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#### RADIOMETRIC ASSESSMENT OF THE RADIOLOGICAL HEALTH IMPLICATIONS ASSOCIATED WITH THE EXPLOITATION OF MAJOR NIGERIAN FOSSIL FUELS

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# ABSTRACT

Gamma spectroscopic technique was used to determine qualitatively and quantitatively the natural radionuclides present in the prominent fossil fuels found in Nigeria (bitumen, coal and crude oil). The radiological health indexes emanating from these radionuclides were then estimated with a view to ascertaining the radiological health consequences associated with the exploitation of these fuels. The results show that the total radioactivity contents are 61.5  $\pm$  8.2  $Bq kg^{-1}$  in bitumen, 32.5  $\pm$  1.5  $Bq kg^{-1}$  in coal and 0.04  $\pm$  0.01  $Bq kg^{-1}$  in crude oil. The mean radium equivalent values are (30.81, 28.22 and 0.06) Bqkg<sup>-1</sup> respectively while the respective external and internal hazard indexes are (0.083, 0.076 and 1.51 x 10<sup>-5</sup>) Bqkg<sup>-1</sup> and (0.159, 0.123 and 3.03 x 10<sup>-5</sup>) Bqkg<sup>-1</sup>. The mean annual dose equivalent values are (0.017, 0.016 and 3.06 x 10<sup>-5</sup>) mSvyr<sup>-1</sup> respectively. These values are below those stipulated in radiation protection. The radiological health consequences associated with the exploitation of the three major Nigerian fossil fuels therefore is insignificant.

Keywords: Radiometric assessment, Exploitation, Nigeria fossil fuels, insignificant radiological health consequence

# INTRODUCTION

Presently the major fossil fuels in Nigeria are coal, crude oil and bitumen of which the most exploited is crude. As geological materials, these are known to be largely associated with naturally Occurring Radioactive Materials (NORM). Thus they contain <sup>238</sup>U /<sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K (Balogun et al, 2003). This radioactive contamination of geological materials has become of concern in radiation protection as these NORM are known to have reached hazardous levels (Eldine et al, 2001). In fact they have been found to be capable of resulting in much larger radiological exposure to the public relative to that caused by the nuclear industry for instance (Mokobia et al, 2006). Thus there are health implications of this contamination both on the exploiters and those living in the immediate environments where exploitation is being carried out.

One aspect of this has to do with <sup>222</sup>Rn (radon) a member of the <sup>238</sup>U/<sup>226</sup>Ra (Uranium) decay series (Mokobia, 2004a: 2010). This short-lived radioactive gas (0.83days) decays into much shorter-lived <sup>218</sup>Po (Polonium) and <sup>214</sup>Pb (Lead) emitting  $\alpha$  particle which causes cancer of the lungs if breathed in (Mokobia, 2004b). Consequent upon these, this study sought to investigate the radiological implications associated with the exploitation of these major Nigerian fossil fuels. Specifically, the qualitative and quantitative determinations of their natural radionuclide contents were carried out using gamma spectrometric technique. The quantitative data obtained were then used to estimate the associated radiological hazard indexes. The values obtained were then compared with radiological health international stipulations and the associated radiological health implications ascertained.

# MATERIALS AND METHOD

Coal samples of the Permian age were obtained from various locations around Enugu (latitude  $6^{\circ} 26' N$  and longitude  $7^{\circ} 30' E$ ) the major location of Nigerian bituminous coal (Mokobia et al, 2006; Mokobia and Balogun, 2004). Surface samples of bituminous sands of the early cretaceous age were collected from locations lying within the East-West belt stretching across Lagos, Ogun, Ondo and Edo States in South Western Nigeria stretching from latitude ( $6^{\circ} 24' to 6^{\circ} 25'$ ) N and longitude ( $3^{\circ} 48' to 6^{\circ} 48'$ )  $E_{\perp}$  The crude oil samples largely

of the cretaceous age were obtained from Mobil Nigeria offshore locations at Eket (latitude

