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Assessment of natural radioactivity in coals and coal combustion residues from a coal-based thermoelectric plant in Bangladesh: implications for radiological health hazards

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Abstract To study the level of radioactivity concentrations from a coal-based power plant (Barapukuria, Bangladesh) and to estimate the associated radiological hazards, coal and associated combustion residuals from the power plant were analyzed by gamma-ray spectrometry with high-purity germanium (HPGe) detector. The results reveal that the mean radioactivity (Bq kg^{-1}) concentrations in feed coal samples are 66.5 ± 24.2, 41.7 ± 18.2, 62.5 ± 26.3, and 232.4 ± 227.2 for U-238, Ra-226, Th-232, and K-40, respectively, while in coal combustion residuals (CCRs), they are 206.3 ± 72.4, 140.5 ± 28.4, 201.7 ± 44.7, and 232.5 ± 43.8, respectively. With the exception of K-40, all the determined natural radionuclides are considerably higher in the investigated feed coal and associated combustion residues as compared with the world soil and world coal mean activities. On the average, CCRs contains 3.10–3.37 times more natural radionuclides than the feed coal, except for K-40. The radioactivity of fly ash and bottom ash is fractionated, and ratio ranges from 1.40 to 1.57. The mean values of the radiological hazard indices in the coal and their associated residuals are 153.1 and 446.8 Bq kg^{-1} for radium equivalent activity, 0.41 and 1.21 for the external hazard index, 70 and 200.1 nGy h^{-1}

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for the absorbed gamma dose rate, 0.09 and 0.25 mSv year^{-1} for the annual effective dose rate, and 3.0×10^{-4} and 8.6×10^{-4} Sv^{-1} for the excess lifetime cancer risk, respectively, most of which exceed the UNSCEAR-recommended respective threshold limits. The outcome of this study suggests a potential radiological threat to the environment as well as to the health of occupational workers and nearby inhabitants from the examined samples.

Keywords Coal-fired thermoelectric plant · Bituminous coal · Fly ash · Bottom ash · Radioactivity · Radiological hazard indices

Introduction

Globally, radiation and the associated potential threat to human health has become a matter of enormous public concern, even though it is an inevitable aspect of environmental materials (UNSCEAR 2000; 2010). The most common radioactive elements such as Th, U and their progenies, and K-40 are inherently present in natural geomaterials (Arbuzov et al. 2011, 2012; Orem and Finkelman 2014; Siegel and Bryan 2014; Chen et al. 2017a, b). Considering the natural abundances of common radionuclides (here, Ra-226, Th-232, U-238, and K-40), assessment of radiological health risk due to environmental radioactivity is more useful than simply studying the volumes of potentially hazardous wastes (Zivotić et al. 2008). Hence, obtaining the information regarding the level of radioactivity concentrations of natural radionuclides in coal and coal combustion residuals (CCRs, a collective term of fly ash, bottom ash and pond ash) is essential (Sajwan et al. 2011; Lauer et al. 2015, 2017) for evaluating the radiological risks.

Coal is the most ubiquitous and intensively used combustible hard fuel and its use in many developed and developing countries including Bangladesh is currently popular and indispensable (e.g., Mishra 2004; Özkul 2016; Khan et al. 2018a; Ozden et al. 2018). Presently, more than 40% of the total world electricity is produced from coal which represents the largest share of worldwide electricity production and this is expected to increase gradually (Amin et al. 2013). Moreover, coal contributes about 28% of the total world energy supply mix (IEA 2017). Previous study reported that ~ 5–20% of solid residuals known as fly ash and bottom ash are produced in coal-based thermal power plants (CTPs) which consist of 85–95% fly ash and 5–15% bottom ash with varying properties (Yao et al. 2015). Although the filtration systems of CTPs trap most of the total fly ash, between 1 and 3% of the total fly ash is released and distributed into the atmosphere and the biosphere (Papastefanou 2010). Geologically, coal carries small amounts of various toxic trace metals and minute amounts of (chemo)radiotoxic radionuclides (Khandekar et al. 1999; Orem and Finkelman 2014; Laraia 2015; Verma et al. 2015; Sengupta and Agrahari 2017), most of which are released from the coal matrix as a gas phase, as well as in solid form (fly ash and bottom ash) and in liquid discharge (leachates from ash ponds) (Bhangare et al. 2011, 2014; Dai et al. 2014; Ram et al. 2015). The combustion processes of a power plant enrich the concentrations of radionuclides in CCRs by 4 to 10 times compared to those of feed coal by reducing the coal volume about 85% (Mondal et al. 2006; Bhangare et al. 2014). The residual discharges from CTPs, which are enriched with heavy metals and radionuclides, are hazardous for human health and all compartments of the surrounding environment (Frontasyeva et al. 2001; Hu et al. 2010; Monir and Hossain 2012;Gupta et al. 2013; Ahamed et al. 2016; Zakir et al. 2017).

