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Full Length Research Paper

Radionuclide concentration: The coal ash effect

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Radionuclide concentrations in coal ash, soil mixed with coal ash and soil were determined using the gamma spectroscopic method. The average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in coal ash samples were found as 35.0, 5.8, and 109.4 Bq/kg, respectively. The average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil mixed with coal ash samples varied from 21.2 to 30.8, 48.6 to 82.7, and 162.5 to 180.1 Bq/kg, respectively. The activity concentrations of radionuclides from the non-mixed soil area, which serves as a control to this study ranged from 123.95 to 128.82, 16.33 to 18.38, and 46.63 to 50.30 Bq/kg, respectively for radionuclides ^{40}K, ^{226}Ra, and ^{232}Th, respectively. From the radiological calculations, it was found that the use of coal ash as a soil additive contributed slightly to the radionuclide concentrations of the area under study.

Key words: Radionuclide, coal ash, radium equivalent, external and internal hazard index.

INTRODUCTION

External exposures to radiation arise from terrestrial radionuclides present at trace levels in soil, rocks and building materials. Only those radionuclides with half-lives comparable to the age of the earth and their decay products exist in significant quantities in these materials. Irradiation is mainly by gamma radiation from radionuclides ^{238}U and ^{232}Th series and from ^{40}K. Reports have shown that the collective occupational exposure to natural sources, significantly above background levels, is higher than that due to man-made sources (UNSCEAR, 2015).

Many processes in non-nuclear industry create a situation where the concentration of naturally occurring radioactive material is additionally enhanced (Solomon, 2006). Such situation usually exists in industrial processes where a significant mass reduction of raw materials occurs, typically with changes of their chemical composition or state of aggregation which might further influence their properties. As a matter of course, these industries of concern are not aimed at the production of natural radionuclides or the deliberate use of radiation. Therefore, radioactive isotopes are usually accumulated in the waste. Such alterations to the natural state result in an increment of radiation risk to people as well as to the

environment in case of disposal of such waste. Each particular occurrence of natural radioactivity presents a unique scenario of exposure, usually different from those caused by artificial radionuclides.

Although alternative energy sources are gaining increasing importance, fossil fuels, especially coal, play a significant role in the global energy production. Unfortunately, coal combustion generates a large amount of Coal Combustion Products (CCPs), including coal ash. Coal ash has substantial negative effect on environment, while its properties could make it a desired secondary raw material in many industry branches (Lloyd, 2002).

Coal ash is often used in agriculture, because of its special physical and chemical properties (Lloyd, 2002). Coal ash has a unique structure and contains almost all the nutrients necessary for proper plants growth and development

It can also be applied as an agent, which increases plant growth and amount of the obtained yields. In addition, coal ash is used to reduce the amount of heavy metals accumulated in plants and to limit the spread of diseases (Szponder and Trybalski, 2011).

Although coal ash application in agriculture has many benefits, there are some disadvantages connected with this application for example heavy metals and radionuclides contamination of soils and surface waters, soil salinity (Szponder and Trybalski, 2011). Therefore, it is necessary to continue research on this topic to clarify the effects of the coal ash addition to the soil on agricultural production and the environment from a radiological view. Therefore, this work is aimed at determining the contribution of coal ash to soil radionuclide concentration by measuring the radionuclide concentrations in farms treated with coal ash and farms with no applications of coal ash within the same geographic location.

MATERIALS AND METHODS

Sample collection and preparation

Samples were collected at Oji-river, Enugu state, Nigeria, where coal is utilized as fuel for the coal fired electric power station. Two coal ash samples were collected from the base of the plant furnace; five soil samples were taken from farms that utilize coal ash as a soil additive. While three soil samples were also collected from farms where coal ash is not utilized for farming purposes. About 0.5 kg of each sample was collected and these were prepared for gamma activity concentration measurement. The samples were labeled as coal ash 1 and 2 for the coal ash samples; MCA1-5 for the soil mixed with coal ash and OJC1-3 for the soil samples with no coal ash contamination. The OJC samples served as the control sample used to compare the radionuclide content contribution of the coal ash in the environment under study.

