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Original Research Article

Radiological Risk Assessment of Technologically Enhanced Naturally Occurring Radioactive Materials in coal wastes from Van Eck Coal-Fired thermal Power Plant, Namibia.

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ABSTRACT

In this study, the concentrations of primordial radioactive nuclides ^{226}Ra, ^{232}Th, and ^{40}K and their potential for causing radiological health hazards were measured using coaxial high-purity germanium (HPGe) gamma detector in coal, bottom, and fly ashes collected from the 120 MW Van Eck thermal power plant, Windhoek, Namibia. The experimental results showed that for coal samples, the activities of ^{226}Ra, ^{232}Th, and ^{40}K were 20.22 ± 3.82 Bq/kg, 26.70 ± 5.90 Bq/kg, and 30.36 ± 6.36 Bq/kg, respectively. For the bottom ash layer samples, the activities of ^{226}Ra, ^{232}Th, and ^{40}K were 88.89 ± 11.91 Bq/kg, 113.28 ± 15.44 Bq/kg, and 97.84 ± 16.55 Bq/kg, respectively whereas for the fly ash the values are 59.18 ± 4.55 Bq/kg, 77.00 ± 5.89 Bq/kg and 77.17 ± 7.30 Bq/kg, respectively. The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K were all enhanced in the bottom and fly ash relative to coal samples. However, the estimated radiological health hazards and the concentration of the various samples from the selected location were within the permissible limits provided for human safety and environmental protection.

1. Introduction

Coal residues and waste produced by the combustion of the coal contain naturally occurring radionuclides such as ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K which can be dispersed into the environment through the combustion processes [1]. The mobilization and dispersion of these radioactive materials into the atmosphere and human environment have been associated with significant human health challenges [2]. In recent decades, there has been a growing awareness by professionals on the impact of thermal power plants which use, for various applications, raw materials such as lignite, bituminous coal, and high ash and S-bearing coals naturally rich in uranium, thorium, or radium [3].

In this study, the focus was on the Van Eck Power Plant that produces electricity based on the combustion of bituminous coal. In most instances, the mechanism used in the combustion of coal will enhance the concentration of long-lived radionuclides in the raw material which is termed as Technologically Enhanced Naturally Occurring Radioactive Materials (TENORMs). These combustion processes generate large amounts of solid residues in the form of fly and bottom ashes [4]. Considering the rapid industrialisations, the Van Eck coal-fired power plant will continue to play an important role in the medium-term energy requirement of Namibia which serves as part of a diverse energy mix to ensure the security of supply, reducing the over-dependence on imports and mitigating the diverse impacts on the balance of payment. However, the management of waste generated presents a major challenge for both the power plant and the surrounding environment [5]. Particularly, fly ash particles, entrained up the stack in the flue gas stream, have a greater tendency to absorb trace elements such as natural radionuclides during combustion owing to their relatively small size and large surface area [6, 7].

According to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), the combustion of coal in a coal-fired power plant can lead

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to an increased concentration of natural radionuclides in the environment through the pathways of ash deposition of non-combustible elements with enrichments factor of 5–10 times the average concentrations of the primordial radionuclides (^{40}K, ^{238}U and ^{232}Th) [8, 9]. However, with changes in environmental consciousness throughout the world, there is also a growing tendency of utilizing industrial by-products such as fly ash. The storage of waste resulting from the combustion of coal in thermal power plants can potentially lead to significant environmental and human health problems. Therefore, utilization of ashes as a secondary raw material such as an additive in concrete construction, and road construction, and asphalt's mixture has clear environmental and economic advantages [10].

However, coal-powered plants (CPPs) that produce TENORMs during their operation should be investigated from the radiological point of view as an additional radiation source. Moreover, for the utilization of these residues, it is of importance to evaluate the physical parameters, chemical composition, and radiological features of these materials. Therefore, monitoring of technologically naturally occurring radioactive materials is an important aspect from a human health and environment protection perspective. This study provides the much-needed data on, enrichment, and radiological characteristics associated with the utilization of waste as secondary raw materials.

2. Material and Methods

2.1 Sampling

The study focused on the Van Eck Coal-fired Thermal Power Plant located in the northern industrial area in Windhoek, Namibia, and is part of an extensive industrial area (heavy and light industry). The plant comprises four generating units with a combined output of 120-Megawatt (MW) per annum. The power plant uses approximately 650 kg of bituminous coal per MWh of electricity generated. The coal used is imported from neighbouring countries Botswana and South Africa based on product costs. The power station has been in operation since the early seventies and is a sub-critical thermal power plant.

2.2 Sample preparation and analysis

In this study, all samples were measured with high-purity germanium (HPGe) detector for the determination of the activities of ^{226}Ra (^{238}U), ^{232}Th, and ^{40}K. About 1000 g of coal, fly, and bottom ash samples were collected from a heap of coal waste where they are stored temporarily within the plant area using purposive random sampling. A total of fifty-four (54) samples comprising of eighteen Coal (numbered C-01 to C-18), eighteen bottom ash samples (numbered CA-01 to CA-18) and fly ash samples (numbered FA-01 to FA-18) were collected and kept in cleaned and numbered polyethylene bags. All collected samples were air-dried at ambient temperature in the laboratory for seventy-two (72) hours to ensure moisture-free samples.

