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Quantification of radionuclides and associated radiological risk estimation of coal combustion residues from a South African coal-fired power plant

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► ***Original article***

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ABSTRACT

Background: Radionuclides occur in coal combustion residues, such as fly ash and bottom ash, which are by-products of coal combustion. They pose potential radiological risks to people present in the surrounding areas. ***Materials and Methods:*** Gamma spectrometry was performed to determine the radionuclide activity concentrations in a coal-driven power plant located in the Limpopo province, South Africa, to assess the radiological impacts of the ash stored in ash dumps adjacent to the plant. ***Results:*** The mean (± SD) activity concentrations were found to be 144.3±4, 62±2.1, and 315.9±4.9 Bq/kg for ^{226}Ra, ^{232}Th, and ^{40}K, respectively, which are comparable to those found in previous studies. The radium equivalent activity was determined to be 258.43 Bq/kg. The average values of internal and external hazard indices were 1.09 and 0.70, respectively. With the exception of the internal hazard index, all the other indices were within the prescribed ranges indicated by the literature. Furthermore, the mean total annual effective dose received by plant workers was found to be 0.070 mSv/y, which is within the limit of 1.0 mSv/y prescribed by the IAEA. The average excess lifetime cancer risk value was 0.49×10^{-3}, which is higher than the UNSCEAR precautionary limit of 0.29×10^{-3} but lower than the ICRP limit of 0.05 for low-level radiation. ***Conclusion:*** Ash dust inhalation was identified as the most significant exposure pathway among plant workers. However, the results demonstrated that storing of ash at this plant does not constitute any radiological threat to people in the adjacent regions.

Keywords: *Radionuclides, flyash, hazard index, excess lifetime cancer risk (ELCR)*

INTRODUCTION

According to the International Energy Outlook (IEO) [1], about 40% of the global energy demand is met by coal-fired power plants (CFPP). Coal is composed of inorganic constituents that include naturally occurring radioactive materials (NORM); NORM are enriched in coal combustion residues, such as fly ash and bottom ash, which are the by-products of coal combustion [2]. The International Atomic Energy Agency (IAEA) [3] refers to the NORM-type industries as those whose operations could incrementally expose the population and employees within their environment to ionizing radiation. The radioactive content of some of these fuels undergo volatilization during combustion, and are emitted into the atmosphere, whereas the contents whose melting points are higher than the combustion temperature are concentrated in the resulting waste that comprises bottom ash and fly ash [4]. More than 50 000 tonnes of coal is consumed daily by a modern CFPP in South Africa; consequently, more than 17 000 tonnes of ash is produced daily, depending on the content of ash and heat, and the quality of coal [5].

Ash is a by-product obtained from the combustion of coal, and is commonly used in cement manufacturing [6]. The huge quantities of ash produced by CFPPs are stored in ponds in heaps

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adjacent to the power plants [4, 7, 8]. Thus, extensive research has been conducted to evaluate the radiological impact of both the radionuclides emitted into the atmosphere and those stored in massive ash deposits [4, 9-11].

The interest in measuring NORM concentrations in coal and the resulting combustion residues arises out of the awareness of health hazards and environmental pollution [12]. The residues produced by CFPP are notable sources of exposure to plant workers and the population near the plants to naturally occurring radionuclides [13, 14]. Naturally occurring radionuclides, specifically ^{40}K, ^{226}Ra, and ^{232}Th released by these plants, pose potential health hazards [15]. Therefore, the risks emanating from coal combustion residues should be evaluated to determine the radiological impact of such residue deposits, and devise functional methodologies and a practical framework for administering control doses to the public and the employees.

From a global perspective, the area of radionuclides in coal and the resulting combustion residues have been significantly studied [2, 11, 13, 14, 16-18]. However, estimation and quantification of the radiological risks posed by radionuclides present in coal combustion residues (especially around CFPP ash dumps) remain to be presented in the public domain. Accordingly, this study is focused on quantifying the radionuclides in the ash dumps surrounding a typical CFPP in the Limpopo Province, South Africa, and further estimating the radiological risk associated with them.

