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Assessment of Natural Radionuclides in Fly Ash Produced at Orji River Thermal Power Station, Nigeria and the Associated Radiological Impact

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Abstract

Coal fired power plants produce significant amounts of ashes, which are quite often being used as additives in cement and other building materials. Coal contains trace quantities of ^{226}Ra, ^{232}Th and ^{40}K. The concentrations of these radionuclides are usually low in the coal, but enriched in fly ash. The activity concentrations of natural radionuclides in fly ash and soil samples in the vicinity of Orji River thermal power station in Nigeria was determined by gamma ray spectroscopy method using NaI(Tl) detector. The mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the fly ash were 40.8 ± 11.6, 49.1 ± 9.3 and 321 ± 17 Bq·kg^{-1}, respectively. Coarse fly ash collected from the dump site had mean activity concentrations of 28.2 ± 8.3, 37.6 ± 5.0 and 335 ± 32 Bq·kg^{-1}, respectively for ^{226}Ra, ^{232}Th and ^{40}K. Soil samples collected at about distances of 10 m from the dump site had 32.7 ± 4.3, 40.0 ± 4.2 and 298 ± 15 Bq·kg^{-1}, respectively. Those collected at about distances of 100 m from the dump site had 39.1 ± 11.2, 34.1 ± 5.2 and 257 ± 19 Bq·kg^{-1}, respectively. Occupational dose received by workers due to exposure to the fly ash ranged between 33.0 and 61.2 µSv·y^{-1} with a mean value of 47.1 ± 8.4 µSv·y^{-1}, which is below the intervention exemption level of 1 mSv·y^{-1}. The radium equivalent activity concentration, external and internal hazard indices of the fly ash were below the recommended maximum values for building materials. The mean outdoor absorbed dose rate and the annual effective dose obtained in the vicinity of the plant were 49.7 ± 4.0 nGy·y^{-1} and 0.30 ± 0.02 mSv (dump site), 51.7 ± 3.6 nGy·y^{-1} and 0.32 mSv (soil 10 m from dump site), 49.4 ± 4.9 nGy·y^{-1} and 0.30 ± 0.03 mSv (soil 100 m away from dump site), which are lower than the world average. The results obtained in this study show that there is no significant radiological impact of the fly ash on both the workers and the public from radiation protection point of view.

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Keywords

Fly Ash, Natural Radionuclides, Gamma Radiation, Occupational Dose, Absorbed Dose Rate

1. Introduction

Measurements of radioactivity in Technologically Enhanced Naturally Occurring Radioactive Materials (TENORMs) are important from radiation protection point of view because more than 50% of the total dose to human population from natural sources of radiation is contributed by these materials [1]. Coal is technologically important material used for power generation and its fly ash is used in manufacturing of bricks, sheet, cement, land filling etc. Coal, like most materials found in nature, contains trace quantities of the naturally occurring primordial radionuclides arising from the U and Th series and ^{40}K. Naturally occurring radionuclides contribute most to environmental radiation. The concentration of these long lived radionuclides is usually low in the coal. When it is burnt in power plant, the fly ash that is emitted through the stack gets enriched in some of the radionuclide [2]. Therefore, the combustion of coal results in the release of some natural radioactive elements into the environment and in the redistribution of these radioactive elements in the surface soil particularly in the vicinity of coal-fired power plants. This can modify ambient radiation fields and population exposures [3]. Thus coal fired power plant is a major contributor towards Technologically Enhanced Natural Radiation (TENR) [4]. The interest in measuring radioactivity in terms of activity concentrations of ^{238}U, ^{232}Th and ^{40}K concentration in fly ash is due to its health hazards and environmental pollution. The concentration of radioactivity in fly ash is extremely variable and depends on the composition of the parent coal, conditions during coal combustion and efficiency of emission control devices etc. [2].