Sample preparation for gamma analysis

All the collected samples were dried under direct sun and humidity condition for 5 days. Soils were well mixed after removing

extraneous materials such as roots, mat portions, pieces of stone and gravel. Samples were weighed and dried into an electric oven at 105°C until a constant dry weight was obtained. The soil samples were sieved to grain size of less than 0.63 mm and were scaled in a plastic cylindrical container of about 250 cm^3 and labeled. The samples were kept for at least 30 days before the measurement to reach secular equilibrium between thorium and radium and their decay products.

Gamma ray spectroscopy

The gamma-ray spectrometry set-up is made up of a 7.62 × 7.62 cm^2 NaI(Tl) detector housed in a 6 cm thick lead shield (to assist in the reduction of the background radiation) and lined with cadmium and copper sheets (CERT manual, 1999). The samples were placed on the detector surface and each counted for about 29,000 s in reproducible sample detector geometry. The configuration and geometry was maintained throughout the analysis, as previously characterized based on well established protocol of the laboratory (at the Centre for Energy Research and Training, Zaria). A computer based Multichannel Analyser (MCA) MAESTRO Programme from ORTEC was used for data acquisition and analysis of gamma spectra. The 1764 keV Gamma-line of ^{214}Bi for ^{238}U was used in the assessment of the activity concentration of ^{226}Ra, while 2614.5 keV Gamma-line of ^{208}Tl was used for ^{232}Th. The single 1460 keV Gamma-line of ^{40}K was used in its content evaluation.

The activity concentration (C) in Bq/kg of the radionuclides in the samples was calculated after decay correction using the expression (Avwiri et al., 2013):

$$C_s(Bq/kg) = \frac{C_a}{\varepsilon_y \times M_s \times t_c \times P_y} \tag{1}$$

where C_s is the sample concentration, C_a is the net peak area of a peak at energy, ε_y is the efficiency of the detector for a γ-energy of interest, M_s is the sample mass, t_c is the total counting time, P_y is the abundance of the γ-line in a radionuclide.

Radiological assessment of coal ash as a soil nutrient supplement

The radiological equivalence activity concentration (Ra$_{eq}$), external hazard (H$_{ex}$), and internal hazard (H$_{in}$) indices were calculated using the following equations, respectively (Ademola and Onyema, 2014; Hasan et al., 2014):

$$Ra_{eq} = 0.077A_k + 1.43A_{Th} + A_{Ra} \tag{2}$$

$$H_{ext} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \tag{3}$$

$$H_{in} = \frac{C_k}{4810} + \frac{C_{Th}}{259} + \frac{C_{Ra}}{185} \tag{4}$$

Radiation doses

The radiation doses were calculated for outdoor annual radiation dose and external outdoor absorbed gamma dose from the radionuclides were calculated using Equations 5 and 6, respectively (Solehah et al., 2016; Faanu et al., 2016).

Table 1. Activity concentration of Radionuclides in coal ash.

Sample	^{40}K	^{226}Ra	^{232}Th
coal ash 1	274.84±1.55	8.57±1.04	33.29±1.25
coal ash2	278.90±0.48	8.94±0.94	39.40±0.19
Mean coal	109.43±4.89	5.75±1.21	35.01±1.20

Table 2. Activity concentration of radionuclides in soil samples of Oji-River mixed soil and coal ash (Bq/kg).

Sample code	$^{40}K_{soil}$	$^{226}Ra_{soil}$	$^{232}Th_{soil}$
MCA01	180.09±6.37	23.75±4.40	48.57±2.16
MCA02	169.36±5.88	30.82±2.31	73.54±1.25
MCA03	172.16±8.08	23.52±6.14	82.66±9.12
MCA04	168.93±5.43	21.23±2.03	76.43±1.03
MCA05	162.47±5.30	24.10±4.29	81.03±4.38
Mean	170.60±6.21	24.68±3.83	72.44±3.58

Table 3. Activity concentration of radionuclides in soil samples of Oji-River- non mixed soil (Bq/kg).