2.3 Gamma-ray Spectrometry

The samples were oven-dried at 80 degrees Celsius for 12 hours to attain constant weight. The dried coal and bottom ash samples were thoroughly pulverised, sieved, and homogenised. About 600 ± 0.001 g of the homogenised samples were carefully packed in well-labelled 500 ml Marinelli beakers. To achieve secular equilibrium in the ^{238}U series (between ^{226}Ra and its daughters) and the ^{232}Th series (between ^{228}Th and its daughters), the samples were sealed hermetically to avoid (^{222}Rn and ^{220}Rn) emanation. The sealed samples were then stored for about +30 days (the equivalent of 7 half-live of ^{222}Rn) for ^{226}Ra and ^{222}Rn and its short-lived gamma-emitting decay products, ^{214}Bi and ^{214}Pb before being measured by gamma-ray spectrometry [11, 12].

The radioactivity in the collected samples was measured using a coaxial (62.80 X 64.80 mm) Canberra gamma-ray spectrometer HPGe detector Model No. GC4520 SN 10882 with 45% relative efficiency and resolution of 2.00 KeV (FWHM) at 1.33 MeV peak of Co-60 and 1.200 keV (FWHM) at 122 keV. The gamma spectrometry system was calibrated for energy and efficiency using a multi-nuclide calibration standard with an initial activity of 40 kBq homogeneously distributed in silicone matrix, which was supplied by Eckert & Ziegler Nuclitec GmbH, Germany, SN. AM 5599 and validated using IAEA NORMs reference material RGK-1, RGTh-1, and RGU-1. A computer-based Multichannel Analyser (MCA) Genie 2000 software from Canberra, Australia was used for data acquisition and analysis of gamma spectra. The samples were counted for 43200 s in a reproducible sample detector geometry and the configuration, and geometry was maintained throughout the analysis.

The 295.22 keV, 351.93 keV for ^{214}Pb and 609.32 keV, 1120.29 keV and 1764.49 keV for ^{214}Bi gamma lines were used in the assessment of activity concentration of ^{226}Ra, while 911.21 keV for ^{228}Ac, 968.97 keV and 238.63 keV for ^{212}Pb were used for ^{232}Th. The single 1460 keV Gamma-line of ^{40}K was used in its content evaluation.

The activity concentration of individual radionuclides in all samples investigated was calculated using the following analytical expression [13].

$$A(Bqkg^{-1}) = \frac{N}{\epsilon_\gamma P_\gamma T_s M} \ldots\ldots\ldots\ldots\ldots\ldots 1$$

where A is the specific activity in Bq/kg of each radionuclide in the sample, N is the net peak count rate of the resulting photo-peak, ε_γ is the detector efficiency of the specific gamma-ray, P_γ is the gamma emission probability of the corresponding gamma energy, T_s is the counting time of the sample and M is the sample mass in kg.

The error associated with every activity calculation was computed by the standard deviation equation derived from the uncertainty budget. The equal counting time for both background and sample was chosen to minimize the uncertainty in the net counts,

$$\Delta A = A\sqrt{(\frac{\Delta N}{N})^2 + (\frac{\Delta \varepsilon_\gamma}{\varepsilon_\gamma})^2 + (\frac{\Delta P_\gamma}{P_\gamma})^2 + (\frac{\Delta M}{M})^2 + (\frac{\Delta T_s}{T_s})^2} \ldots\ldots\ldots 2$$

where ΔA is the uncertainty of the sample measured and ΔN, Δεγ, ΔPγ, ΔM, and ΔTs are the uncertainties of the net count rate, efficiency, gamma emission probability, sample weight, and counting time respectively.

2.4 Radiological assessment of bottom and fly ash as secondary raw material in construction industries

The radiological equivalence activity concentration (Ra_{eq}), the absorbed gamma dose rate (DR), annual gonadal effective dose equivalent (AGDE), annual effective dose equivalent (AEDE), activity utilization index (AUI), external hazard index (H_{ex}), internal hazard index (H_{in}), representative gamma index (I_γ) and excess lifetime cancer risk (ELCR) were computed using the following equations, respectively.

2.4.1 Radium equivalent activity (Ra_{eq})

Ra_{eq} defined based on the assumption that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th, and 4810 Bq/kg of ^{40}K produce the same gamma-ray dose is calculated as follow:

$$Ra_{eq}(Bqkg^{-1}) = A_{Ra} + 1.43A_{Th} + 0.077A_K \ldots\ldots\ldots 3$$

The Ra_{eq} is also used as the screening tool for radioactivity evaluation of construction materials.

