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Natural radioactivity analysis and radiological impact assessment from a coal power plant

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ABSTRACT

Background: Through combustion, a coal-fired power plant produces by-products, such as fly ash and bottom ash, which contain significant concentrations of radionuclides that cause environmental contamination, leading to health problems. ***Materials and Methods:*** This study investigated the specific activity and radiological impacts of naturally occurring radioactive materials (NORMs) in the coal, fly ash, and bottom ash at a coal-fired power plant. Samples were collected from a coal-fired power plant in Malaysia. ***Results:*** Gamma spectrometry was used to determine the specific concentrations of NORMs, namely ^{40}K, ^{232}Th, ^{238}U, in each sample. The radioactivity ranges for ^{238}U, ^{232}Th, and ^{40}K in the soil were 22.7–150.7 Bq/kg, 20.7–153.6 Bq/kg, and 68.6–1594.4 Bq/kg, respectively. The coal, fly, and bottom ash samples contained 67.54–189.18 Bq/kg of ^{238}U, 50.2–134.57 Bq/kg of ^{232}Th, and 327.54–1114.40 Bq/kg of ^{40}K. The radium equivalent activities (Ra_{eq}) in the samples were 164.55, 467.42, and 429.09 Bq/kg, respectively. Meanwhile, the absorbed dose rate (ADR) in the air ranged from 76.04 to 217.44 nGy/h. Internal and external hazards ranged from 0.44 to 1.26 and 0.63 to 1.77, respectively. The annual gonadal dose equivalent (AGDE) value fluctuated between 521.28 and 1,496.99 µSv. ***Conclusion:*** The excess lifetime cancer risk (ELCR) oscillated from 1.30×10^{-3} to 3.75×10^{-3} indoors and 0.32×10^{-3} to 0.95×10^{-3} outdoors.

INTRODUCTION

Coal is mainly used in combustion to produce electricity in coal-fired power plants by heating water into steam, and the steam drives turbine generators to generate electricity. As of 2007, 50,000 coal-fired power plants remained active, and the total number is expected to continue increasing globally. The electricity generated by coal-fired plants was 21.6 trillion kilowatt-hours (T-kWh) in 2012 and 25.8 T-kWh in 2020. This number is expected to grow higher, reaching approximately 36.5 T-kWh in 2040 (1). Coal-fired power generation represents a major source of electricity in many countries, accounting for around 45% of total power generation in the US, 40% in Germany, and 25% in Japan. In China, the share is around 80%, while India and Serbia get around 70% of their power from coal (2).

Unfortunately, the combustion of coal produces carbon monoxide (CO) as its main product, followed by nitrogen dioxide (NO_2) and sulfur dioxide (SO_2). Therefore, the large amount of CO_2 emitted from coal-fired power plants constitutes one of the largest causes of global warming, contributing significantly to climate change.

The combustion of coal involves high temperatures and potentially poses risks to environmental and human health (3). For example, coal combustion discharges gaseous particulates and by-products known as bottom ash and fly ash. These by-products contain radionuclides that are regularly released into the atmosphere (4). Coal, fly ash, and bottom ash contain trace impurities of the higher radionuclides uranium-238 (^{238}U), thorium-232 (^{232}Th), and potassium-40 (^{40}K). Since these naturally occurring radioactive materials (NORMs) have exceptionally long half-lives (up to 1000 years), their existence can simply be considered permanent. As a result, the amounts of radioactivity in the world's land, water, and air vary by location (5).

In this study, the selected coal power plant is one of Malaysia's largest thermal power plants. It generates 3,100 MW of electricity per year, making it Peninsular Malaysia's largest power producer to date. The plant will remain the country's largest electric power producer into the foreseeable future, representing 30% of the power generation capacity in Peninsular Malaysia [6].

The power plant uses coal as a source to fuel its furnace and consumes up to 15 million tons (Mt) of coal per year, an amount which is increasing more rapidly than other energy resources [7]. Elements such as carbon and silicon are redistributed into bottom ash, electrostatic precipitator fly ash, and stack-emitted materials during coal or solid waste burning. One ton of ash is produced for every four tons of coal burned, and this amount of ash can spread over 150,000 square kilometers (approximately 60,000 square miles). Fly ash can travel up to 40 to 50 km in the direction of the wind. When it settles, it can cause land degradation and severe air and water pollution [8].

Fly ash contains radionuclides, such as ^{238}U, ^{232}Th, and ^{40}K, which could fly a few kilometers. Within a 20 -kilometer radius of a coal-fired power plant station, the concentrations of ^{238}U, ^{232}Th, and ^{40}K in the upper layer of soil grow approximately 0.03% to 0.12% annually, similar to typical natural concentrations in soil [1]. Harmful substances have been discovered in cow's milk in such coverage areas. The inhalation of this ash by workers and the public in the surrounding area and its deposition in the soil can present a potential health risk to the population through its emanation of radon [9].