Sample code	$^{40}K_{soil}$	$^{226}Ra_{soil}$	$^{232}Th_{soil}$
OJC01	123.95±8.55	16.33±1.96	46.63±1.82
OJC02	124.37±5.03	18.38±1.20	48.82±1.38
OJC03	128.82±7.14	17.33±0.32	50.30±2.10
Mean	125.71±6.90	17.34±1.16	48.58±1.76

$$D(nGy.h^{-1}) = 0.0417A_k + 0.604A_{Th} + 0.462A_{Ra} \quad (5)$$

$$E(mSv.y^{-1}) = D \times 24 \times 365.25 \times 0.2 \times 0.7 \times 10^{-6} \quad (6)$$

RESULTS AND DISCUSSION

From the gamma spectroscopic analysis of the coal ash, mixed soil, and coal ash samples and the non-mixed soil and coal ash samples, the results obtained for the radionuclides concentrations are shown in Tables 1, 2, and 3. From the gamma spectroscopic analysis, no anthropogenic radionuclides was obtained, only the primordial radionuclides ^{40}K, ^{226}Ra of the ^{238}U decay series and ^{232}Th were observed.

Two samples of the coal ash taken from the bottom of the furnace, showed mean radionuclide concentrations for ^{40}K, ^{226}Ra and ^{232}Th to be 109.43, 5.75, and 35.01 Bq/kg, respectively. This activity concentration in the coal ash from Oji-River thermal power plant was found to be lower than what was obtained in Brazilian coal (Flues et al., 2007), when also compared with the works of (Mondal et al., 2006), the coal ash from the Enugu coal also showed to have less radionuclides than their Indian counterparts. The result also, when compared with the

work of Mokobia (2010) on the radiometric assessment of Nigerian fossil fuels were higher than what was obtained for the raw coal. This suggests an increment in the radionuclide concentration in the burned coal ash.

The radionuclides concentrations for the mixed soil and coal ash ranged from 162.47 to 180.09, 21.23 to 30.82 and 48.57 to 82.66 Bq/kg for the radionuclides ^{40}K, ^{226}Ra, and ^{232}Th, respectively. This result was found to be similar to what Amakom et al. (2017) found in the soils around the Enugu coal mines. The result is also comparable to what was obtained around a cement factory in Ghana by Addo et al. (2013) and also similar to what was found in quarry dust around the Abakaliki quarries by Ugwu et al. (2008).

The activity concentrations of radionuclides from the non-mixed soil area, which serves as a control to this study ranged from 123.95 to 128.82, 16.33 to 18.38, and 46.63 to 50.30 Bq/kg for the radionuclides ^{40}K, ^{226}Ra and ^{232}Th, respectively. This was found to be lower than those found for the mixed soil and coal ash area. The result was also similar to the results of Avwiri et al. (2011, 2013).

This suggests that using coal ash as a soil additive may have contributed significantly in the radionuclide burden of the area.

Table 4. Calculated radiation and radiological parameters.