2.4.2 External hazard index (H_{ex})

The quantification of the incurred radiation hazard due to external exposure to gamma-ray from the environmental sample was assessed by the external hazard index as follow:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \ldots\ldots\ldots\ldots\ldots\ldots 4$$

2.4.3 Internal hazard index (H_{in})

The radiation hazard administered to the respiratory organs from radon and its short-lived radionuclides was quantified by assessing the internal hazard index (H_{in}) as given by [14]

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \leq 1 \ldots\ldots\ldots 5$$

2.4.4 Representative gamma index (I_γ)

The gamma radiation hazard due to the respective concentration of the investigated natural radionuclides was assessed by the representative gamma index. The index serves as a screening parameter for the material of possible radiation health challenges.

$$I_\gamma = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \leq 1 \ldots\ldots\ldots 6$$

2.4.5 Activity utilization index (AUI)

The natural radionuclides activity utilization index (AUI) was used to express the dose rate in the air from different combinations of the three primordial radionuclides in the studied sample. The appropriate factor to the measured specific activity of the respective nuclides was applied and the AUI was calculated from the equation [15, 16]

$$AUI = \left[\frac{A_{Ra}}{50\ Bqkg^{-1}}\right] f_u + \left[\frac{A_{Th}}{50Bqkg^{-1}}\right] f_{Th} + \left[\frac{A_K}{500Bqkq^{-1}}\right] f_K \ldots\ldots 7$$

where A_{Ra}, A_{Th}, and A_K are the specific activities of ^{226}Ra, ^{232}Th, and ^{40}K respectively and f_u (0.041), f_{Th} (0.604), and f_K (0.462) are the respective fractional contribution from the actual activities of these radionuclides to the total gamma radiation dose rate in air.

2.5 Radiation doses

The radiation doses were calculated for total annual effective dose equivalent (AEDE), external outdoor absorbed gamma dose, and excess lifetime cancer risk (ELCR) from the radionuclides were calculated using Equations 8-10, respectively

2.5.1 Annual effective dose equivalent (AEDE)

The outdoor and indoor AEDE in mSv from the radioactivity content of the sample was calculated applying two conversion coefficients provided by UNCEAR (2000) as follow:

$$AEDE(mSvy^{-1}) = D_R(nGyhr^{-1}) \times 8760\frac{h}{y} \times \frac{10^{-6}}{10^{-9}}\frac{mGy}{Gy} \times 0.7 \times 0.2\frac{Sv}{Gy}$$

or

$$AEDE(mSvy^{-1}) = D_R \times 1.226 \times 10^{-3} \ldots\ldots\ldots 8$$

The two conversions are the coefficient factor from absorbed dose in the air to an effective dose, given as 0.7Sv/Gy, and the outdoor occupancy factor of 0.2 based on the assumption that an individual spends on average of 80% of his time indoors.

2.5.2 Gamma absorbed dose rate (D_R)

The gamma absorbed dose rate 1 m above the air was computed as per the UNSCEAR report (2000) using the equation:

$$D_R(nGyh^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \ldots\ldots\ldots 9$$

2.5.3 Excess lifetime cancer risk (ELCR)

After the evaluation of the annual effective dose equivalent, excess lifetime cancer risk (ELCR) was estimated using the equation [13, 16]

$$ELCR = AEDE \times DL \times RF \ldots\ldots\ldots 10$$

where AEDE, DL, and RF are the annual effective dose equivalent, duration of life (70 years), and risk factor (0.05 Sv^{-1}) for stochastic effects in any given population respectively. Data obtained from this study were subjected to Pearson's correlation analysis using statistical software; the statistical package for social sciences (SPSS 22.0). This was necessary to understand and establish interdependency and mutual relationships that may exist among the measured radiological variables.

2.6 Enrichment factor

The enrichment factor (EF) defined as the ratio of activity concentration of the radionuclides [X] and ^{40}K in bottom ash or fly ash sample divided by the corresponding ratio of the feed coal sample was determined by the formula used by [16, 17] as follow

$$EF = \frac{([x]ash\ sample/40_K ash\ sample)}{([X]feed\ coal/40_k feed\ coal)} > 1 \ldots\ldots\ldots 11$$

This has the effect of normalizing the apparent enrichment resulting from the loss of carbon during the combustion process. The ^{40}K is used for the normalization process because its activity concentration remains constant in all types of fly ashes and is assumed to be a tracer for the aluminosilicate-dominated fly ash matrix.

3. Results and Discussion

3.1 Activity Concentration in Bq/kg of ^{226}Ra, ^{232}Th, and ^{40}K in Coal, Bottom ash and fly ash.

The statistical description of activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, comprising the mean and standard deviation (SD) are presented in Figure 1 for the studied samples. Table 1 represents the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K for the present study compared to other similar work. From Figure 1 and Table 1, the mean activity concentration of 20.22 ± 3.82 Bq/kg for ^{226}Ra, 26.70 ± 5.90 Bq/kg for ^{232}Th, and 30.36 ± 6.36 Bq/kg for ^{40}K in coal samples were in accordance with the world average values and with studies of [4, 5, 9]. Meanwhile, in Table 2 the relatively enhanced activity concentration of 88.89 ± 11.91 Bq/kg for ^{226}Ra, 113.28 ± 15.44 Bq/kg for ^{232}Th and 97.84 ± 16.55 Bq/kg for ^{40}K in bottom ash, and 59.18 ± 4.55 Bq/kg for ^{226}Ra, 77.00 ± 5.89 Bq/kg for ^{232}Th, and 77.17 ± 7.30 Bq/kg for ^{40}K in fly ash samples were in agreement with studies of [9, 18].