The Oji River Power Station is a thermal power station in Oji River town in Enugu State, Nigeria. The plant reportedly last generated power in 2004. The Oji River thermal power station was effective with power generation to commercial towns and improved the industrial base of the eastern region since it was established by the British colonial administration in the country. The efficacy of its operation is based on the fact that there are constructed mechanical devices that enhance intake of water from the fast flowing Oji River into the plant house that is powered by natural coal to provide electrical energy, which is then transmitted to various towns in the former eastern region. At present, the thermal power station is not functional. However, there is fly ash in the burning chambers and dump of fly ash in the vicinity of the station. This could pose radiation risk to the workers and people in the vicinity of the thermal station. This study seeks to determine the activity concentration of natural radionuclides in fly ash and soil samples collected in the vicinity of the power station and assess the radiological implication.

2. Material and Method

2.1. Gamma Activity

2.1.1. Sample Preparation

A total of 20 samples of fly ash were collected from the thermal plant. In order to investigate the radioenvironmental impact due to discarded fly ash from the power plant, samples were collected from dump site where fly ash is discarded and soil samples in the vicinity of the power plant. These composed of 10 coarse fly ash samples from the dump site, 10 soil samples at distances of about 10 m away from the dump site and 10 soil samples at distances of about 100 m away from the dump site. The soil samples were collected from the surface layer (0 - 10 cm depth). The samples were oven dried at a temperature of 100˚C until a constant weight was attained. The coarse fly ash from the dump site and the soil samples were pulverized. A mass of 130 g each of the fly ash and coarse fly ash and 150 g each of the soil samples were packed in cylindrical plastic containers of uniform size of base diameter 7 cm which could sit on the 7.6 cm by 7.6 cm NaI(T1) detector. The plastic containers were sealed tightly with caps and wrapped with thick cellotape around their screw necks and kept for more than four weeks in order for ^{226}Ra, ^{232}Th and their daughter products to attain secular equilibrium.

2.1.2. Activity Concentration Measurement

The activity concentration of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K were measured with gamma ray spectrometry system having a 7.6 cm by 7.6 cm NaI(T1) detector (Model No. 802 series, Canberra Inc.) shielded in a lead. The detector is coupled through a preamplifier base to Canberra series 10 plus multichannel analyzer (MCA) (Model No.1104) and has a resolution of about 8% at energy of 0.662 MeV (^{137}Cs). This is capable of distinguishing the gamma ray energies considered during the measurements. The uranium-238 and thorium-232 activities were determined indirectly through the activities of their daughter products. The choice of the reference nuclides for their activity determination was made based on the fact that NaI(Tl) detector has a poor resolution, hence, the peaks of interest to be considered would be sufficiently discriminated and intense. The ^{226}Ra and ^{232}Th contents were determined from 1.760 MeV (^{214}Bi) and 2.614 MeV (^{208}Tl), respectively. The potassium content was determined from 1.461 MeV photopeak of ^{40}K.

The energy calibration of the counting system was done as described by Ademola and Olatunji (2013) [5]. The detection efficiency of the detector was determined using a reference source of known activity concentrations of each of the radionuclides prepared from Rocketdyne Laboratories, California USA, which is traceable to a mixed standard gamma source (No. 48772-356) by Analytic Inc., Atlanta, Georgia. Each of the sample containers was placed on top of the detector and counted for 10 hours. Background radiation was corrected for all the samples.

2.2. Occupational Dose Assessment

Occupational exposure could be external, internal or both. For the assessment of the occupational dose at the power plant station due to the fly ash, the following exposure pathways were considered:

1. external exposure to gamma radiation
2. inhalation of dust

The external exposure to gamma radiation is calculated using the equation [6]

$$E_{ext} = \sum_R A_R DC_{ext,R} T \quad (1)$$

where A_R is the activity concentration of radionuclide R (Bq·kg^{-1}), $DC_{ext,R}$ is the effective dose coefficient for radionuclide R in Sv·h^{-1} per Bq·kg^{-1} and T is the annual exposure time assumed to be 2000 h·y^{-1}.