Therefore, despite the use of filtration systems, heavy metals and radiogenic pollutants from the combustion of coal are partly released into the atmosphere in the finest particulates form, most of which then fall to earth and enter the soil and surface water (Bhuiyan et al. 2010a, b; Halim et al. 2013, 2015; Krylov and Sidorova 2013; Dai et al. 2014; Al-Masri et al. 2015; Hossain et al. 2015). Furthermore, such pollutants may move from the disposal site (ash mound) to the groundwater mainly through leaching and may thus accumulate in the soil (Mahur et al. 2008; Dai et al. 2012; Howladar 2013; Hasani et al. 2014). They can also migrate through the water and air and enter the food chain (Megalovasilis et al. 2013; Skoko et al. 2017). Their presence (radionuclides) modifies the composition of environmental materials and elevates the level of natural background radiation through alpha and beta energy, gamma rays and spontaneous gaseous radon (Rn-222) release and the total dose rate to which all living things are exposed (Gupta et al. 2013). Radon is the most dangerous decomposition product of U (Amin et al. 2013; Bhangare et al. 2014). Thus, they may reach the human body through the intake of contaminated water and food, the inhalation of particulate pollutants, and

exposure to external radiation, which can cause various diseases, e.g., cell damage, lung, and bone cancer (Dai et al. 2012; Amin et al. 2013; HosgoodIII et al. 2013; Asaduzzaman et al. 2015; Munawer 2018). Thus, it is quite important to calculate the radiation risk to the population from the radioactivity of coal and associated CCRs.

Bearing in mind the adverse impact on the environment and the hazards to public health caused by radioactive elements from CTPs, there has been interest in studying naturally occurring radioactive materials (NORMs) since the 1960s (Eisenbud and Petrow 1964). Such investigations continue because of their relatively long half-lives of radionuclides, (eco)radiotoxicity, chemotoxicity, harmful effects of ionizing radiation exposure, and their potentiality to cause unavoidable environmental contamination, especially those associated with CTPs (Papastefanou 2010; Mahur et al. 2013; Swanson et al. 2013; Laraia 2015; Hower et al. 2016; Li et al. 2017). There has been extensive research around the world in the context of radionuclides derived from the combustion of coal and many studies have detected radiogenic signatures in materials originating from CTPs (Flues et al. 2006, 2007; Cevik et al. 2007, 2008; Turhan et al. 2010; Aytekin and Baldik 2012; El-Mekawy et al. 2015; Boukhair et al. 2016; Feng and Lu 2016; Hower et al. 2016; Sahu et al. 2017; Campaner et al. 2018; Ozden et al. 2018; Pak et al. 2018; Turhan et al. 2018; other references cited therein). Since, the burning of coal plays a central role in power production around the world, the harmful environmental impacts of radioactivity and the adverse human health effects from CTPs have received much attention from researchers among the world's scientific community (e.g., Atwood 2013; Michalik et al. 2013; Liu et al. 2015; Li et al. 2017; Sengupta and Agrahari 2017).