Indeed, the study shows that the combustion of coal significantly enhances the activity concentration of the radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K (Table 1). These variations in activity concentration could be attributed to the radionuclide's enrichment and volatilization before combustion due to ^{226}Ra and ^{232}Th which may have been in equilibrium in the feed coal samples. After combustion, the equilibrium state may be disturbed resulting in the disturbance of the degree of portioning of the radionuclides between the bottom and fly ash which is normally influenced by their volatility and geochemical association [19, 20].

These results further indicate the well-known pattern that uranium and its progenies are usually associated with the organic compound of coal, while thorium and its decay radionuclides together with potassium are associated with the inorganic material of coal [20]. The variations are also found in studies around the world Table 2. The activity concentration ranged from 97.3 Bq/kg (India) [21] to 1909 Bq/kg (Brazil) [19] for ^{226}Ra; from 32 Bq/kg (Turkey) [8] to 455 Bq/kg (Tanzania) [22] for ^{232}Th and 323 Bq/kg (Turkey) [8] to 3069 Bq/kg (Tanzania) [22] for ^{40}K.

Table 1: Activity concentrations (Bq/kg) of ^{226}Ra, ^{232}Th, and ^{40}K in coal with the world's average values and those of other published works

References	Coal		
	^{226}Ra	^{232}Th	^{40}K
[UNSCEAR 2008]	20	20	50
Present study	20.22	26.71	30.36
Nigeria [5]	34.18	18.18	70
Nigeria [18]	31.51	26.91	90.67
Botswana [9]	18.1	27.43	177.38
Kolo [4]	8.0	7.0	27.4

Figure 1: Mean activity concentrations in the bottom and fly ash samples.

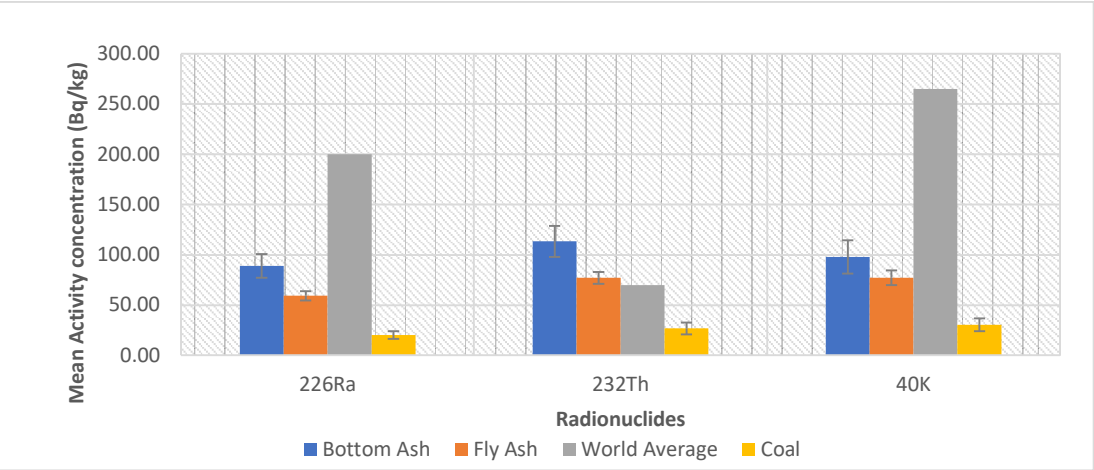


Table 2: Activity concentrations (Bq/kg) of ^{226}Ra, ^{232}Th, and ^{40}K in ashes with the world's average values and those of other published works

References	Ashes		
	^{226}Ra	^{232}Th	^{40}K
[UNSCEAR, 2008]	200	265	70
Present study	59.18	77.00	77.70
Brazil [19]	1909	58	650
Nigeria [18]	28.2	37.6	335
Botswana [9]	49.37	64.54	40.08
India [21]	97.3	107.5	315.8
Tanzania [22]	448	455	3069
Turkey [8]	815	32	323

3.2 Radiological Assessment of Bottom and Fly ash

The utilization of ashes as a secondary raw material or supplementary materials in the production of construction materials either for building or as aggregate in concrete can result in external gamma exposure to members of the public due to the presence of natural radionuclides. The radiological assessment was estimated based on human health and environmental protection perspective. Radium equivalent activity Ra_{eq} and gamma representative index I_γ were used as the screening parameters. The average activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were used to estimate the radiological hazards. The results obtained are shown in Table 3.