The inhalation of contaminated dust was calculated using the relation given in Weiss *et al.* (2005) [7];

$$E_{int} = TV \sum_R g_{inh,R} C_R \quad (2)$$

where V is the breathing rate, $g_{inh,R}$ is the inhalation dose coefficient for radionuclide R (Sv·Bq^{-1}) and C_R is the ambient activity concentration of radionuclide R (Bq·m^{-3}).

$$C_R = A_R \cdot k \quad (3)$$

where k is the dust loading factor in kg·m^{-3}.

2.3. Radiological Hazard Assessment of Fly Ash as Building Material

Fly ash has been shown to be a very good substitute of soil for geo-technical applications, substitute of cement for mortar and concrete and substitute of clay for manufacture of bricks and various ceramic products [8]. The radiological hazard of materials used as building materials is assessed by the radium equivalent activity concentration (Ra_{eq}), external (H_{ext}) and internal (H_{int}) hazard indices. Ra_{eq}, H_{ext} and H_{int} are calculated using the following relations, respectively [9]-[11];

$$Ra_{eq} = A_{\mathrm{Ra}} + 1.43A_{\mathrm{Th}} + 0.077A_{\mathrm{K}} \quad (4)$$

$$H_{ext} = A_{\mathrm{Ra}}/370 + A_{\mathrm{Th}}/259 + A_{\mathrm{K}}/4810 \le 1 \quad (5)$$

$$H_{int} = A_{\mathrm{Ra}}/185 + A_{\mathrm{Th}}/259 + A_{\mathrm{K}}/4810 \le 1 \quad (6)$$

where A_{Ra}, A_{Th} and A_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq·kg^{-1}, respectively. Radium equivalent activity concentration is based on the estimation that 1 Bq·kg^{-1} ^{226}Ra, 0.7 Bq·kg^{-1} ^{232}Th or 13 Bq·kg^{-1} ^{40}K produce the same gamma dose rate [12]. For safe use of building materials as far as radiation is concerned, the maximum value of Ra_{eq} in the materials must be less than 370 Bq·kg^{-1}. H_{ext} is obtained from the Ra_{eq} expression by assuming that its maximum value allowed (equal to unity) corresponds to the upper limit of Ra_{eq}

(370 Bq·kg^{-1}). H_{int} takes into consideration the radiation hazard to respiratory organs. The numerical quantities at the denominator of Equations (5) and (6) are in the units of Bq·kg^{-1}; hence the indices are dimensionless quantities. The indices should be less than unity for safe use of building material.

2.4. Outdoor Radiation Dose

The outdoor exposure due to gamma radiation from soil in the vicinity of the plant was calculated using the relation [13] [14];

$$D\left(\text{nGy}\cdot\text{h}^{-1}\right)=0.462A_{\text{Ra}}+0.604A_{\text{Th}}+0.0417A_{K} \quad (7)$$

where A_{Ra}, A_{Th} and A_{K} are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively in Bq·kg^{-1}. This equation accounts for external absorbed dose rate at 1 m above ground surface. The annual effective dose was estimated taking into account the conversion coefficient (CC), from absorbed dose to effective dose, 0.7 Sv·Gy^{-1} and occupancy factor (OF) [13] [15];

$$E\left(\text{mSv}\cdot\text{y}^{-1}\right)=D\times CC\times OF\times 8670\ \text{h}\cdot\text{y}^{-1}\times 10^{-6} \quad (8)$$

3. Results and Discussion

3.1. Activity Concentrations

The activity concentrations of the samples examined are presented in **Table 1** and **Table 2**. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the fly ash varied from 21.5 ± 2.8 to 65.7 ± 6.4, 33.6 ± 5.8 to 63.7 ± 10.4 and

Table 1. Activity concentrations of natural radionuclides in fly ash.