Parameter	Radiological equivalence of radionuclides in coal ash (Ra$_{eq}$)	H$_{ext}$ of radionuclides in coal ash	H$_{in}$ of Radionuclides in coal ash	Absorbed dose rate $D(nGy.h^{-1})$	Annual effective dose $E(mSv.y^{-1})$
Coal ash	51.6586	0.1594	0.1890	28.36	0.3480
Mixed coal ash and soil	141.35	0.3818	0.4484	62.26	0.7640
Non mixed soil	96.489	0.2604	03074	42.5953	0.5227
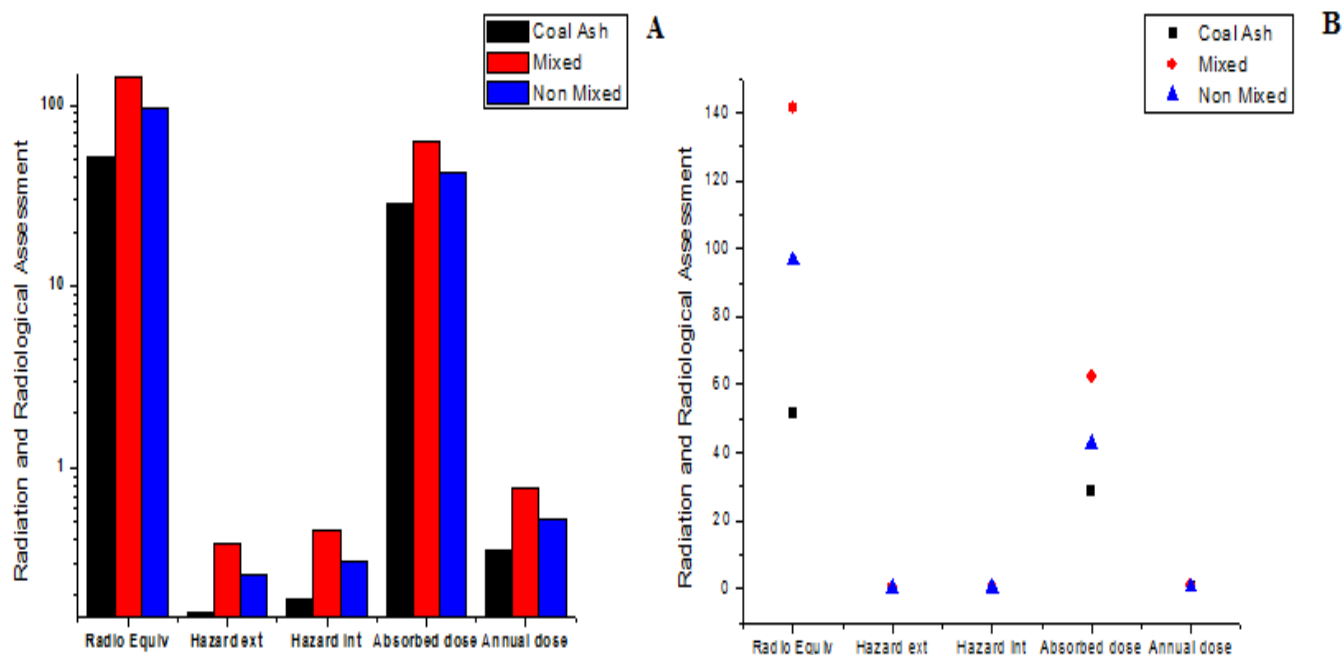
Figure 1. Pattern recognition for radiation and radiological parameters.

Radiological assessment

The average values of the activity concentration were used to calculate the radium equivalent, absorbed dose rate, annual effective dose, and external and internal hazard indices. The results obtained are shown in Table 4 and is statistically expressed in Figure 1. Figure 1A shows that mixed coal ash has the highest determined value in all the parameters calculated and followed a trend of mixed coal ash > non mixed > coal ash for the primordial radionuclides observed in the samples. Additionally, among the parameters determined, the trend pattern was observed to be radium equivalence > absorbed dose > annual dose > hazard internal > hazard external. The same trend is also shown on the scatter plot of Figure 1B. There was no significant difference between the determined levels of hazard internal, hazard external and annual dose of the samples obtained from coal ash, mixed coal ash and non-mixed coal ash, respectively.

The annual effective doses obtained for mixed coal ash and non-mixed coal ash were found to exceed the world average value of 0.46 mSvyear^{-1} (Hasan et al., 2014), this could be a source of possible health concern when used for agricultural purposes over a long period.

The mean values of the samples radium equivalence were below the limit of 370 Bqkg^{-1} limit; hence, the coal ash may be safe when applied as agricultural supplement (Hasan et al., 2014). This also suggests its safe used as a construction material (Amakom and Aghamelu, 2013). Amongst the samples, only the mixed coal ash has absorbed dose greater than the world average value of 55 nGyh^{-1}.

Finally, the external and internal indices of the three samples of coal ash, mixed soil and non-mixed soil were below the standard of 1 which is the acceptable international standard (Solehah et al., 2016). Thus, there is no significant risk posed by the coal ash and non-mixed soil as similarly observed by Kolo et al. (2016) and Ademola and Onyema (2014). On the other hand,

immediate radiological risk exists for mixed soil due to soil and coal ash interaction (Kolo et al., 2016).

From the radiological point of view, this study was able to show that using coal ash for agricultural activities, somewhat contributed to the radionuclide concentration of the area. This could be as a result of mass reduction of the chemical components of the coal itself during the firing operations in the power plants. Care should be taken when using coal ash as a soil additive for agricultural purposes as its continuous application might lead to concentrations of these radionuclides to the proportion to which it constitutes a serious radiation hazard to the environment.

CONFLICT OF INTERESTS

The authors have not declared any conflict of interests.

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