The estimated values of Ra_{eq} ranged from 175.23 Bq/kg to 258.42 Bq/kg and are presented in Table 3. The average values obtained were within the world precautionary limit of 370 Bq/kg. The elevated activity concentration observed in ashes samples resulted in an estimated mean absorbed dose rate 1 m above the ground exceeding the recommended world average value of (60 nGy/y) (Figure 1). However, their corresponding total annual effective dose equivalent of 0.14 mSv/y and 0.09 mSv/y were within the public's annual effective dose rate threshold of 1 mSv/y and 20 mSv/y recommended for occupational exposure. The estimated values for AUI, H_{ex}, H_{in}, and $I_{\gamma r}$ were all comparable to the world average value of unity. In addition, the estimated lifetime cancer risk was relatively within the world safety limit of 0.05 for low-level radiation limit by the International Commission on Radiation Protection (ICRP).

The study found that the radiological indices of Ra_{eq}, I_γ, H_{in}, and H_{ex} were generally high for bottom ash than for fly ash samples. These relatively high values of Ra_{eq}, I_γ, H_{in} and H_{ex} could be attributed to the fact that when coal is combusted, most of the non-combustible material, which includes the natural radionuclides, remained, and concentrates in the bottom and fly ash thereby enhancing the Ra_{eq}, I_γ, H_{in} and H_{ex} values [17,20, 23].

3.3 Statistical analysis

Based on the data set of the activity concentrations and estimated radiation hazard indices for the studied samples, a correlation matrix was calculated and presented in Table 4-6. The results showed that there is a strong positive correlation of ($r \geq +0.66$) with radiation hazard indices and with ^{226}Ra, ^{232}Th, and ^{40}K activity for coal and fly ash samples. These results demonstrate the presence of radiation in coal and fly ash at the Van Eck power plant which is homogeneously distributed among the primordial radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K. It can, therefore, be inferred that ^{226}Ra, ^{232}Th, and ^{40}K are the major contributors to radiation emissions in fly ash from Van Eck Power Plant. However, the existence of a positive correlation between ^{226}Ra, ^{232}Th, and ^{40}K may be independent of the nature of radium and thorium decay series origin. A very weak positive degree of association was observed in the bottom ash sample between $^{40}K/^{226}Ra$ (r=0.26), $^{40}K/^{232}Th$ (r=0.30), and ^{40}K ($0.37<r<0.32$) activity with all the radiation hazard indices. These results indicate that the gamma-ray emissions in bottom ash are predominantly due to the activity of ^{226}Ra and ^{232}Th activity.

Table 3: Estimated radiological hazards

Parameters	Ra_{eq} (Bq/kg)	DR (nGy/h)	AEDE (mSv/y)	AUI	H_{ex}	H_{in}	I_γ	ELCR (10^{-3})
Bottom Ash	258.42	113.60	0.14	1.53	0.70	0.94	1.79	0.48
Coal	60.55	26.75	0.03	0.37	0.16	0.22	0.42	0.11
Fly Ash	175.23	77.09	0.09	1.05	0.47	0.63	1.22	0.33
World Average	370	60	1 & 20	≤ 1	≤ 1	≤ 1	≤ 1	

Note: The ELCR world safety limit for low-level radiation is [0.05]

Table 4: Correlation relationship (95 % confidence level) of measured variables for coal samples from Van Eck power plant.

	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	DR	AEDE	AGDE	AUI	H_{ex}	H_{in}	I_γ	ELCR
^{226}Ra	1.00											
^{232}Th	0.89	1.00										
^{40}K	0.91	0.83	1.00									
Ra_{eq}	0.95	0.99	0.88	1.00								
DR	0.95	0.99	0.89	1.00	1.00							
AEDE	0.95	0.99	0.89	1.00	1.00	1.00						
AGDE	0.95	0.99	0.89	1.00	1.00	1.00	1.00					
AUI	0.91	1.00	0.86	0.99	0.99	0.99	0.99	1.00				
H_{ex}	0.95	0.99	0.89	1.00	1.00	1.00	1.00	0.99	1.00			
H_{in}	0.97	0.97	0.90	1.00	1.00	1.00	1.00	0.98	1.00	1.00		
I_γ	0.95	0.99	0.89	1.00	1.00	1.00	1.00	0.99	1.00	1.00	1.00	
ELCR	0.95	0.99	0.89	1.00	1.00	1.00	1.00	0.99	1.00	1.00	1.00	1.00

Table 5: Correlation relationship (95 % confidence level) of measured variables for fly ash samples from Van Eck power plant.