 $5^{\circ} 40' N$  and longitude  $8^{\circ} 01' E$  Known masses of these fuels were separately weighed into Marinelli beakers. These beakers were then securely sealed and left for 28 days in accordance with conventional practice (Balogun et al, 2003; Mokobia et al, 2006; Mokobia, 2005). Natural radioactivity measurement was performed using a Canberra vertical highpurity germanium coaxial detector (HPGe) based gamma spectrometer. This semiconductor device having a volume of 155 mm<sup>3</sup> was enclosed in a 100 mm thick lead shield to minimize the interfering effects of the radiation from sources other than the samples and natural radiation from the environment where the counting was carried out. Energy as well as efficiency calibrations of the device was carried out using a mixed source soil standard obtained from the International Atomic Energy Agency (IAEA). Each of the samples as well as the standard and the background were counted for 10 hrs. It was ensured that the sample containers and that of the standard have the same configuration so as to minimize error. Spectra evaluation was carried out using a PC based SAMPO 90 computer program capable of matching the y- energies at various levels to a library of possible isotopes. The specific activities of the radionuclides detected in each of the studied samples were calculated using the comparative approach:

$$A_s = \frac{N(E_{\gamma})_s M_d A_d}{N(E_{\gamma})_d M_s}$$
[1]

 $A_s$  is the specific activity in Bqkg<sup>-1</sup> of the identified nuclide having a net photopeak area  $N(E_{\gamma})_s$  contained in a sample of mass  $M_s$  in kg.  $A_d$  is the activity of this nuclide contained in the mixed standard of mass  $M_d$  in kg.  $N(E_{\gamma})_d$  is the net photopeak area of the said nuclide. This approach was adopted because it is acclaimed to be relatively more accurate compared with the absolute method (Gilmore and Hemingway, 2002).

Radium – equivalent activity was calculated using the relationship (Tufail et al 2000; Kharter t al, 2001; Frame, 2009):

$$A_{Ra_{ea}} = A_{Ra} + 1.43A_{Th} + 0.077A_{K}$$
<sup>[2]</sup>

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The external and internal health hazard indexes arising from the use of this mineral was determined by employing the equations used in (Mokobia, 2010; Tufail et al, 2000):

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810}$$
[3]

$$H_{\rm int} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_{K}}{4810}$$
[4]

 $A_{Ra}$ ,  $A_{Th}$  and  $A_K$  represent the activity concentrations of  ${}^{238}U/{}^{226}Ra$ ,  ${}^{232}Th$  and  ${}^{40}K$  in Bqkg<sup>-1</sup> respectively. The annual effective dose rate (mSvy<sup>-1</sup>) was determined using the relationship:  $E = TOD * 10^{-6} * F.$  [5]

 $D_t$ , the outdoor dose rate in air (nGyh<sup>-1</sup>) was obtained from the relation (Farai and Jibiri, 2000):  $D_t = 0.446A_{Ra} + 0.662A_{Th} + 0.048A_K$  [6]

 $10^{\text{-6}}$  represents a conversion factor, T is 8760 hours per year, Q = 0.7  $\text{SvGy}^{\text{-1}}$  and  $F_{t}$  = 0.2 is the outdoor occupancy factor.

### **RESULTS AND DISCUSSION**

In Table 1, the specific activities of the primordial radionuclides detected in these fuels, their determined radium equivalent activities and the consequent external and internal hazard indexes as obtained in this work for each of them are presented. The radionuclides detected belong to the <sup>226</sup>Ra/<sup>238</sup>U; <sup>228</sup>Ra/<sup>232</sup>Th decay series with specific activities ranging from 0.04  $\pm$  0.01 Bqkg<sup>-1</sup> in crude to 28.2  $\pm$  3.7 Bqkg<sup>-1</sup> in bitumen and 7.5  $\pm$  0.4 Bqkg<sup>-1</sup> in coal to 33.3  $\pm$  4.5 Bqkg<sup>-1</sup> in bitumen respectively. The non decay series <sup>40</sup>K (mean specific activity 7.8  $\pm$ 0.2 Bgkg<sup>-1</sup>) was only detected in the coal samples. This shows that <sup>40</sup>K is not the major contributor to the radioactivity in these fossils unlike the results obtained for some other geological materials (Turhan et al, 2008; Ngachin et al 2008). The values of the mean specific activities of the primordial radionuclides detected in these studied samples are lower than the values obtained for some other geological materials such as marble (Tufail et al, 2000; Mokobia, 2008; Turhan, 2009), kaolin (Zoltan et al, 2005) and liquid paints (Mokobia et al, 2003). The total radioactivity contents of the fossils are  $(61.5 \pm 8.2, 32.5 \pm 1.5 \text{ and}$  $0.04 \pm 0.01$ ) Bgkg<sup>-1</sup> for bitumen, coal and crude respectively. Radioactivity content is least in the crude samples. This low radioactivity observed for the crude falls within the range 0.02 to  $0.2 \text{ Bg}(^{226}\text{Ra})\text{kg}^{-1}$  given in the literature (SPIM, 1993).