MATERIALS AND METHODS

Sample collection

Thirty-three coal samples (1 kg each) were collected from the ash dumps (containing both fly ash and bottom ash) of a CFPP located in the Limpopo Province, South Africa. Each sample was collected at a distance of 20 m from the other at a depth of 30 cm from each heap sampled. This process ensured satisfactory representation of the ash dumps by the samples. The bituminous coal used in this power plant is a blend of different qualities from a single mine that processes up to six coal zones. Typically, the middling produced from an advanced coal beneficiation plant are supplied to the power station. The samples were packed and sealed in polyethylene bags, carefully labelled, and subsequently transported to an independent ISO certified laboratory in Pretoria, South Africa.

Sample processing and analyses

Gamma spectrometry was performed to determine the radionuclides of interest (^{226}Ra, ^{232}Th, and ^{40}K) in the samples. The ash samples were dried for 24 h in an air-circulation oven at 110 °C. The samples were further pulverized to obtain a fine powder and were sieved for homogeneity. Then, 100 g of each sample was placed in plastic containers of 6.5 cm (diameter) × 7.5 cm (height), which were sealed to become airtight. The samples were left in this state for a month in a designated laboratory cupboard to ascertain secular equilibrium between ^{226}Ra and ^{238}U with their progeny, and prevent Rn loss. A high-resolution, p-type coaxial HPGe γ-ray spectrometer (Canberra, USA) protected with cylindrical lead was used to determine the specific radionuclides of the samples, i.e. ^{232}Th, ^{226}Ra, and ^{40}K. At 1.33 MeV Co^{60} peak, the energy resolution of the detector was 1.67 keV at full width at half maximum (FWHM), and its relative efficiency was 28.2%. The detector was coupled to a 16 k MCA to determine the photo-peak area of the γ-ray spectrum, which was then analysed using the Genie 2K software (Canberra, USA). A cylindrical multi-nuclide source was used for detector energy calibration and efficiency determination [19]. The measured detection efficiencies were fitted using a polynomial fitting function described by Khandaker *et al.* [20], and the fitted efficiencies were used in activity determination of the samples. The minimum detectable activity (MDA) of the γ-ray measurement system at 95% confidence level was calculated based on the procedure devised by Khandaker *et al.* [20]. Each sample was counted for 24 h, and similarly for background counts, to obtain the net activity. All the experiments were conducted in triplicate.

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were estimated using the certified reference material IAEA-447 obtained from IAEA, Vienna, for quality assurance in this study. The measured values were in good agreement with the certified values; the mean measured and certified values were respectively 24.2±2.8

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Bq/kg and 25.1±2.0 Bq/kg for ^{226}Ra, 36.2±1.8 Bq/kg and 37.3±2.0 Bq/kg for ^{232}Th, and 535±30 Bq/kg and 550±20 Bq/kg for ^{40}K.

Radiological hazard assessment

The following parameters were evaluated using the activity concentrations of the radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) quantified by gamma spectrometry.

Radium equivalent activity (Ra_{eq})

In most naturally occurring radioactive materials, the radionuclides ^{226}Ra, ^{232}Th, and ^{40}K are not in secular equilibrium; therefore, the parameter Ra_{eq} is demarcated in terms of exposure to radiation. The radium equivalent accounts for the effective dosage from Rn and its decay products [21]. Furthermore, it is measured in Bq/kg, and its definition is primarily based on the assumption that the specific activity of 370 Bq/kg of ^{226}Ra, which is uniformly distributed in any naturally occurring sample, can produce an annual effective dosage of 1 mSv at 1 m above the ground level [22]. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [23] defines Ra_{eq} quantitatively by the use of equation 1:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (1)$$

Where A_{Ra}, A_{Th}, and A_K represent the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The constants in Eq. 1 represent the corresponding activity conversion rates of ^{226}Ra, ^{232}Th, and ^{40}K, which result in the same gamma dose rate at a maximum permissible Ra_{eq} of 370 Bq/kg.