	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	DR	AEDE	AGDE	AUI	H_{ex}	H_{in}	I_γ	ELCR
^{226}Ra	1.00											
^{232}Th	0.61	1.00										
^{40}K	0.68	0.76	1.00									
Ra_{eq}	0.83	0.95	0.82	1.00								
DR	0.84	0.95	0.83	1.00	1.00							
AEDE	0.84	0.95	0.83	1.00	1.00	1.00						
AGDE	0.83	0.95	0.83	1.00	1.00	1.00	1.00					
AUI	0.66	1.00	0.80	0.97	0.96	0.96	0.97	1.00				
H_{ex}	0.83	0.95	0.82	1.00	1.00	1.00	1.00	0.97	1.00			
H_{in}	0.90	0.89	0.81	0.99	0.99	0.99	0.99	0.92	0.99	1.00		
I_γ	0.82	0.95	0.83	1.00	1.00	1.00	1.00	0.97	1.00	0.99	1.00	
ELCR	0.84	0.95	0.83	1.00	1.00	1.00	1.00	0.96	1.00	0.99	1.00	1.00

Table 6: Correlation relationships (95 % confidence level) of measured variables for bottom ash samples from Van Eck power plant.

	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	DR	AEDE	AGDE	AUI	H_{ex}	H_{in}	I_γ	ELCR
^{226}Ra	1.00											
^{232}Th	0.85	1.00										
^{40}K	0.26	0.30	1.00									
Ra_{eq}	0.93	0.98	0.33	1.00								
DR	0.94	0.98	0.34	1.00	1.00							
AEDE	0.94	0.98	0.34	1.00	1.00	1.00						
AGDE	0.94	0.98	0.34	1.00	1.00	1.00	1.00					
AUI	0.86	1.00	0.37	0.99	0.98	0.98	0.99	1.00				
H_{ex}	0.93	0.98	0.33	1.00	1.00	1.00	1.00	0.99	1.00			
H_{in}	0.96	0.96	0.32	1.00	1.00	1.00	1.00	0.97	1.00	1.00		
I_γ	0.93	0.98	0.34	1.00	1.00	1.00	1.00	0.99	1.00	0.99	1.00	
ELCR	0.94	0.98	0.34	1.00	1.00	1.00	1.00	0.98	1.00	1.00	1.00	1.00

3.4 Radionuclide Partitioning in Bottom (BA) and Fly ash (FA)

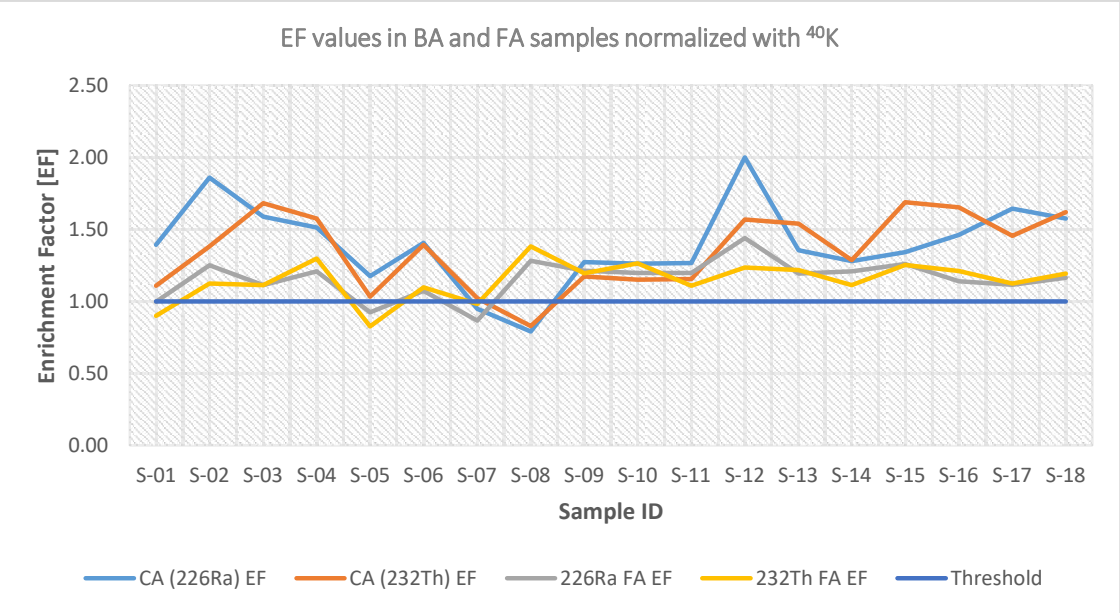
The degree of radionuclides partitioning between the bottom and fly ash due to the difference in physical and chemical characteristics and their association with the alumino-silicate in coal were determined by estimating the enrichment factor and activity concentration comparison of bottom ash and fly ash samples [24, 25].

3.4.1 Enrichment factor (EF)

The analysis showed an EF value > 1 which indicates enrichment in the ash's samples relative to feed coal. The EF values variations among the ash samples were considered the same with little differences. Generally, the EF in the FA samples is higher than the EF in BA samples however, in this study bottom ash EF values were moderately higher than those of Fly ash.