Currently, Bangladesh suffers from a deficiency of energy and many comprehensive initiatives have been taken to solve that shortage where coal-based power production is and will play a significant role (Ahamad 2016; Zaman et al. 2018). The energy sector in Bangladesh emphasizes the generation of electricity from coal. As of now, eight coal power projects are being constructed in the country with installed capacity of 6543 MW (BPDB 2017; Zaman et al. 2018). Therefore, the major portion of the total energy production in coming years is expected to be derived from coal energy (BPDB 2017). The current installed power generation capacity of the country is more than 12,780 MW which it is planned to increase up to 39,000 MW by 2030, of which about 50% would be derived from local and imported coal fuel source (PSMP 2010, 2016; Islam and Khan 2017; Zaman et al. 2018). Presently, the Barapukuria bituminous coal-based sub-critical thermal power station, BTPS, contributes only 3.75% of the total power while the largest portion of electricity(about 69.7%) is generated from local natural gas, which is being rapidly depleted (Mondal et al. 2010; PSMP 2010; 2016; BPDB 2017; Zaman et al. 2018).

According to the present development data, there are five coal basins in northwestern part of Bangladesh having probable coal reserves total 3.3 billion tons. The only Barapukuria coal basin (reserve 377 million tons, Mt) is in active commercial operation (Bakr et al. 1996; Islam and Hayashi 2008; Islam and Khan 2017). BTPS has been in operation since 2005, which possesses a generating capacity of 250 MW (2×125), combusts around 0.72 Mt. of local coal and generates nearly 0.08 Mt. of CCRs per annum (Hossain et al. 2015; Howladar and Islam 2016). These CCRs are usually sluiced into a nearby ash pond. The volume of these CCRs is expected to increase as third BTPS unit with a capacity of 275 MW has started since 2017. The release of CCRs into the atmosphere has been identified as the ultimate source of elevated levels of natural radioactivity in the environment and particularly affect inhabitants living close to CTPs. Previously several studies have been carried out on the Permian Gondwana coal mined at Barapukuria, Bangladesh, in the context of its palynological (Akhtar and Kosanke 2000), geological, sedimentological, petrological (Norman 1992; Bakr et al. 1996; Islam and Hayashi 2008; Farhaduzzaman et al. 2012; Hossain et al. 2014), and geochemical (Podder et al. 2004; Haider et al. 2011; Islam et al. 2011) properties. Earlier studies (Islam et al. 2011) on feed coal, fly ash, and bottom ash from BTPS have shown that the average (ranges) concentrations of Th were 9.6 (9.4–10.6 ppm), 61.4 (53.9–66.4 ppm), and 44.8 (41.8–47.0 ppm); of U, 2.1 (1.3–2.3 ppm), 13.7 (10.7–17.0 ppm), and 10(7.3–12.2 ppm); and of K, 0.10(0.8–0.15%), 0.57(0.50–0.62%), and 0.44(0.41–0.64%), respectively. Despite the long and extensive history of previous studies of radionuclides in different parts of the world, the levels of radioactivity and radiation doses in coal and associated CCRs from BTPS have not hitherto been

investigated. However, the systematic measurement and assessment of radionuclides and the quantification of the potential risk of radiation exposure are of prime importance for the environment and the health protection from the radiological hazards.

Therefore, the main objectives of the present study are to detect the levels of radioactivity and their distribution in coal and associated CCRs from Barapukuria coal mine (BCM) and BTPS and to estimate the hazard indices due to radioactivity from concentrations of radionuclides in coal and CCRs based on their radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), external gamma absorbed dose (D), annual effective dose (E), and excess lifetime cancer risk (ELCR) in order to assess the radiological health hazards risk in order to protect the environment, occupational workers, and local inhabitants from deleterious radiological effects arising from CCRs. The findings of the study will be useful in assessing public radiation doses and in monitoring environmental radioactivity. The results of this study are also anticipated to be applicable to the effective management of radiogenic pollutants containing CCRs originating from BTPS.