External hazard index

The external hazard index (H_{ex}) is used for quantifying the gamma ray-acquired radiation hazards. The maximum value of 1, corresponding to the upper limit of radium equivalent at 370 Bq/kg, constitutes the optimum acceptable value for external hazard index [21, 24]. Equation 2 is used for computing H_{ex}:

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (2)$$

Where A_{Ra}, A_{Th}, and A_K represent the activity concentrations of the respective radionuclides, i.e. ^{226}Ra, ^{232}Th, and ^{40}K. It is assumed that the same rate of gamma dose can be obtained from 4810 Bq/kg of ^{40}K, 259 Bq/kg of ^{232}Th, and 370 Bq/kg of ^{226}Ra [25-27].

Internal hazard index

Radon and its carcinogenic decay products are hazardous to respiratory organs [27-29]. The internal exposure to radon and its decay progenies is quantified by the internal hazard index H_{in}, which is given by equation 3 (from [23]):

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \quad (3)$$

Where A_{Ra}, A_{Th}, and A_K represent the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The values of both H_{ex} and H_{in} must be less than one for radiation hazards to be negligible [23].

Representative gamma index ($I_{\gamma r}$)

The representative gamma index, $I_{\gamma r}$, is a common parameter used for screening materials that present potential health issues because of radiation [30]. The Nuclear Energy Agency (NEA) proposed equation 4 for computing $I_{\gamma r}$:

$$I_{\gamma r} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \quad (4)$$

Where A_{Ra}, A_{Th}, and A_K denote the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively; $I_{\gamma r}$ is measured in Bq/kg. The specific rates of exposure for ^{40}K, ^{232}Th, and ^{226}Ra are denoted by the denominators 1500, 100, and 150, respectively. The European commission indicates that for the materials used in large quantities, the exemption dose criterion (0.3 mSv/y) corresponds to $I_{\gamma r} < 0.5$, whereas the dose criterion 1 mSv/y corresponds to $I_{\gamma r} \leq 1$ [32]. On the other hand, for superficial and other materials, the corresponding values of $I_{\gamma r}$ should be between 2 and 6 [29].

Excess lifetime cancer risk

The possibility of contracting cancer by individuals surrounded by coal combustion

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products can be evaluated using the excess lifetime cancer risk (ELCR) parameter, even in the absence of outbreak of radioactive components. In this study, the ELCR was estimated using equation 5, as described by Taskin *et al.* [33] and Ravisankar *et al.* [34]:

$$ELCR = AEDE \times DL \times RF \quad (5)$$

Where AEDE (mSv/y), DL, and RF represent the annual effective dose equivalent, duration of life (70 years), and risk factor (Sv^{-1}) (fatal cancer risk per sievert), respectively. For stochastic effect, the International Commission on Radiological Protection (ICRP) [35] uses the values 0.05 for the public.

The value of AEDE is proposed to be calculated using equation 6, as described by Ravisankar *et al.* [34]:

$$AEDE = ADRA \times DCF \times OF \times T \quad (6)$$

where ADRA represents the absorbed dose rate in air (nGy/h) at 1 m above the ground level, and is based on the radioactivity of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs in the sample; further, DCF, OF, and T represent the dose conversion factor (0.7 Sv/Gy), outdoor occupancy (0.2), and time (8760 h/y), respectively. The ADRA was calculated using Eq. 7 [23]:

$$ADRA=0.461A_{Ra}+0.623A_{Th}+0.041A_{K}+0.1243A_{Cs} \quad (7)$$

Where A_{Ra}, A_{Th}, and A_K denote the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, respectively. Preliminary analyses revealed that Cs^{137} was not detected in any sample, and the concentrations are thus not presented in the results. This is rational because Cs^{137} spreads in the atmosphere via nuclear activity, and most of the fallout radiation accumulates in the soil [23].