The results for ^{226}Ra in bottom ash samples further appear to indicate an increased activity concentration in the bottom ash rather than the finer fly ash as most literature suggests [25, 26] whose observation suggest that uranium and its progenies have the greatest small particle enrichment among elements that were neither lithophiles nor chalcophiles (elements associated with sulphide minerals).

Figure 3: Comparison of EFs of ^{226}Ra (^{238}U series) and ^{232}Th in the bottom and fly ash samples.

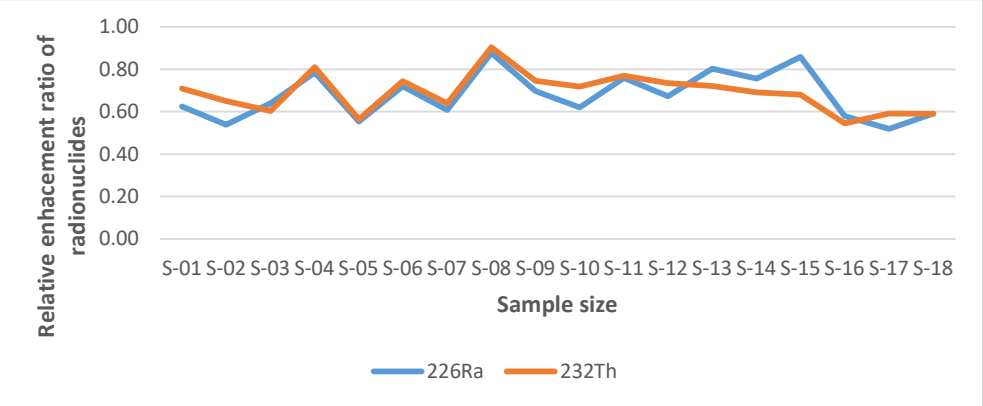


3.4.2 Relative Enhancement of Radionuclides Based on AC_{FA}/AC_{BA} Values

All the samples have activity concentration in fly ash relative to bottom ash of less than one for all the radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K with the mean values of 0.68, 0.69, and 0.80 respectively Figure 4. Based on the AC_{FA}/AC_{BA} values in Figure 4, ^{226}Ra and ^{232}Th were found to be evenly partitioned and distributed between bottom ash and fly ash although they differ in their volatility with ^{226}Ra reported to be more volatile than ^{232}Th.

The relative enhancement of ^{40}K is less than one in all the collected samples which confirms its behaviour classification as readily incorporated into bottom ash and are normally partitioned between the two common waste products of combustion [17]. However, according to [6] an increase in the furnace temperature of the power plant can decrease the enhancement in bottom ash resulting in an increased enhancement in the fly ash relative to the bottom ash of the plant.

Figure 4: Relative enhancement of radionuclide activity in fly ash samples.



4. Conclusion

This study evaluated the radiological hazards associated with the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in coal and coal combustion residues generated at Van Eck Coal-Fired power plant when used as secondary raw materials in construction industries. The results show that coal and ashes of Van Eck Power plant present high mean activity concentrations ^{226}Ra, ^{232}Th, and ^{40}K when compared to Morupule Power Station in Botswana and below that of Brazil, India, and Tanzania. However, the activity concentrations values obtained in this study, agree with other reported findings of reviewed literature in this work. As was expected, the activity of the three radionuclides was higher in the ashes samples than in the parent coal. This is because the activity concentrations accumulate when the coal is burned. The same observations were reported in coal and ashes samples reported elsewhere [9, 18, 19, 21, 22]. The statistical analysis demonstrates the presence of radiological hazards homogeneously distributed among the primordial radionuclides of ^{226}Ra, ^{232}Th, and ^{40}K in coal and fly ash, whereas for the bottom ash the radiological hazards are predominantly due to ^{226}Ra and ^{232}Th. Hence, continuous radiological screening and monitoring of the ashes utilized as a secondary raw material by other industries for any purpose whatsoever is recommended to keep the possible radiation hazards as low as reasonably achievable (ALARA). The study has provided the much-needed data on technologically enhanced naturally occurring radionuclides concentration in ashes fractions arising from Van Eck coal-fired power plant in Namibia. The results obtained in this study can serve as a baseline for further radiological studies of Van Eck Power Plant and as valuable data for the plant management and regulatory authority in waste management, drafting of policies, emission control regulations, and future developments.

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References

Zhang, X. (2014). Management of coal combustion wastes. *IEA Clean Coal Centre*, 2-68

Mamurekli, D. (2010). Environmental impacts of coal mining and coal utilization in the UK. *Acta Montanistica Slovaca*, *15*(2), 134.

Boukhair, A., Belahbib, L., Azkour, K., Nebdi, H., Benjelloun, M., & Nourreddine, A. (2016). Assessment of the radiological impact on the environment near a storage site of coal ashes in a thermal power plant. *World*, *6*, 206-216.

Kolo, M. T., Amin, Y. M., Khandaker, M. U., & Abdullah, W. H. B. (2017). Radionuclide concentrations and excess lifetime cancer risk due to gamma radioactivity in tailing enriched soil around Maiganga coal mine, Northeast Nigeria. *International Journal of Radiation Research*, *15*(1), 71.).