Experimental

Study area

BTPS and BCM are situated in a humid subtropical region in an agriculture dominant area in the northwestern part of Bangladesh (Fig. 1). The population density is 823/km^2. The mean annual rainfall is 1800 to 2000 mm of which 85% falls from May to September and the relative humidity is 80 to 90%. The prevailing dominant local wind direction is from east to west (40%) followed by west to east (25%) and northeast (18%). Wind speeds rarely exceed 8 m/s (BBS 2011). The major features of the BTPS and BCM are presented in Table S1. Properties of the feed coals from Barapukuria, Bangladesh, are presented in Table S2.

Sampling and sample preparation

Core and bulk coal, pulverized coal samples from coal seam VI, coal storage pile, and pulverizer source, respectively, were taken for radioactivity measurement. Samples of fly ash (FA) from the electrostatic precipitator (ESP), bottom ash (BA) from the bottom of the boiler of thermal plant units 1 (FA 1 and BA 1) and 2 (FA 2 and BA 2), and pond ash from the unified disposal mound (in single ash pond) were taken from BTPS at ten different times (i.e., ten sub-samples for each item from same location) at regular order and interval between March and April 2017. In order to prepare bias-free (spatial, geological, technical etc.), more representative and reduced sample number for each item, dried-up sub-samples (ten for each item) were thoroughly mixed and milled into powder (except fly ash and pulverized coal which were directly processed because they were already in powder form) by silicon nitride ball miller. Each item was homogenized using a 500-μm mesh size sieve and re-sampled as required. Further sample drying was conducted after air-drying for several samples by using oven at 105 °C for 24 h; however, the weight difference between pre- and post-oven drying is negligible for both coal and ash samples, except for bottom ash samples. It is probably because the coal and fly ash samples naturally contain very low amount of moisture and are furthermore dried by air-drying in the winter season with very low humidity.

The dried powder samples were packed in a U8 vessel with a dimension of 5 cm effective height and 5 cm effective diameter of its cap, mass weighed, and then hermetically sealed tightly around their necks with black electrical tape to prevent the loss of the radionuclides in the form of gaseous radon (Rn) and stored for at least 4 weeks to reach a stable equilibrium between the long half-life parent and the short half-life daughter radionuclides (U-238 and Th-232 chain radionuclides and their daughter products) prior to being measured. This procedure has been followed in many previous studies around the world (e.g., Hasani et al. 2014; Habib et al. 2018).

Measurement of NORMs

The bulk/core coal, pulverized coal, fly ash, bottom ash, and pond ash samples were investigated for the activity measurements of the natural radionuclides U-238, Ra-226, Th-232, and K-40 indirectly by means of a gamma-ray spectrometer with a low background HPGe semiconductor detector, (GEM 30-70, ORTEC) at the Radioisotope Center (RI), Hiroshima University, Japan, at 0 cm distance from the detector, i.e., the sample was placed exactly above the detector, although there were a