Occupational risk estimation

Large volumes of coal combustion products (fly ash and bottom ash) consisting of fine particles, which are relatively loose and non-compacted, are continuously dumped from the conveyor belts in the ash dumps surrounding the power plant. Consequently, workers are (potentially) constantly exposed to high doses of radiation via three major pathways:

1. Gamma radiation from the ash dumps (external exposure)
2. Inhalation of ash from the ash dumps (internal exposure)
3. Accidental ingestion of ash from the dumps (internal exposure)

The radiation dose, which could be received via any of the exposure pathways described above, can be calculated by applying the dose conversion coefficients prescribed by the ICRP, which are discussed in the forthcoming sections. They cumulatively present the total effective dose of radionuclides and any health hazard posed because the radiation exposure depends on them.

External radiation

Equation 8, as described by several authors, is used to estimate the external exposure to gamma radiation [36, 37]:

$$D_{ext} = \Sigma A_i C_{ext,i} T_e \quad (8)$$

Where A_i denotes the activity concentration of a specific radionuclide *i* (measured in Bq/kg), and $C_{ext,i}$ represents the effective dose coefficient related to a specific nuclide *i* in the contaminated surface (measured in Svh^{-1}/Bqg^{-1}). In this study, the values 9.929, 0.003, and 1.175 $nSv/Bqkg^{-1}$ were used for ^{226}Ra, ^{232}Th, and ^{40}K, respectively [36]. T_e denotes the duration of exposure in years (2000 years) [36].

Inhalation dose

Equation 9 is used to calculate the internal exposure due to inhalation of ash [36]:

$$D_{inh} = \Sigma A_i C_{inh,i} \eta_{inh} D_f T_e \quad (9)$$

Where $C_{inh,}$ denotes the dose coefficient for inhalation of a specific radionuclide *i* (measured in Sv/Bq) (the values 2.2×10^{-6}, 2.9×10^{-5}, and 3×10^{-9} Sv/Bq were used for ^{226}Ra, ^{232}Th, and ^{40}K, respectively, as described in ICRP 119 [38]);

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η_{inh} denotes the breathing rate, which is measured in m^3/h (1×10^{-3}) [36]; D_f is the dust loading factor (1×10^{-3}) [39]; A_i and T_e are the same as those defined in equation 8.

Ingestion dose

The internal dose received by accidental ingestion of radionuclides is estimated using equation 10.

$$D_{inh} = \Sigma A_i C_{ing,i} \eta_{ing} T_e \quad (10)$$

where $C_{ing,}$ denotes the dose coefficient for ingestion of a specific radionuclide *i* (measured in Sv/Bq) (the values 28×10^{-7}, 2.2×10^{-7}, and 6.2×10^{-9} Sv/Bq were used for ^{226}Ra, ^{232}Th, and ^{40}K, respectively, as described in ICRP 119 [38]); η_{ing}, which is measured in kg/h, denotes the ingestion rate typical for adults, i.e. 5×10^{-6} kg/h [36]; A_i and T_e are the same as those defined in equation 8.

Statistical analysis

Descriptive statistical tools, such as minimum, maximum, mean, and standard deviations, were calculated for the obtained data. The interdependency of the radiological variables measured in this study was evaluated using the Pearson's correlation matrix with an alpha-testing level of $P<0.05$ for the 33 ash samples. The matrix (table 2) was produced using the statistical program for social science (SPSS 25.0.0.0).

RESULTS

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K and their corresponding uncertainty levels of ±σ, involving the minimum, maximum, mean, and standard deviations of the samples, are presented in table 1. The range of activities obtained for the ash samples are between 99±5–183±5, 40±2–71±3, and 229±8–388±8 Bq/kg for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The mean values (±SD) were 144.3±4.0, 62±2.1, and 315.9±4.9 Bq/kg for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. In addition, the radium equivalent and radiological hazard indices are presented using the activity concentrations and equations 1–4. The mean (± SD) Ra_{eq} for the samples was 258.43 Bq/kg and the corresponding hazard indices for the H_{ex}, H_{in} and $I_{\gamma r}$ were 0.70, 1.09 and 1.80 respectively. The possibility of contracting cancer by individuals surrounded by coal combustion products was evaluated using the ELCR parameter which was calculated by combined usage of equations 5–7. and was found to be 0.49×10^{-3}.