When people are exposed to specific levels of ^{238}U, ^{232}Th, and ^{40}K for an extended period of time, they risk developing cavity and bone cancer [10]. In addition, the ash may irritate or block the delicate and fine airways of the lungs, thereby causing asthma and chronic bronchitis. Furthermore, when radium, which is the progeny of ^{238}U, enters the body through ingestion and inhalation, it deposits in the bone because its metabolic behavior is similar to calcium faction. This deposition may result in teeth fracture, anemia, and cataracts and may even cause various types of cancer. Meanwhile, thorium exposure can cause lung, pancreatic, hepatic, bone, and kidney cancers as well as leukemia [11, 12].

This is the first study aimed at measuring the specific activity concentration of naturally radioactive materials (NORMs) in coal, bottom ash, and fly ash at one of Malaysia's coal power plants, which comprises five generating units, one of which uses ultra-supercritical technology. From the specific radionuclide activity, this study determined the radium equivalent activity (Ra_{eq}), absorbed dose rate (ADR) in the air, internal and external hazards, annual gonadal dose equivalent (AGDE), total effective dose, representative gamma index (IYr), and excess lifetime cancer risk (ELCR).

MATERIALS AND METHODS

Sampling and sample preparation

Soils, sediment, and water were collected within a 4–5 km radius of the coal power plant, as shown in figure 1. Table 1 lists the coordinates of each sample. Meanwhile, the coal, fly ash, and bottom ash samples were taken from one of the coal power plant's combustion units. For direct gamma spectrometry of Canberra hyper pure germanium (HPGe), all the samples taken were dried in an air circulation oven (Berkeley BSK-1700X-M) at 70ºC for 48 hours. The samples were then ground into a fine powder 500 µm in size, placed in a polyethylene bottle and sealed hermetically. Due to the possibility of ^{222}Rn escape during handling, the sample was held for at least 30 days for homogenization before counting to re-establish the secular radioactive equilibrium between ^{226}Ra and its short-lived daughter products. This method was adapted from the technical IAEA document, the same standard used in reference [13].

Figure 1. Location of sampling of soil, sediment, and water.

Table 1. Coordinates of soil (T), water (A), and sediment (S) samples were recorded surrounding the coal power plant. These coordinates were recorded using GPS and samples for each of the coordinates were collected.

Sample	Coordinate		Sample	Coordinate	
	Latitude	Longitude		Latitude	Longitude
T1	4.17495	100.66750	T16	4.18458	100.65775
T2	4.17439	100.66442	T17	4.18301	100.65618
T3	4.17527	100.66209	T18	4.18159	100.65478
T4	4.17628	100.65723	T19	4.18711	100.6496
T5	4.17691	100.65311	S1	4.1832	100.64575
T6	4.17128	100.63893	S2	4.17705	100.653
T7	4.17337	100.63633	S3	4.17697	100.65149
T9	4.1782	100.64099	S4	4.16932	100.6404
T10	4.17787	100.64528	A1	4.18313	100.64562
T11	4.17726	100.65032	A2	4.18469	100.65796
T12	4.18291	100.64543	A3	4.17699	100.65149
T13	4.18652	100.63750	A4	4.17705	100.653
T14	4.18086	100.63775	A5	4.16933	100.64043
T15	4.18494	100.63853			

Calculation of specific radionuclide activities

The specific activities of natural radionuclides were measured using a Canberra HPGe detector with a relative efficiency of 10%. At the Nuclear Science

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Program facility, Universiti Kebangsaan Malaysia (UKM), a ^{60}Co-ray energy line with a full width at half maximum (FWHM) of 1.9 keV was used at 1,332 keV. The specific activities were measured for 43,200 seconds (12 hours) per sample through direct high-resolution γ-ray spectrometry. The activities of ^{238}U and ^{232}Th were determined from the photo peaks of ^{214}Bi (1764.49 keV) and ^{214}Pb (351.5 keV), and ^{208}Tl (2614.53 keV) and ^{228}Ac (911.32 KeV), respectively. Meanwhile, ^{40}K was determined directly using the photopeak at 1460.8 keV. The radionuclide activity was calculated using the following equation 5 as shown in equation 1):

$$A\ (\mathrm{Bq/kg}) = \frac{N_A \ln 2}{T_{1/2} \times M} \quad (1)$$

Where; A is the concentration activity, N_A is the Avogadro constant (6.022×10^{23}), $T_{1/2}$ is the half-life of the sample, and M is the molar mass of the sample (g). Equation 2 was used to determine the specific radionuclide activity in water, η:

$$\eta = N/A \quad (2)$$

Where; N = net counting and A = specific activity. The specific activity, A, as shown in Equation (3) is defined as

$$A = \frac{\text{Activiti (cps)}}{\varepsilon\left(\frac{\text{cps}}{\text{bq}}\right) \times \delta \times \text{sample weight}} \quad (3)$$

Where; ε = efficiency of the equipment (cps/Bq) and δ = abundance of the radionuclide.