S/N	^{226}Ra (Bq·kg^{-1})	^{232}Th (Bq·kg^{-1})	^{40}K (Bq·kg^{-1})
1	59.3 ± 5.9	39.3 ± 6.6	331 ± 18
2	38.9 ± 4.2	41.7 ± 7.0	293 ± 16
3	52.4 ± 5.3	42.1 ± 7.1	299 ± 16
4	47.3 ± 4.9	41.0 ± 6.1	329 ± 18
5	40.6 ± 4.3	44.8 ± 7.5	321 ± 17
6	46.5 ± 4.8	61.0 ± 10.0	340 ± 18
7	21.5 ± 2.8	57.1 ± 9.4	316 ± 17
8	35.7 ± 3.9	33.6 ± 5.8	300 ± 16
9	42.5 ± 4.5	43.0 ± 7.2	313 ± 17
10	39.2 ± 4.2	40.6 ± 6.8	294 ± 16
11	47.6 ± 4.9	58.0 ± 9.5	320 ± 17
12	26.7 ± 3.2	48.7 ± 8.1	332 ± 18
13	44.4 ± 4.6	49.1 ± 8.1	318 ± 17
14	23.8 ± 3.0	42.3 ± 7.1	350 ± 19
15	27.5 ± 3.2	41.5 ± 7.0	309 ± 17
16	49.4 ± 5.0	54.5 ± 5.0	310 ± 17
17	41.6 ± 4.4	56.3 ± 9.2	356 ± 19
18	65.7 ± 6.4	62.9 ± 10.3	321 ± 17
19	28.5 ± 3.3	63.7 ± 10.4	330 ± 18
20	37.5 ± 4.0	61.1 ± 10.0	328 ± 18
Mean ± Std Dev	40.8 ± 11.6	49.1 ± 9.3	321 ± 17

Table 2. Activity concentrations of natural radionuclides in coarse fly ash from dump site and soil samples in the vicinity of the plant.

Sample	No.	^{226}Ra (Bq·kg^{-1})		^{232}Th (Bq·kg^{-1})		^{40}K (Bq·kg^{-1})	
		Mean ± Std. Dev	Range	Mean ± Std. Dev	Range	Mean ± Std. Dev	Range
Coarse fly ash	10	28.2 ± 8.3	18.1 - 38.8	37.6 ± 5.0	31.6 - 44.7	335 ± 32	287 - 385
Soil (10 m)	10	32.7 ± 4.3	26.3 - 38.4	40.0 ± 4.2	32.1 - 46.6	298 ± 15	278 - 324
Soil (100 m)	10	39.1 ± 11.2	14.6 - 52.4	34.1 ± 5.2	25.2 - 40.2	257 ± 19	223 - 286

293 ± 16 to 356 ± 19 Bq·kg^{-1}, with mean values of 40.8 ± 11.6, 49.1 ± 9.3 and 321 ± 17, respectively. Coarse fly ash from the dump site ranged between 18.1 ± 2.7 and 38.8 ± 4.4, 31.6 ± 5.6 and 44.7 ± 7.6, and 287 ± 16 and 385 ± 21 Bq·kg^{-1}. The mean values were 28.2 ± 8.3, 37.6 ± 5.0 and 335 ± 32 Bq·kg^{-1}, respectively. Soil samples, about distances of 10 m from the dump site ranged between 26.3 ± 3.3 and 38.4 ± 4.3, 32.1 ± 5.7 and 46.6 ± 7.8 and 278 ± 16 and 324 ± 18 Bq·kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The mean values were of 32.7 ± 4.3, 40.0 ± 4.2 and 298 ± 15 Bq·kg^{-1}, respectively, for ^{226}Ra, ^{232}Th and ^{40}K. Soil samples collected at about distances of 100 m from the dump site had activity concentrations in the range of 14.6 ± 2.5 and 52.4 ± 5.4, 25.2 ± 4.7 and 40.2 ± 6.9, and 223 ± 13 and 286 ± 16 Bq·kg^{-1} with mean values of 39.1 ± 11.2, 34.1 ± 5.2 and 257 ± 19 Bq·kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively. When compared with those of the world population weighted value for soil, ^{226}Ra in the fly ash and soil samples collected 100 m from the dump site are higher than the world population weighted value of 33 Bq·kg^{-1}. This could be due to the fact that uranium compounds are soluble and the enhanced level in the fly ash could be partially leached from the dump site to the soil [16]. The other two radionuclides in all the samples are within the world population weighted value of 45 Bq·kg^{-1} for ^{232}Th and 420 Bq·kg^{-1} for ^{40}K [13]. They are also lower than the global average for building material, 50 Bq·kg^{-1} for ^{226}Ra, 50 Bq·kg^{-1} for ^{232}Th and 500 Bq·kg^{-1} for ^{40}K [15].