The interdependency of the radiological variables measured in this study is presented in table 2 which was produced using the statistical program for social science (SPSS 25.0.0.0) with an alpha-testing level of $P < 0.05$ for the ash samples. The degree of association existing between the radionuclides and the radiological hazards are presented in terms of their correlation coefficients (r).

The radiation doses received by those exposed to ^{226}Ra, ^{232}Th, and ^{40}K via the three exposure pathways (external, inhalation, and ingestion) were calculated using equation 8–10. The results of the calculated radiation doses and the total effective dosage are presented in table 3 which illustrates that the calculated effective dosage due to external exposure to the ash dumps varies between 2.60–4.46 μSv/y. The effective dose delivered by inhalation to those exposed to the ash was in the range 46.68–83.24 μSv/y, whereas that from incidental ingestion of radionuclides had a mean value of 0.06 μSv/y. The total effective dosage received via all three pathways was 0.050–0.088 mSv/y.

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Table 1. Activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, their corresponding radiation hazard indices, and the ELCR of coal combustion products (combination of fly ash and bottom ash) from the ash dumps around the CFPP in Limpopo Province, South Africa.

SAMPLE	Activity Concentration(Bq/kg)				Hazard Indices			
	^{226}Ra	^{232}Th	^{40}K	Ra_{eq}	H_{ex}	H_{in}	I_{yr}	ELCR ($\times10^{-3}$)
1	166±5	68±3	319±3	287.80	0.78	1.23	2.00	0.57
2	165±2	68±2	329±3	284.71	0.77	1.22	1.98	0.57
3	180±3	70±2	337±3	300.33	0.81	1.30	2.08	0.60
4	101±5	43±3	312±5	222.26	0.60	0.87	1.56	0.37
5	150±3	60±1	259±8	248.59	0.67	1.08	1.72	0.50
6	155±2	62±2	292±5	257.56	0.70	1.11	1.79	0.52
7	182±2	71±3	287±5	268.45	0.73	1.22	1.85	0.60
8	154±5	61±2	315±4	255.48	0.69	1.11	1.78	0.52
9	142±2	53±2	330±5	223.18	0.60	0.99	1.56	0.48
10	111±4	47±3	295±8	232.39	0.63	0.93	1.63	0.40
11	140±5	52±3	333±7	264.31	0.71	1.09	1.85	0.48
12	168±3	68±2	311±5	297.77	0.80	1.26	2.07	0.57
13	99±5	41±1	270±3	198.44	0.54	0.80	1.39	0.35
14	130±4	46±2	312±3	254.12	0.69	1.04	1.77	0.44
15	172±4	64±2	318±6	290.87	0.79	1.25	2.02	0.57
16	116±5	48±3	297±4	223.24	0.60	0.92	1.56	0.41
17	115±3	47±3	337±6	242.48	0.66	0.97	1.70	0.41
18	111±3	44±2	289±4	219.05	0.59	0.89	1.53	0.39
19	183±2	71±1	311±3	301.33	0.81	1.31	2.09	0.61
20	131±2	50±2	292±6	232.13	0.63	0.98	1.62	0.45
21	134±5	48±2	324±5	253.33	0.68	1.05	1.77	0.45
22	112±5	47±1	342±3	215.55	0.58	0.89	1.51	0.41
23	177±2	69±3	388±8	288.39	0.78	1.26	2.01	0.60
24	171±4	68±2	287±7	277.47	0.75	1.21	1.92	0.57
25	183±5	71±3	352±8	305.91	0.83	1.32	2.12	0.61
26	132±4	49±3	294±6	239.01	0.65	1.00	1.67	0.44
27	145±5	58±1	341±3	271.36	0.73	1.12	1.89	0.50
28	155±2	62±2	319±5	291.10	0.79	1.21	2.03	0.53
29	165±4	67±3	300±6	266.75	0.72	1.17	1.85	0.56
30	144±4	53±2	229±8	254.58	0.69	1.08	1.76	0.47
31	100±3	40±2	367±4	228.36	0.62	0.89	1.61	0.37
32	131±5	49±2	387±3	262.33	0.71	1.06	1.84	0.46
33	144±5	53±1	350±5	269.62	0.73	1.12	1.88	0.49
Min	99±5	40±2	229±8	198.44	0.54	0.80	1.39	0.35
Max	183±5	71±3	388±8	305.91	0.83	1.32	2.12	0.61
Mean ± SD	144.3±4	62±2.1	315.9±4.9	258.43	0.70	1.09	1.80	0.49