The enrichment factor (EF) can be calculated using equation 4. This enrichment factor gives information about the activity concentration that has been increased from a material to another material.

EF = activity concentration of ash/
activity concentration of coal (4)

Radiation indices

Radiation indices are important for assessing the health effects caused by radiation hazards. The index is a reference point that defines relative radiation dose levels [14].

Radium equivalent activity (Ra_{eq})

The Ra_{eq} index as shown in equation 5 was created to represent the activity levels of ^{238}U, ^{232}Th, and ^{40}K while considering the radiological dangers. The United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) has mathematically defined it as follows [5]:

$$Ra_{eq}\ (\mathrm{Bq/kg}) = A_U + 1.43A_{Th} + 0.077A_K \quad (5)$$

Where A_U, A_{Th}, and A_K are the specific activities of ^{238}U, ^{232}Th, and ^{40}K, respectively. Meanwhile, 1.43 and 0.077 are conversion factors for thorium and potassium. Therefore, the maximum value of Ra_{eq} must be less than or equal to 370 Bq/kg.

Absorbed dose rate (ADR)

The ADR is defined as the ratio of an incremental dose (dD) over a particular time interval calculated one meter above the ground as shown in equation (6). The elements ^{238}U, ^{40}K, and ^{232}Th are assumed to be uniformly distributed. In addition, other radionuclides are assumed to have fewer effects on the total environmental background dose [15]. The absorbed dose can be computed as follows, based on the UNSCEAR guidelines [5]:

$$\mathrm{ADR\ (nG/h)} = 0.462A_U + 0.621A_{Th} + 0.0417A_K \quad (6)$$

Where 0.462, 0.621, and 0.0417 are the conversion factors for ^{238}U, ^{232}Th, and ^{40}K, respectively. Another assumption is that the naturally occurring radionuclide contribution can be overlooked as it has an insignificant effect on the total dose from background radiation. The world average for ADR is 57 nGy/h [5].

Annual effective dose equivalent (AEDE)

AEDE is used to estimate annual effective doses. It considers (a) the conversion coefficient from the absorbed in the air to the effective dose and (b) the indoor occupancy factor as shown in equation (7-8). The following formulas represent the components of the annual effective dose [16]:

Indoor (nSv) = absorbed dose nGy/h × 8760 h × 0.8 × 0.7 SvG/y (7)

Outdoor (nSv) = absorbed dose nGy/h × 8760 h × 0.2 × 0.7 SvG/y (8)

The UNSCEAR 1993 Report [16] used 0.7 SvG/y as the conversion coefficient from the absorbed dose in the air to the effective dose received by adults. It used 0.2 and 0.8 for the outdoor and indoor occupancy factors, respectively. Thus, the recommended values of AED for outdoor and indoor are 70 µSv/y and 450 µSv/y, respectively [17].

Hazard index

External and internal exposure are indicated by widely used hazard indices, namely the external hazard index (H_{ex}) and internal hazard index (H_{in}) as shown in equation (9, 10). The indices can be calculated using the following equations (5):

$$H_{ex} = \frac{Au}{370} + \frac{ATh}{259} + \frac{Ak}{4810} \quad (9)$$

$$H_{in} = \frac{Au}{185} \quad \frac{ATh}{259} + \frac{Ak}{4810} \quad (10)$$

Where A_U, A_{Th}, and A_K are the specific activities of ^{238}U, ^{232}Th, and ^{40}K, respectively. It can be safely accepted that the same gamma dose rate can be achieved from 4,810 Bq/kg ^{40}K, 259 Bq/kg of ^{232}Th, and 370 Bq/kg of ^{226}Ra. The conversion factor for indoor hazards is half that for outside hazards for ^{238}U. The value of the hazard index should not exceed 1 for both external and internal hazards.

Annual gonadal dose equivalent (AGDE)

To estimate annual effective doses as shown in equation (11), consideration must be made of (a) the conversion coefficient from the absorbed dose in the air to the effective dose and (b) the indoor occupancy factor. Thus, the components of the annual effective dose are determined as follows [16]:

$$\text{AGDE } (\mu Sv/year) = 3.09A_U + 4.18A_{Th} + 0.314A_K \quad (11)$$

Where A_U, A_{Th}, and A_K are the specific activities of ^{238}U, ^{232}Th, and ^{40}K, respectively. The 3.09, 4.18, and 0.314 values are the conversion factors for the specific activity. The average world value of AGDE is 300 µSv/y [17].