3.2. Occupational Dose Assessment

The total dose to workers is the summation of the external dose and the internal dose that is Equations (1) and (2). The following parameters were used for the calculation of the occupational dose:

1. The effective dose equivalents for contaminated surface, 9.929, 0.003 and 1.175 nSv·h^{-1} per Bq·g^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively [6].
2. Inhalation dose coefficients, 2.2 × 10^{-6}, 2.9 × 10^{-5} and 3.0 × 10^{-9} Sv·Bq^{-1}, respectively for ^{226}Ra, ^{232}Th and ^{40}K. These are the values for the activity median aerodynamic diameter (AMAD) of dust particle 5 μm for normal working condition. The conversion factor corresponds to M (moderate residence time in lungs) for ^{226}Ra and ^{232}Th and F (short residence time in lungs) for ^{40}K [17].
3. The breathing rate for industrial scenario was taken as 1.5 m^3·h^{-1} [18].
4. Dust loading factor for industrial activity, 10 mg·m^{-3} was used [19].

The mean activity concentrations of the three radionuclides in the fly ash had been used to estimate the occupational exposure using Equations (1) and (2), and the parameters stated above. The results obtained are presented in **Table 3**. The effective dose due to external exposure varied from 1.2 to 2.1 μSv·y^{-1} and that of internal exposure varied from 31.6 to 59.1 μSv·y^{-1}, showing a higher inhalation dose. The total annual effective dose varied between 33.0 and 61.2 μSv·y^{-1}, with a mean value of 47.1 ± 8.4 μSv·y^{-1}. The mean of the estimated annual effective doses is below the intervention exemption level (1.0 mSv·y^{-1}) given in the International Commission on Radiological Protection Publication 82 [20]. However, it is recommended that adequate protective means should be ensured in order to minimize the occupational dose.

3.3. Radiation Hazard of Fly Ash as Building Material

Equations (4)-(6) had been used to determine the suitability of the fly ash for use as building material as far as radiation is concerned and the results are shown in **Table 4**. The value of Ra_{eq} ranged from 107 to 180 Bq·kg^{-1}. The values of the hazard indices ranged between 0.29 and 0.49 for H_{ext} and 0.36 and 0.66 for H_{int}. The mean values of Ra_{eq}, H_{ext} and H_{int} are 136 ± 19 Bq·kg^{-1}, 0.37 ± 0.05 and 0.48 ± 0.08, respectively. The errors quoted are the standard deviations from the mean values. All the results obtained are lower than the maximum recom-

Table 3. Calculated effective dose ($\mu Sv \cdot y^{-1}$) for workers.