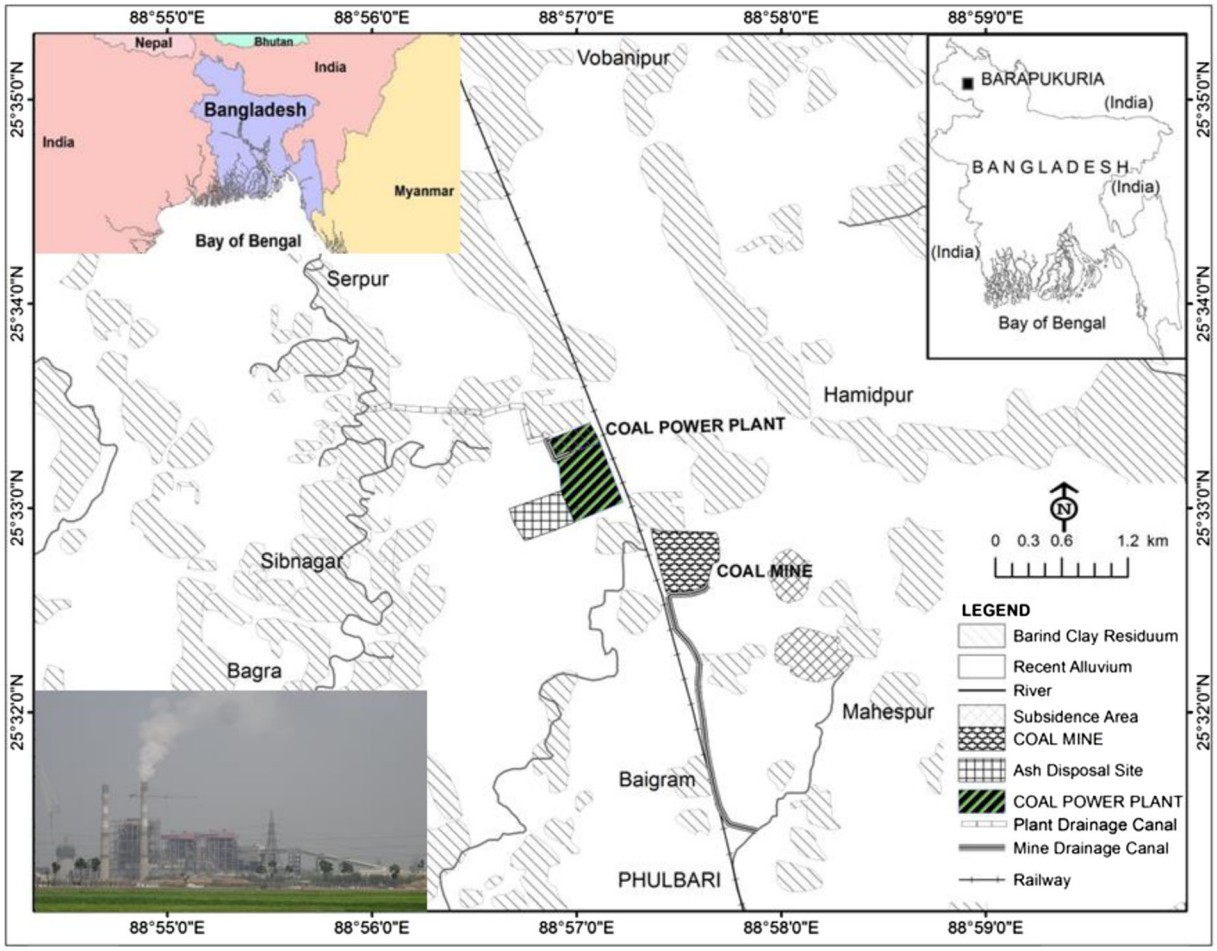

Fig. 1 The location of Barapukuria coal-fired thermal power plant (BTPS) and Barapukuria coal mine (BCM) area in Bangladesh

cover on the detector and bottom of U8 vessel between sample and detector. Detection efficiency calibration of the gamma-ray spectrometer was conducted by using the set of standard sources (MX033U8PP) which consist of radionuclides with known radioactivity emitting from low- to high-energy gamma ray, manufactured by the Japan Radioisotope Association (JRA). The set of standard sources that were used have different thicknesses, 5, 10, 20, 30, and 50 mm to consider the geometry of the samples. In principle, the radioactivity measured by using a gamma-ray spectrometer with decay correction was compared to the initial radioactivity of standard source measured by the JRA to obtain the detection or counting efficiency. The gamma-ray spectrometer counting efficiency for NORMs were estimated by using curve-fitting of energy and counting efficiency and furthermore counting efficiency and thickness of the sample to consider the geometry of the sample.

Regarding the traceability of the measurement, the screening measurement was conducted to analyze whether the radionuclides which are important in NORM analysis can be detected in the sample. It aims to evaluate whether the radionuclide of interest is traceable or not to compare with its background value. In the screening process, the background of the coal and ash samples were measured and compared. The background was measured in 3.82 days, one coal sample of Barapukuria about 60 g in 1.83 days and one fly ash sample about 80 g in 2.32 days. The quoted uncertainties (1σ) were calculated by error propagation calculation which included the data relating to the samples and the background and the efficiency calibration uncertainty.