Representative gamma index (Iϒr)

Iϒr as shown in equation (12) is the gamma radiation hazard due to the specific concentrations of the studied natural radionuclides. The index is a screening factor for materials with the potential to pose radiation health risks. It can be estimated using the Ousif *et al.* [18] equation below:

$$I\Upsilon r = \frac{AU}{300} + \frac{ATh}{200} + \frac{Ak}{3000} \quad (12)$$

300, 200, and 3000 are the final activity concentration index of radium, thorium and potassium, respectively. These values are rounded to the nearest full 100 Bq/kg for radium and thorium or 1000 Bq/kg for potassium. Values of Iϒr less than or equal to 2 (Iϒr ≤ 2) are related to a dose rate criterion of 0.3 mSv per year, whereas Iϒr ≤ 6 corresponds to a dose rate criterion of 1 mSv/year [18]. Therefore, the value of Iϒr should be equal to or less than 6, which corresponds to an annual effective dose of ≤ 1 mSv/y [15].

Excess lifetime cancer risk (ELCR)

In today's world, cancer has emerged as a major disease facing communities. One cause of cancer involves the effect of radiation on the biological cell. Hence, another important parameter is ELCR, which can be estimated using equation (13) [12]:

$$\text{ELCR} = \text{AEDE} \times \text{DL} \times \text{RF} \quad (13)$$

Where AEDE is the annual effective dose, DL is life duration (70 years), and RF is the risk factor (0.05 Sv^{-1}). As per the recommendation of the International Commission on Radiological Protection (ICRP), the public's stochastic effects risk factor, RF, is 0.05 [19].

XRD analysis

The elements of fly ash, bottom ash, and coal were determined using XRD (Bruker D8 Advance) with a Cu Kα source (λ = 0.1542 nm) from 2θ = 10° – 80° by increasing the rate 0.02° every 40 seconds.

Statistical analysis

Basic descriptive statistics such as mean and standard deviation have been provided regarding a dataset's central tendency and variability. An overview of the data's features and distribution has been shown. The average or typical value of a variable can be found by computing the mean, which is helpful for understanding the overall trend. The standard deviation calculates how widely apart the data points are from the mean and provides a measure of how much the values differ from the mean. Equations (14 and 15) have been used to obtain the mean and standard deviation.

$$\bar{X} = \frac{\sum X}{N} \quad (14)$$

$$\sigma = \sqrt{\frac{\sum_i^n (X_i - \bar{X})^2}{n-1}} \quad (15)$$

Where; $\bar{X}$ is the mean, ∑X is the sum of value and N is the number of samples. Meanwhile, σ is the standard deviation, X_i is each value and n is total number of samples.

Specific activity of the soil, water, and sediment

Based on table 2, the ranges of activity concentration for ^{238}U, ^{232}Th, and ^{40}K were 22.7–150.7 Bq/kg, 20.7–153.6 Bq/kg, and 68.6–1594.4 Bq/kg, respectively. The highest activity concentration was found at T14, located near the palm estate. The use of fertilizer by the palm estate contributes to the higher activity concentration of radionuclides in the soil. Meanwhile, figure 2 shows the activity concentration of radionuclides in sediment and water.

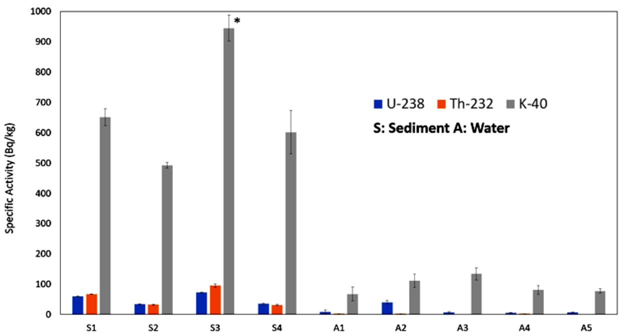

Figure 2. Activity concentration of radionuclides in sediment and water. *Indicates the significant value of the specific activity at S3 Point.

Generally, the activity concentrations of all samples were below the average for Malaysia, ranging from 16 to 46 Bq/kg, 28 to 87 Bq/kg, and 171 to 690 Bq/kg for ^{238}U, ^{232}Th, and ^{40}K, respectively.

Figure 3 shows the specific activities of ^{238}U, ^{232}Th, and ^{40}K for coal, fly ash, and bottom ash at the coal power plant. The specific activities of ^{238}U, ^{232}Th, and ^{40}K for coal were 67.54, 50.20, and 327.54 Bq/kg, respectively. Meanwhile, for fly ash, the specific

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activities of ^{238}U, ^{232}Th, and ^{40}K were 189.18, 134.57, and 1,114.40 Bq/kg, respectively. Finally, the bottom ash specific activities of ^{238}U, ^{232}Th, and ^{40}K were 186.32, 126.35, and 806.40 Bq/kg, respectively.

Table 2. Activity concentrations (Bq/kg) in each point of the soil (T) of U-238, Th-232, and K-40 surround the coal power plant. The average of activity concentrations of U-238, Th-232 and K-40 are 82.3, 82.3, and 677.95 Bg/kg. K-40 shows the highest value since it is the most abundant radionuclide in the earth.