S/N	E_{ext}	E_{int}	Total annual effective dose
1	2	38.2	40.2
2	1.5	38.9	40.4
3	1.7	40.1	41.8
4	1.7	38.8	40.5
5	1.6	41.7	43.3
6	1.7	56.1	57.8
7	1.2	51.1	52.3
8	1.4	31.6	33
9	1.6	40.3	41.9
10	1.5	37.9	39.4
11	1.7	53.6	55.3
12	1.3	42.2	43.5
13	1.6	45.6	47.2
14	1.3	38.4	39.7
15	1.3	37.9	39.2
16	1.7	50.7	52.4
17	1.7	51.8	53.5
18	2.1	59.1	61.2
19	1.3	57.3	58.6
20	1.5	55.6	57.1
Mean ± Std Dev	1.6 ± 0.2	45.5 ± 8.2	47.1 ± 8.4

mended limits for building materials; 370 $Bq \cdot kg^{-1}$ for Ra_{eq}, and 1 (unity) for H_{ext} and H_{int}. This is an indication that the use of the fly ash as building material does not pose any significant hazards as far as radiation is concerned.

3.4. Outdoor Radiation Dose

The outdoor radiation dose at 1 m above the ground was calculated using the mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the fly ash from the dump site and soil samples in the vicinity of the plant and Equation (7). The results obtained are presented in **Table 5**. The absorbed dose rate was 49.7 ± 4.0 $nGy \cdot h^{-1}$ for coarse fly ash collected from the dump site. Those of the soil samples were 51.7 ± 3.6 and 49.4 ± 4.9 $nGy \cdot h^{-1}$, respectively for 10 m and 100 m distances. All are below world population weighted mean of 60 $nGy \cdot h^{-1}$ [13].

The annual effective doses had been calculated assuming an occupancy factor of 0.2 in Equation (8). The results are within the range of 0.30 ± 0.02 and 0.32 ± 0.02 mSv, which is lower than the annual 1 mSv recommended for the public by the International Commission on Radiological Protection [21].

4. Conclusion

Ashes produced in thermal power plants may contain high levels of natural radioactivity and constitute a potential health hazard to the power plant personnel, and to the population living in the vicinity, due to fly-ash releases, fly ash depositions and fly-ash industrial utilization. The radioactivity of fly ash samples and soil samples in

Table 4. Radium activity concentrations (Ra_{eq}), external (H_{ext}) and internal (H_{int}) hazard indices of fly ash.

S/N	Ra_{eq} (Bq·kg^{-1})	H_{ext}	H_{int}
1	141	0.38	0.54
2	121	0.33	0.43
3	136	0.37	0.51
4	131	0.35	0.48
5	129	0.35	0.46
6	160	0.43	0.56
7	127	0.34	0.4
8	107	0.29	0.39
9	128	0.35	0.46
10	120	0.32	0.43
11	155	0.42	0.55
12	122	0.33	0.4
13	139	0.38	0.5
14	111	0.3	0.36
15	111	0.3	0.37
16	151	0.41	0.54
17	150	0.4	0.52
18	180	0.49	0.66
19	145	0.39	0.47
20	150	0.41	0.51
Mean ± Std Dev	136 ± 19	0.37 ± 0.05	0.48 ± 0.08

Table 5. Outdoor absorbed dose rate (D) and annual effective dose (H).

Sample	No.	D (nGy·h^{-1})	H (mSv·y^{-1})
Coarse fly ash	10	49.7 ± 4.0	0.30 ± 0.02
Soil (10 m)	10	51.7 ± 3.6	0.32 ± 0.02
Soil (100 m)	10	49.4 ± 4.9	0.30 ± 0.03

the vicinity of the thermal plant was determined using gamma ray spectroscopy method. The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the fly ash were higher than other samples. However, they were below the world average for soil and building materials. The occupational dose obtained was very low compared to the recommended occupational dose. The radium equivalent activity, the external and internal hazard indices of the fly ash were below the recommended limit. The annual effective doses estimated were within the recommended limit of 1 mSv for the public. From radiological point of view, it can therefore be concluded that workers and the people in the vicinity of the thermal plant are not at risk to significant radiological hazard.

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