The calculated mean values for the external hazard indexes are (0.08, 0.08 and 1.51 x10<sup>-4</sup>) Bqkg<sup>-1</sup> for coal, bitumen and crude oil respectively. These values are each less than unity. This suggests that the exploitation of these fossils does not constitute any radiological effect of threat to the public around the locations of the exploitation. Indoor <sup>226</sup>Ra accumulation has been established to cause exposure to persons through the inhalation of its progeny <sup>222</sup>Rn (Mokobia, 2004b; Hizem et al, 2005). The estimated internal hazard indexes following the indoor exposure of members of the public around the immediate locations where exploitation

is carried out are (0.12, 0.16 and 3.03 x  $10^{-4}$ ) Bqkg<sup>-1</sup> in that order. These values are each less than unity. This indicates that indoor radiological health implication is insignificant.

The mean annual effective dose equivalent determined for coal and bitumen are each 0.02 mSvy<sup>-1</sup>. This value is less than the recommended value of 1mSvy<sup>-1</sup> (2%) for non radiation workers (members of the public) by the International Commission on Radiological Protection (ICRP, 1990). The value obtained for the crude samples is 0.0003 mSvy<sup>-1</sup> (.03%). The fact that these values of dose equivalent are less than the recommended unity also suggests that radiation dose to members of the public resulting from the exploitation of these Nigeria fossils has negligible radiological consequence. As shown in Figure 1, of these three fossil fuels studied, the exploitation of crude oil is associated with the least radiological hazard relative to coal and bitumen.

# CONCLUSION

From the results, it is concluded that the radiological health consequences associated with the exploitation of the three major Nigerian fossil fuels from the standpoint of radiation protection is insignificant.

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 Table 1: Activities of Nuclides in Nigerian Fossils and the Corresponding Determined Hazard

 Indexes

Sample	<sup>40</sup> K	<sup>238</sup> U/ <sup>226</sup> Ra	<sup>232</sup> Th/ <sup>228</sup> Ra	External	Internal hazard	Annual
	(Bq kg <sup>-1</sup> )	(Bq kg⁻¹)	(Bq kg⁻¹)	hazard	index	effective
				index	(Bqkg⁻¹)	dose
				(Bqkg <sup>-1</sup> )		(mSvy <sup>-1</sup> )
Coal						
FC1	7.1 ± 0.02	13.2 ± 0.4	7.1 ± 0.1	0.065	0.100	0.013
FC2	8.7 ± 0.02	19.0 ± 1.6	6.4 ± 0.4	0.078	0.129	0.016
FC3	7.6 ± 0.03	$16.1 \pm 0.6$	6.0 ± 0.5	0.068	0.112	0.014
Fc4	8.0 ± 0.01	17.5 ± 0.4	9.5 ± 0.3	0.086	0.133	0.018
FC5	7.5 ± 0.01	20.1 ± 1.8	7.5 ± 0.5	0.085	0.139	0.018
Mean	7.8 ± 0.2	17.2 ± 0.9	7.5 ± 0.4	0.076	0.123	0.016
Crude Oil						
FCO1	ND	$0.05 \pm 0.004$	ND	1.35 x 10 <sup>-4</sup>	2.70 x 10 <sup>-4</sup>	2.73 x 10 <sup>-4</sup>
FCO2	ND	$0.07 \pm 0.008$	ND	1.89 x 10 <sup>-4</sup>	3.78 x 10 <sup>-4</sup>	3.83 x 10 <sup>-4</sup>
FCO3	ND	$0.04 \pm 0.003$	ND	1.08 x 10 <sup>-4</sup>	2.16 x 10 <sup>-4</sup>	2.19 x 10 <sup>-4</sup>
FCO4	ND	$0.07 \pm 0.004$	ND	1.89 x 10 <sup>-4</sup>	3.78 x 10 <sup>-4</sup>	3.83 x 10 <sup>-4</sup>
FCO5	ND	$0.05 \pm 0.006$	ND	1.35 x 10 <sup>-4</sup>	2.70 x 10 <sup>-4</sup>	2.73 x 10 <sup>-4</sup>
Mean	ND	$0.04 \pm 0.005$	ND	1.51 x 10 <sup>-4</sup>	3.03 x 10 <sup>-4</sup>	3.06 x 10 <sup>-4</sup>
Bitumen						
FB1	ND	29.1 ± 4.6	35.9 ± 5.4	0.086	0.165	0.018
FB2	ND	28.2 ± 3.3	36.9 ± 5.7	0.084	0.160	0.018
FB3	ND	26.7 ± 3.2	30.7 ± 3.5	0.079	0.151	0.016
FB4	ND	28.1 ± 4.0	32.6 ± 3.5	0.083	0.159	0.017
FB5	ND	29.1 ± 3.6	30.6 ± 4.6	0.085	0.154	0.018
Mean	ND	28.2 ± 3.7	33.3 ± 4.5	0.083	0.159	0.017