Sample	U-238	Th-232	K-40
T1	81.8 ± 3.7	93.24 ± 4.2	579.3 ± 16.3
T2	27.0 ± 0.5	28.7 ± 1.4	323.9 ± 20.2
T3	23.4 ± 1.5	25.2 ± 0.2	230.1 ± 12.6
T4	22.7 ± 1.0	20.7 ± 0.6	284.5 ± 5.6
T5	88.0 ± 5.8	100.6 ± 6.9	656.4 ± 27.4
T6	77.7 ± 1.7	42.1 ± 3.7	739.7 ± 10.9
T7	118.4 ± 7.4	67.1 ± 3.1	936.0 ± 17.6
T9	81.6 ± 6.9	76.2 ± 3.7	844.8 ± 28.5
T10	92.3 ± 8.1	85.7 ± 4.0	588.8 ± 3.8
T11	77.2 ± 4.0	106.1 ± 2.1	503.8 ± 16.2
T12	69.7 ± 1.8	73.6 ± 3.7	68.6 ± 2.0
T13	146.4 ± 3.0	145.7 ± 7.9	463.2 ± 5.6
T14	150.7 ± 9.1	153.6 ± 7.3	409.9 ± 5.7
T15	146.8 ± 3.4	85.9 ± 3.0	1387.1 ± 4.7
T16	83.2 ± 0.6	113.3 ± 2.8	798.5 ± 53.0
T17	69.6 ± 3.0	80.9 ± 4.7	1022.6 ± 24.8
T18	69.3 ± 4.2	92.1 ± 2.1	1594.4 ± 1219.6
T19	56.4 ± 3.0	90.9 ± 3.4	771.5 ± 29.1
Minimum	22.7 ± 0.5	20.7 ± 0.2	68.8 ± 2.0
Maximum	150.7 ± 9.1	153.6 ± 7.9	1594.4 ± 1219.6
Average	82.3 ± 3.8	82.3 ± 3.6	677.95 ± 83.5
Malaysia	67	82	31
World	35	40	400

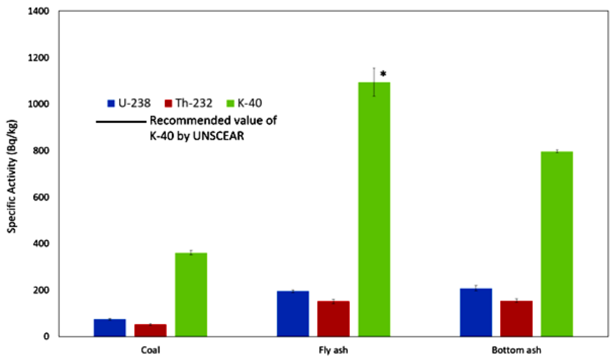

Figure 3. Activity concentration of radionuclides in coal, fly ash, and bottom ash. *Indicates the significant value of K-40 in S3 fly ash.

Radiation indices

The Ra_{eq}, ADR, IYr, AEDE, and AGDE values are tabulated in table 3. The Ra_{eq} index can be used to characterize the radiation risk posed by various radionuclide combinations in a particular material. The Ra_{eq} in this study oscillated between 164.55 and 467.42 Bq/kg with a mean of 353.69 Bq/kg. The values for fly ash (467.42 Bq/kg) and bottom ash (429.09 Bq/kg) exceeded the safety limit of 370 Bq/kg. The Ra_{eq} value for coal of 164.55 Bq/kg was below the acceptable safety limit.

Table 3. A specific value of radium equivalent (Raeq,) absorbed dose rate (ADR), iγr, annual effective dose equivalent (AEDE), and annual gonad dose equivalent (AGDE) have been measured according to the specific equations. Based on these data, it can be concluded that the radiation exposure (AEDE) is below 1 mSv/y as stated in Malaysian regulation even though there are increasing values of activity concentration of fly ash and bottom ash.

Sample	Raeq (Bq/kg)	ADR (nG/h)	Iγr	AEDE (mSv/y)		AGDE (µSv/year)
				Indoor	Outdoor	
Coal	150.90 ± 20.87	81.53 ± 1.02	0.49 ± 0.11	0.40 ± 0.01	0.09 ± 0.01	559.32 ± 6.69
Fly ash	421.57 ± 62.43	228.94 ± 2.11	1.38 ± 0.32	1.12 ± 0.01	0.28 ± 0.01	1573.63 ± 13.75
Bottom ash	416.30 ± 64.55	224.51 ± 2.79	1.44 ± 0.33	1.10 ± 0.02	0.28 ± 0.02	1533.66 ± 18.58
Mean	329.59 ± 49.28	178.33 ± 1.97	1.10 ± 0.25	0.87 ± 0.12	0.22 ± 0.12	1408.54 ± 13.01

Radiologically, the operation of the studied coal power plant does not appear to pose an immediate threat to its workers. This study found ADR values of 76.04 nG/h for coal, 217.44 nG/h for fly ash, and 198.17 nG/h for bottom ash. The average ADR worldwide is 57 nG/h (5). All of the coal, fly ash, and bottom ash samples exceeded the world average value. The bottom ash value was two times higher than the world average.