Table 2. Correlation matrix of the radiological variables for coal combustion products (fly and bottom ashes) from the ash dumps around the CFPP in Limpopo Province, South Africa.

Correlations								
Variables	**^{226}Ra**	**^{232}Th**	**^{40}K**	**Ra_{eq}**	**H_{ex}**	**H_{in}**	**I_{yr}**	**ELCR**
^{226}Ra	1.00							
^{232}Th	0.97	1.00						
^{40}K	0.07	0.05	1.00					
Ra_{eq}	0.98	0.98	0.13	1.00				
H_{ex}	0.99	0.98	0.13	0.99	1.000			
H_{in}	0.99	0.98	0.10	0.98	0.99	1.00		
I_{yr}	0.98	0.98	0.14	0.98	0.98	0.99	1.00	
ELCR	0.99	0.98	0.14	0.99	0.98	0.99	0.98	1.000

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Table 3. Calculated effective dose received by workers by radiation from the surrounding ash dumps in the CFPP in the Limpopo Province, South Africa.

SAMPLE	Effective dose (μSv/y)			Total effective dose (mSv/y)
	D_{ext}	D_{inh}	D_{ing}	
1	4.05	79.03	0.06	0.083
2	4.05	78.96	0.06	0.083
3	4.37	82.03	0.06	0.086
4	2.74	49.69	0.04	0.052
5	3.59	69.99	0.06	0.074
6	3.76	72.33	0.06	0.076
7	4.29	83.16	0.06	0.087
8	3.80	71.28	0.06	0.075
9	3.60	62.54	0.06	0.066
10	2.90	54.35	0.04	0.057
11	3.56	61.41	0.06	0.065
12	4.07	79.18	0.06	0.083
13	2.60	47.58	0.04	0.050
14	3.32	54.79	0.04	0.058
15	4.16	75.55	0.06	0.080
16	3.00	55.71	0.04	0.059
17	3.08	54.65	0.04	0.058
18	2.88	51.41	0.04	0.054
19	4.37	83.23	0.06	0.088
20	3.29	58.78	0.04	0.062
21	3.42	57.05	0.06	0.060
22	3.03	54.43	0.04	0.057
23	4.43	80.83	0.06	0.085
24	4.07	79.40	0.06	0.084
25	4.46	83.24	0.06	0.088
26	3.31	57.88	0.04	0.061
27	3.68	67.67	0.06	0.071
28	3.83	72.33	0.06	0.076
29	3.98	77.97	0.06	0.082
30	3.40	62.68	0.06	0.066
31	2.85	46.68	0.04	0.050
32	3.51	57.81	0.04	0.061
33	3.68	62.69	0.06	0.066
Min	**2.60**	**46.68**	**0.04**	**0.050**
Max	**4.46**	**83.24**	**0.06**	**0.088**
Mean	**3.61**	**66.25**	**0.06**	**0.070**

DISCUSSION

The range of activities obtained for the ash samples indicated in table 1 showed mean values (± SD) of 144.3±4.0, 62±2.1, and 315.9±4.9 Bq/kg for ^{226}Ra, ^{232}Th, and ^{40}K, respectively. The values of the measured concentrations of ^{226}Ra, ^{232}Th, and ^{40}K were similar to those measured for other CFPPs in South Africa that use coals from other local mines [11]. In addition, the values are comparable to those reported by Mora *et al.* [40] and Baeza *et al.* [4]. Furthermore, the average activity levels defined in the UNSCEAR report [41] for fly ash are 240, 70, and 265 Bq/kg for ^{226}Ra, ^{232}Th, and ^{40}K, respectively, which are similar to the average values obtained for the ash dumps in this study.