Omale, P. E., Ibrahim, B. M., Okeniyi, S. O., Ngokat, A. B., & Babarinde, A. O. (2017). Radiological assessment of coal samples from selected coal mines in Nigeria. *Chemistry Research Journal*, *2*(3), 22-28.

Lauer, N. E., Hower, J. C., Hsu-Kim, H., Taggart, R. K., & Vengosh, A. (2015). Naturally occurring radioactive materials in coals and coal combustion residuals in the United States. *Environmental science & technology*, *49*(18), 11227-11233.

Ozden, B., Guler, E., Vaasma, T., Horvath, M., Kiisk, M., & Kovacs, T. (2018). Enrichment of naturally occurring radionuclides and trace elements in Yatagan and Yenikoy coal-fired thermal power plants, Turkey. *Journal of environmental radioactivity*, *188*, 100-107

Gür, F., & Yaprak, G. (2010). Natural radionuclide emission from coal-fired power plants in the southwestern of Turkey and the population exposure to external radiation in their vicinity. *Journal of Environmental Science and Health Part A*, *45*(14), 1900-1908.

Mudiwa, J. (2015). *Dose assessment of natural radioactivity in fly ash and environmental materials from Morupule a coal-fired power station in Botswana* (Master's thesis, University of Ghana, graduate school of nuclear and allied sciences).

IAEA. (2011). *Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards* (General Safety Requirements Part 3 No. GSR Part 3 (Interim)). Vienna, Austria

Khandaker, M. U. (2011). High purity germanium detector in gamma-ray spectrometry. *International Journal of Fundamental Physical Sciences*, *1*(2), 42-46.

Knoll, G. F. (2010). *Radiation detection and measurement*. John Wiley & Sons.

Amin, Y. M., Khandaker, M. U., Shyen, A. K. S., Mahat, R. H., Nor, R. M., & Bradley, D. A. (2013). Radionuclide emissions from a coal-fired power plant. *Applied Radiation and Isotopes*, *80*, 109-116.

UNSCEAR (2008). *Sources and effects of ionizing radiation*, Report to the General Assembly with Scientific Annexes Vol. 1, United Nations, New York

UNSCEAR (2000). *Sources and effects of ionizing radiation*, Report to General Assembly, with Scientific Annexes, Vol. 1, United Nations, New York.

Dinis, M. L., Fiúza, A., Góis, J., Carvalho, J. M. S., & Castro, A. C. M. (2014). Modeling radionuclides dispersion and deposition downwind of a coal-fired power plant. *Procedia Earth and Planetary Science*, *8*, 59-63

Parami, V. K. (2008). *Determination of technologically enhanced naturally occurring radioactive material (TENORM) in ashes from coal-fired thermal power plants in the Philippines*.

Ademola, J. A., & Onyema, U. C. (2014). Assessment of natural radionuclides in fly ash produced at Orji River thermal Power Station, Nigeria, and the associated radiological impact. *Natural Science*, *6*(10), 752

Flues, M., Camargo, I.M.C., Silva, P. S.C., & Mazzilli, B.P. (2006). Radioactivity of coal and ashes from Figueira coal power plant in Brazil. *Journal of Radioanalytical and Nuclear Chemistry*, *270* (3), 597–602

Papastefanou, C. (2010). Escaping radioactivity from coal-fired power plants (CPPs) due to coal burning and the associated hazards: a review. *Journal of Environmental Radioactivity*, *101*(3), 191-200.

Mondal, T., Sengupta, D., & Mandal, A. (2006). Natural radioactivity of ash and coal in major thermal power plants of West Bengal, India. *Current Science*, 1387-1393.

Makundi, I., Nyaki, M., & Mohammed, N. (2018). Assessment of radioactivity levels in coal and coal ash in Kiwira coal mine using gamma-ray spectrometry. *Tanzania Journal of Science*, *44*(3), 1-11.

Izquierdo, M., & Querol, X. (2012). Leaching behaviour of elements from coal combustion fly ash: an overview. *International Journal of Coal Geology*, *94*, 54-66.

Sahu, S. K., Tiwari, M., Bhangare, R. C., Ajmal, P. Y., & Pandit, G. G. (2017). Partitioning behaviour of natural radionuclides during combustion of coal in thermal power plants. *Environmental Forensics*, *18*(1), 36-43.

Flues, M., Sato, I. M., Scapin, M. A., Cotrim, M. E. B., & Camargo, I. M. C. (2013). Toxic elements mobility in coal and ashes of Figueira coal power plant, Brazil. *Fuel*, *103*, 430-436.

Çam, N. F., Yaprak, G., & Eren, E. (2010). The natural radioactivity contents in feed coals from the lignite-fired power plants in western Anatolia, Turkey. *Radiation protection dosimetry*, *142*(2-4), 300-307

Ahmaruzzaman, M. (2010). A review on the utilization of fly ash. *Progress in energy and combustion*, 327-363