Meanwhile, IYr is associated with the annual dose rate due to the extra external gamma radiation created by superficial material. The study found IYr values of 0.56 for coal, 1.58 for fly ash, and 1.45 for bottom ash, for an average of 1.20. The values of IYr for coal, fly ash, and bottom ash were all below 2. Values of IYr ≤ 2 correspond to a dose rate criterion of 0.3 mSv/y, whereas IYr ≤ 6 relates to a standard of 1 mSv/y. In addition, the indoor and outdoor AEDEs were calculated. The indoor and outdoor AEDEs for coal were 0.37 mSv/y and 0.09 mSv/y, respectively; for fly ash these values were 1.07 mSv/y and 0.27 mSv/y, respectively, and for bottom ash they were 0.97 mSv/y and 0.24 mSv/y. Furthermore, the value of AGDE oscillated between 521.28 and 1,496.99 µSv/y with an average of 1,125.12 µSv/y, while the worldwide average value of AGDE is 300 µSv/y (17).

Hazard index and excess lifetime cancer risk (ELCR)

The hazard index values and calculated ELCR for coal, fly ash, and bottom ash are tabulated in table 4. The calculated external hazard index values for coal, fly ash, and bottom ash ranged from 0.44 to 1.16 with an average of 0.95. The internal hazard index values ranged from 0.63 to 1.66 with an average of 1.35. The ELCR depends on the AEDE. The indoor count's values ranged from 1.30×10^{-3} to 3.75×10^{-3} with an average of 2.82×10^{-3}. The outdoor calculation for ELCR fluctuated from 0.32×10^{-3} to 0.84×10^{-3} with a mean value of 0.71×10^{-3}.

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Table 4. Hazard index (indoor and external) and Excess Lifetime Cancer Risk (ELCR) to know the possibility that the public would obtain cancer due to coal, fly ash, and bottom ash. These hazard indexes and ELCR show how much the public could have a health risk from the coal, fly ash and bottom ash.

Sample	Hazard index		ELCR ($\times10^{-3}$)	
	Indoor	External	Indoor	Outdoor
Coal	0.67 ± 0.01	0.48 ± 0.01	1.40 ± 0.02	0.35 ± 0.01
Fly ash	1.86 ±0.02	1.33 ± 0.01	3.93 ± 0.04	0.98 ± 0.01
Bottom ash	1.88 ± 0.03	1.32 ± 0.02	3.85 ± 0.05	0.96 ± 0.02
Mean	1.47 ± 0.02	1.04 ± 0.01	3.06 ± 0.04	0.76 ± 0.01

XRD analysis

Figure 4 shows the XRD peaks for coal, fly ash, and bottom ash. An XRD analysis was used to determine the elements contained before and after coal combustion. As indicated in figure 4(a), the coal mainly contained carbon elements. The analysis detected carbon at nearly 72 wt% in the coal, while the other elements were H, O, and N [25].

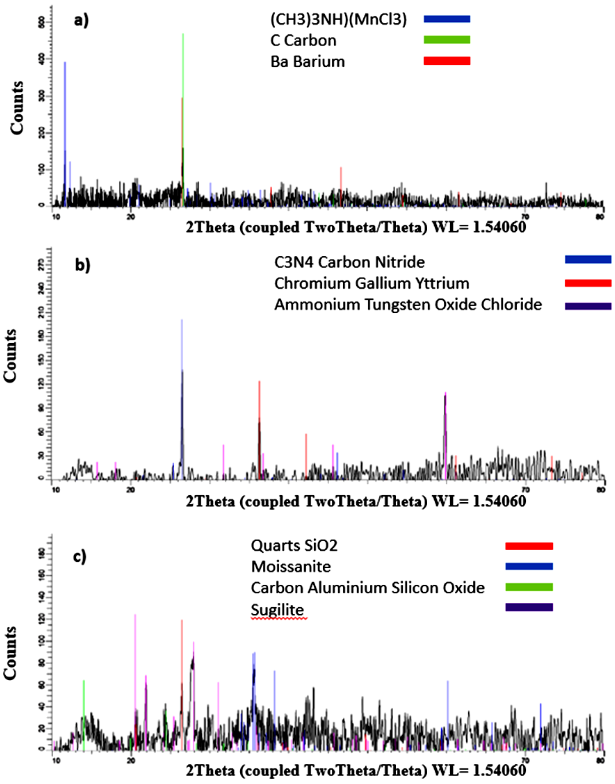

Figure 4. XRD analysis of **a)** coal **b)** fly ash and **c)** bottom ash.