The calculated values for the Ra_{eq} activity index were in the range 198.44–305.91 Bq/kg, with an average value of 258.43 Bq/kg; H_{ex} had an average of 0.70. The Ra_{eq} and H_{ex} values were lower than the corresponding values of 370 Bq/kg and 1, which are the values recommended by UNSCEAR [23]. The degree of internal exposure of Rn and its decay products is quantified by the internal hazard index (H_{in}), and was recorded as 1.09 on average—this is slightly higher than the limit prescribed by UNSCEAR [23], which is less than 1. The gamma index ($I_{\gamma r}$), recorded as (average) 1.80, and should be between 2 and 6 for materials such as coal ash, according to Hilal *et al.* [29]. The calculated ELCR for the ash dumps in the Limpopo CFPP varied between 0.35×10^{-3} and 0.61×10^{-3} with an average value of 0.49×10^{-3}. The average ELCR value was thus found to be higher than the UNSCEAR [23] precautionary limit of 0.29×10^{-3}, but lower than the ICRP [35] limit of 0.05 prescribed for low-level radiation. As mentioned earlier, the radiation hazard indices in table 1 are calculated based on the radionuclide concentrations that are similar to those of the ashes from other local CFPPs, as reported by Ahmed and Joubert [11]. It could, therefore, be stated that other CFPPs in South Africa are expected to have similar Ra_{eq}, H_{ex}, H_{in}, $I_{\gamma r}$, and ELCR values.

Table 2 indicates that a very strong degree of association exists between ^{226}Ra and ^{232}Th ($r = 0.97$), which could be attributed to the fact that Ra and Th are both heavy radionuclides that exist together in nature [36]. On the contrary, very weak correlations exist between ^{40}K, ^{226}Ra ($r = 0.07$), and ^{232}Th ($r = 0.05$). Furthermore, very strong positive correlations were found not only between all the estimated radioactive

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variables (Ra_{eq}, H_{ex}, H_{in}, I_{yr}, and ELCR) but also between ^{226}Ra and ^{232}Th ($r \geq 0.98$). This indicates that gamma radiation is predominantly radiated by ^{226}Ra and ^{232}Th in the ash dumps.

The total effective dosage received via all three pathways (external exposure, inhalation and ingestion) as demonstrated in table 3 was 0.050–0.088 mSv/y. These results indicate that the most significant occupational exposure pathway is the inhalation of ash, which accounts for approximately 94% of the total average annual effective dose. It could be concluded, with regards to the occupational exposure, that the total annual effective dose from the ash dumps is less than the precautionary limit set by the IAEA [42], which is 1 mSv/y. Therefore, storing of ash at the Limpopo CFPP does not pose any harmful radiological threat to the people in adjacent areas.

CONCLUSIONS

The mean activities for ^{226}Ra, ^{232}Th, and ^{40}K in the ash samples analysed were 144.3±4.0, 62±2.1, and 315.9±4.9 Bq/kg, respectively. The H_{ex} value was within the precautionary limits set by UNSCEAR, whereas H_{in} was slightly higher than UNSCEAR limit of 1. The average ELCR value was found to be 0.49×10^{-3}, which is higher than the UNSCEAR precautionary limit of 0.29×10^{-3}. Furthermore, the mean total annual effective dose of 0.070 mSv/y received by the plant workers was found to be below the safety criterion of 1.0 mSv/y set by the IAEA.

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