351
3.	G/MaiGanGan	0.07544	0.00319	117.325	4.96112	0.0278	0.00066	32.21321	0.764774	0.0409	0.0013	46.63626	1.482326
4.	G/Akwati	0.0164	0.00119	25.50544	1.8507	0.0251	0.00156	29.08459	1.807648	0.0126	0.001	14.36716	1.140251

A first modification to be considered is to make sure that there is no loss of part of the radon gas that is present inside the coals, in the processes of sample preparation and analysis. However usually all radon isotopes are supposed to be emitted to the atmosphere by the stacks, considered in secular equilibrium with their parents but in determining the appropriate radioactivity of the samples, the radon are been carefully saved. Therefore, Radioactivity of the coal samples were determined using Sodium Iodide Detector (NaI), this is to show the possible radionuclides present in the coal samples. Table 3 shows the radioactivities from K-40, Ra-226 and Th-232 at various locations of the sample sites. The average values for K-40, Ra-226 and Th-232 in the coal samples are : K-40; Okaba coal 70.622Bq/kg ± 5.583Bq/kg, Enugu coal 90.669Bq/kg ± 6.983Bq/kg, Garim Maiganga coal 117.325Bq/kg ± 4.961 Bq/kg, Gindi Akwati 25.505 Bq/kg ± 1.851Bq/kg, Ra-226; Okaba coal 34.183Bq/kg ± 3.893 Bq/kg, Enugu coal 31.518Bq/kg ±3.430Bq/kg, Garin Maiganga coal 32.213Bq/kg ± 0.765 Bq/kg, Gindi Akwati

29.085 Bq/kg ± 1.808Bq/kg, Th-232; Okaba coal 18.826 Bq/kg ± 2.623 Bq/kg, Enugu coal 26.910Bq/kg ± 1.596Bq/kg, Garin Maiganga coal 46.636 Bq/kg ± 1.482 Bq/kg, Gindi Akwati coal 14.367 Bq/kg ± 1.140Bq/kg.
The measurements indicate a very low K-40, Ra-226 and Th-232 radioactivities in the samples. K-40 has the highest radioactivity except for Gindi Akwati that is low and Th-232 has the lowest radioactivity while Garin Maiganga valueis high. Furthermore, High concentration of radon in coal sample from turkey was reported by Abdallah *et al.*, (2009) [6]. Tsikritzis *et al.*, (2008) [7] reported The mean concentrations of $^{(238)}$U and $^{(226)}$Ra in lignite were found to be higher than that in steriles from coal samples obtained from the West Macedonia Lignite Center. Cile *et al.*, (2010) [8] reported that the results obtained from Radon concentrations in three underground lignite mines in Turkey are far under the action levels. The annual effect limit obtained from this research is below the world average natural dose which is 370Bq/kg per year.

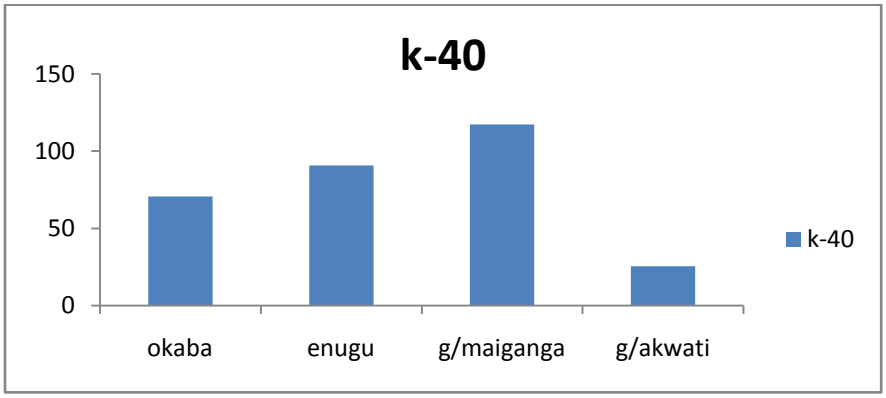

Figure 3: Chart showing K-40 in all the sample locations

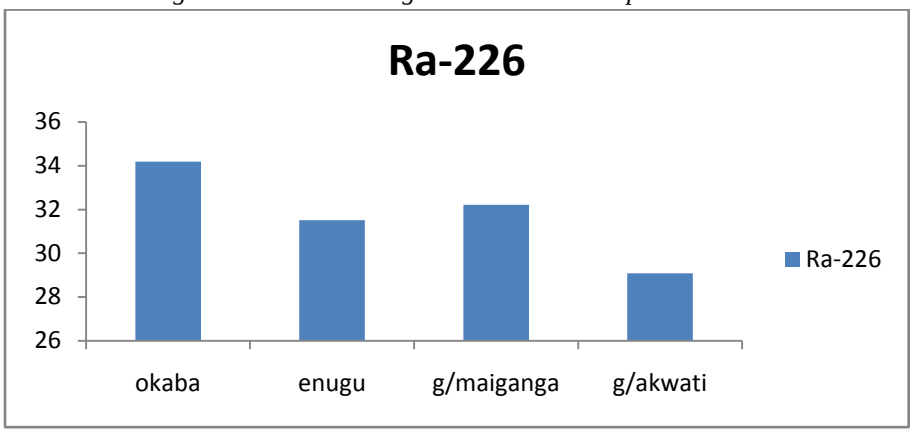

Figure 4: Chart showing Ra-226 in all the sample locations

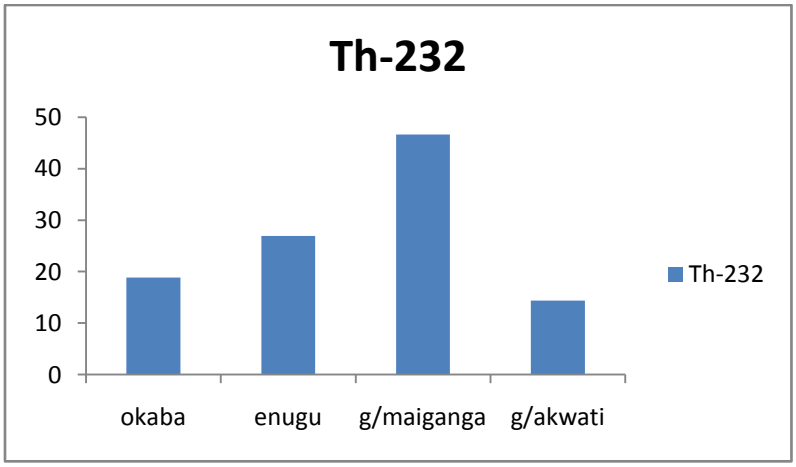

Figure 5: Chart showing Th-232 in all the sample locations

Conclusion

The radiological assessment of coal sample from the various study locations have been carried out and the results obtained have proved that the radioactivities arising from them are below the permissible level.

The populace can continue to explore the usage of coal for domestic and industrial purposes in Nigeria. The federal government of Nigeria can as a matter of urgency begin to diverse to non-renewable energy to save the country from the eminent collapse of the oil industry.

The coal samples from various locations are deemed fit for human domestic use as the radioactivities arising from them are below world average natural dose. This observation provides a useful geologic perspective for addressing societal concerns regarding possible radiation and radon hazard.

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