DISCUSSION

The enrichment factor (EF) can be determined using equation (4) and the specific activity results for coal, fly ash, and bottom ash. Based on the calculation, the EFs for ^{238}U, ^{232}Th, and ^{40}K in the fly ash were 2.80, 2.68, and 3.40, and the EFs in the bottom ash were 2.76, 2.51, and 2.46, respectively. These results shows that the specific activities of fly ash were higher than in coal and bottom ash. Much research has found similar results, with fly ash specific activities that are higher than those for coal and bottom ash [13]. Table 4 compares this study's results to the results from coal-fired power plants in various countries. The enrichments of ^{238}U and ^{226}Ra in fly ash particles are high due to their large surface-to-volume ratios and diminutive size [13]. The average specific activity values recommended by reference [5] for ^{238}U, ^{232}Th, and ^{40}K are 30, 35, and 400 Bq/kg, respectively. The present study found that all specific activity values for ^{238}U, ^{232}Th, and ^{40}K were higher than the worldwide average. Fly ash had the highest specific activity among these, at double the worldwide average.

Table 5. Tabulated data of radionuclide activities of coal, fly ash, and bottom ash in other countries and been compared with this study. These data show that activity concentration of fly ash and bottom ash have been increased after the incineration process in several countries.

Samples	Country	Radionuclide activities (Bq/kg)				Authors
		U-232	Ra-226	Th-232	K-40	
Fly ash	Malaysia	157.71± 8.55	27.42± 2.27	134.41± 4.44	321.65± 12.73	[13]
Fly ash	China	-	46.4-1799.4	59.3-202.4	123.3-906.3	[28]
Fly ash	Turkey	-	77.0-272.0	9.0-696.0	12.0-2974.0	[29]
Coal	Hungary	-	43.0±6.0	26.0±6.0	210.0± 25.0	[20]
Fly ash		-	178.0± 31.0	55.0± 19.0	387.0± 48.0	
Bottom ash		-	144.0± 19.0	84.0±40.0	260.0± 25	
Coal	India	-	20.25-32.23	31.75-44.5	505.56-612.12	[9]
Fly ash		-	14.05-26.12	23.72-44.55	532.33-929.46	
Coal	Spain	-	64.0	18.0	104.0	[30]
Coal	South Africa	-	21.69-52.63	19.91-22.97	88.43-110.76	[31]

It can be concluded that the specific activity concentration in the fly ash is significantly higher than in the coal and bottom ash. This high concentration occurs because NORM enrichment in fly ash increases as the fly ash particle size decreases [5]. The filtering system collects smaller particles less efficiently, and these particles thus preferentially escape from the plant and appear in the environment. Kardos *et al.* [20] reported that the fly ash from a coal-fired power plant in Hungary had a higher specific value than bottom ash and coal. The study stated that the specific value for a sample comprising grain sizes smaller than 0.1 mm was 45% higher than that a sample with grain sizes of 1.6 mm. The natural radioelements from the fly ash spread into the atmosphere as sediment and fall onto the soil surface. The radionuclide activity in this study was compared with that of coal power plants in other countries; the results are tabulated in Table 3. Most countries, including China and India, had higher specific activity radionuclides of ^{40}K than the world average, which is 400 Bq/kg. Research by Al-Areqi *et al.* [13] reported

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that the specific activities of ^{238}U, ^{232}Th, and ^{40}K were 20.05, 5.87, and 321.65 Bq/kg, respectively, which are lower than the worldwide averages recommended by UNSCEAR. This study found that the fly ash from the coal power plant had a higher NORM concentration value than that of industrial incinerators.

The acceptable safety limit calculated for Ra_{eq} is 370 Bq/kg (OECD, 1979; UNSCEAR, 1988). In addition, based on UNSCEAR 1988, a Ra_{eq} value of 370 Bq/kg in building materials will produce an annual dose rate of about 1.5 mSv/y for the inhabitants. In a study by Hasan *et al.* [21] on a Bangladesh coal power plant, the average Ra_{eq} values for coal, fly ash, and bottom ash were 44.26, 251.37, and 170.38 Bq/kg, respectively. These values are below the worldwide average limit. Meanwhile, in Nigeria's fertilizer industries, the Ra_{eq} activity was found to be 215.59 to 607.29 Bq/kg [22]. Even though the fly ash and bottom ash values exceeded the safety limit of 370 Bq/kg, the mean value of Ra_{eq} was still below the safety limit.

Meanwhile, the Ra_{eq} for fly ash was three times higher than the world average value. A study conducted by Asaduzzaman *et al.* [23] on radiological risk in common building materials in Bangladesh reported that one of the building materials was fly ash, a by-product of coal combustion. Fly ash can be used as an additive in cement and other materials such as cement, brick, clinker, and white and red sand. The ADR value for fly ash as an additive is 304.7±35.5 nG/h, which is 1.5 higher than the ADR value of fly ash in the coal power plant studied here. However, the mean ADR levels in the cement, brick, and sand samples were found to fall within the typical worldwide range for building materials, which is 20 to 200 nG/h [5]. Therefore, the fly ash analyzed in this study could potentially be considered for use as an additive material in building construction as the ADR value was lower than the safety limit for building materials.