#### a) Coal, Bitumen and Crude oil



# b) Crude oil

Fig. 1: Graph of the Relative Hazard Indexes of Three Nigerian Prominent Fossil Fuels

### REFERENCES

- Balogun F A, Mokobia C E. Fasasi M K, Ogundare F O (2003). "Natural radioactivity associated with bituminous coal mining in Nigeria". Nuclear Instruments and Methods in Physics Research A . 505, 444-448.
- El Dine N W, EL-Shershaby A. Ahmed F. Abdel-Haleem A (2001). Measurement of radioactivity and radon exhalation rate in different kinds of marble and granites. Applied Radiation and Isotopes, 55 (6) 853-860.
- Farai I P, Jibiri N N (2000). Baseline Studies of Terrestrial Outdoor Gamma Dose Rate Levels in Nigeria. Radiat. Prot. Dosim., 88 (3) 247 – 254.
- Frame P (2009). Calculation of radium equivalent activity. <u>http://wwwhps.org/publicinformation/ate/q543.html, Assessed 2/11/10</u>.
- Gilmore G, Hemingway (2002) J. Practical Gamma–Ray Spectrometry. John Wiley and Sons, New York, pp 1 – 177.
- Hizem N, Ben Fredj A, Ghedira L. "Determination of Natural Radioactivity in Building Materials used in Tunisian Dwellings by Gamma Ray Spectrometry". Radiation Protection Dosimetry, 2005. 114 (4), 533-537
- ICRP. Radiation Protection. Publication 60. 1990 Recommendations of the International Commission on Radiological Protection, Ann. ICRP, 1991. 21 (1 – 3) Khater A E M, Higgy R H, Pimpl M (2001). Radiological impacts of natural radioactivity in Abu – Tartor phosphate deposits, Egypt. Journal of Environmental Radioactivity,. 55,255 – 267.
- Mokobia C E (2004a). Natural Radioactivity in Nigerian Functional Coal Mines M. Phil thesis, Obafemi Awolowo University Ile- Ife,.
- Mokobia C E (2004b). The effect of moisture on <sup>222</sup>Rn Emanation. Journal of Science and Technology Research, 3 (1), 71-73.
- Mokobia C E (2008). A Study of Gamma Induced Thermally Stimulated Luminescence (TSL) of Natural Dolerite and Marble. Ph.D Thesis, Obafemi Awolowo University, Ile-Ife.
- Mokobia C E (2010). Radioanalysis of Natural Kaolin Using Nuclear Spectroscopy. Proceedings of Delta State University Faculty of Science Conference In Press.
- Mokobia C E and Balogun F A (2004). Background Gamma Terrestrial Dose Rate in Nigerian Functional Coal Mines. Radiation Protection Dosimetry, 108.2, 169-173.

- Mokobia C E, Tchokossa P, Olomo J B, Balogun F A (2003) "Assessment of the Natural Radioactivity Content of some Liquid Paints Available in Nigeria". Nig. Journ. of Phys, 15 (1), 14-16.
- Mokobia C E, Adebiyi F M, Akpan I, Olise F S, Tchokossa P (2006). Radioassay of prominent Nigerian fossil fuels using gamma and TXRF spectroscopy. Fuel , 85, 1811-1814.
- Ngachin M, Garavaglia M, Glovani C, Nourreddine A, Kwato Njock M G, Scruzzi E, Lagos L (2008). <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K contents and radon exhalation rate from materials used for construction and decoration in Cameroon. Journal of Radiological Protection, 2008. 28 (369).
- SPIM (1993). Shell Internationale Petroleum Maatschappij, Ionizing radiation safety guide, Shell

Safety and Health Committee, p. 41 (B.V.)

- Tufail M, Iqbal M, Mirza S M. "Radiation doses due to the natural radioactivity in Pakistan marble". Radioprotection, 2000. 35, 299 310
- Turhan S, Baykan U N, Sen K (2008). Measurement of natural radioactivity in building materials used in Ankara and assessment of external doses. Journal of Radiological Protection, 2008. 28 (83)
- Turhan S (2009). Radiological impacts of the usability of clay and kaolin as raw materials in manufacturing of structural building materials in Turkey. Journal of Radiological Protection, 29 (75).
- Zoltan A, Richard B W. Bentonite, kaolin and selected clay minerals. International Labour Organisation, United Nations Environmental Programme, World Health Organisation, 2005,5- 6.