The gamma index is used as a screening tool for identifying materials that might be of concern when used in construction. Materials with $I\Upsilon r > 6$ should be avoided because this limit indicates dose rates of more than 1 mSv/y [18]. Since their values of $I\Upsilon r$ were below 6, the fly ash and bottom ash in this study might be safe for use as a concrete mixture for construction [20, 24]. According to one study, the fly ash used as an additive in building materials in Bangladesh has a $I\Upsilon r$ of 1.7± 0.19 [23]. This value is almost the same as the $I\Upsilon r$ of fly ash in this study.

The worldwide average AEDE dose is 0.48 mSv, implying that individual countries are generally within the 0.3–0.6 mSv/y range [5]. The recommended value for AEDE outdoors is 0.07 mSv/y, while the recommended value indoors is 0.45 mSv/y [17]. The outdoor values of AEDE for the coal, fly ash, and bottom ash in this study exceeded the equivalent dose of 0.07 mSv/y. For indoor AEDE, only coal AEDE did not exceed 0.45 mSv/y, while the fly ash and bottom ash values exceed the worldwide average of 0.48 mSv. Since coal combustion is released outside the building, it is considered safe as it does not exceed the limit. Moreover, ICRP has recommended an AEDE limit of 1 mSv/y for the public and 20 mSv/y for radiation workers [19].

All of the coal, fly ash, and bottom ash AGDE values in this study exceeded the average world value of 300 µSv/y, which is significantly higher than the global population-weighted average value. Kolo *et al.* [15] assessed the radiological dangers posed by natural radioactivity in the vicinity of the Lynas Advanced Material Plant (LAMP) in Kuantan, Pahang, Malaysia. LAMP, the world's largest rare earth refinery project, is based in Malaysia and is solely responsible for the production and recovery of rare earth elements (REEs) from concentrated raw materials imported from Australia. In the study, the AGDE values in the soil outside LAMP oscillated from 24.52 to 151.74 µSv/y with a mean of 77.58 µSv/y. This value is lower than the values for coal and the ashes at our studied coal power plant.

The external hazard index (H_{ex}) is used to calculate the radiation risk posed by gamma rays emitted externally by the samples. In contrast, the internal hazard index (H_{in}) is used to calculate the internal radiation risk posed by the samples, as suggested by UNSCEAR [5]. The value of the hazard index, either internal or external, must not exceed 1. For coal, H_{in} and H_{ex} were 0.63 and 0.44, respectively, which are both below the limit and thus considered safe. For fly ash and bottom ash, the index values exceeded 1. In 1999 the European Commission stated that an area that exceeds 1 is considered unsafe, posing a radiological threat to the population. The H_{ex} values for coal, fly ash, and bottom ash at Bangladesh's Barapukuria Coal Fired power plant are 0.16, 0.68, and 0.46, respectively, which are lower than those of the studied power plant.

For ELCR, ICRP [19] assigned a value of 0.05 (5×10^{-2}) for the public in terms of stochastic effects. Meanwhile, 0.29×10^{-3} is the average world value fixed by UNSCEAR [5]. In this study, the value of ELCR did not exceed 0.05. Nonetheless, the outdoor and indoor values for fly ash and bottom ash exceeded the value of 0.29×10^{-3}, the limit set by UNSCEAR. This result indicates that the lifetime cancer risk is low both for workers and the public. Similarly, the study by Kolo *et al.* [15] estimated that the mean value of ELCR for soil at LAMP was 0.049×10^{-3}. Therefore, even though the value of ELCR in the studied coal power plant was higher than at Lynas, it was still below the acceptable limit. Hence, the cancer risk among the population living outside the coal power plant is insignificant.

The XRD results showed that after the coal went through the combustion process in the power plant

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at temperatures over 500°C, carbon nitride was observed in the XRD peak for fly ash (figure 4(b)). This phenomenon occurs due to the calcination process that requires heating carbon and nitrogen to more than 600°C for 5 hours [26]. In addition, quartz was detected in the bottom ash sample (figure 4(c)) at a very low intensity. A similar result was reported by Varinporn *et al.* [27], where the XRD peak showed quartz peaks with low counts near the background.

CONCLUSION

In this study, the highest specific activities of ^{238}U, ^{232}Th, and ^{40}K were found in fly ash, followed by bottom ash and then coal. The fly and bottom ash values also exceeded the maximum value of radium equivalent and absorbed dose at nine times the acceptable value. The annual effective outdoor (public) dose was below the global limit. Thus, the effective dose is acceptable for the public and workers. The ELCR counts were below the permitted limit, implying that the risk of cancer among people living outside coal power plants is negligible.

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