Activity concentrations of NORMs were calculated by considering net count, counting efficiency, and emission rate of certain radionuclides and weight of sample. Equations (1) and (2) are as follows:

$$A = \frac{\mathrm{cps}_{\mathrm{sample}} - \mathrm{cps}_{\mathrm{BG}}}{\varepsilon \times I_\gamma} \quad (1)$$

$$\mathrm{AC} = \frac{A}{w} \quad (2)$$

where A is the activity(Bq), AC is the activity concentration (Bq kg^{-1}), cps_{sample} is the counts per second of the sample (s^{-1}), cps_{BG} is the counts per second of the background (s^{-1}), ε is the count efficiency of the HPGe detector, I_γ is the intensity of the gamma rays, and w is the sample weight (kg).

The radionuclides of concern in this measurement, which are long half-life radionuclides including Th-232, U-238, U-235, and Ra-226, were estimated based on the activity concentration of gamma rays of their progenies in the samples, with the exception of K-40, which can be measured directly. The activity concentrations of U-238 and Th-232 were determined indirectly by analyzing the full-energy peaks emitted by their progenies. In the Th-232 decay series, Ac-228, Tl-208, Pb-212, and Bi-212 were used to estimate the Th-232 (L'Annunziata 2003). In the U-238 decay series, Pb-214 and Bi-214 were employed to estimate the Ra-226. The activity of Ra-226 was estimated from the average value of the activity of Pb-214 and Bi-214 on four peaks. In the U-235 decay series, U-235, which emits gamma ray at 185 keV, was detected with an overlapping emission at 186 keV from Ra-226. The activity of U-238 was calculated based on the natural abundance ratio of U-235 and U-238. This radiochemical analysis technique is described elsewhere (Habib et al. 2018).

Enhancement ratio and enrichment factor

The enhancement ratio (ER) of radioisotopes was calculated as the ratio of activity concentrations in CCRs to its corresponding specific activity in feed coal (Sahu et al. 2014). The ER > 1.6 is considered for enrichment, whereas ER < 0.6 is considered for depletion (Patra et al. 2012; Usmani and Kumar 2017).

Enrichment factor (EF) of the natural radionuclides was calculated for ash samples to characterize their transformation behaviors. It was calculated as the ratio of the activity concentration of the radionuclide X and of K-40 in the CCRs divided by the corresponding ratio in the feed coal (Coles et al. 1978) by the following Eq. (3),

$$\mathrm{EF} = \frac{\left(\frac{A_X}{A_{K40}}\right)_{\mathrm{CCRs}}}{\left(\frac{A_X}{A_{K40}}\right)_{\mathrm{Coal}}} \quad (3)$$

where $_X$ denotes the radionuclides, and A_x is the corresponding specific activity (Bq kg^{-1}). The K-40 is used as a radio tracer since its concentration remains constant in the samples (Coles et al. 1978; Papastefanou 2010). The estimated EF values are categorized: EF < 2, deficiency to slight; 2 < EF < 5, moderate; 5 < EF < 20, significant; 20 < EF < 40, very high; and EF > 40, extremely high enrichment (Usmani and Kumar 2017).

Partition ratio, PR, was calculated with respect to the activity concentration of radionuclides in fly ash and bottom ash (and between fly ash and pond ash) (Table 1). The ratio PR > 1 indicates enrichment of radionuclides. Additionally, higher PR value also denotes the higher affinity of radionuclides with fly ash than bottom ash (pond ash) (Bhangare et al. 2011; Patra et al. 2012; Usmani and Kumar 2017).

Estimation of radiological hazards

Inherently, the activity concentrations of Ra-226, Th-232, and K-40 radionuclides in the environmental substances (e.g., coal, fly ash) are not uniform. In order to overcome the non-uniformity of the radionuclides, a common index called "radium equivalent activity (Ra_{eq})" is employed to attain the representing radioactivity and also to evaluate the potential gamma radiation exposure hazard risk due to different radionuclides in the geomaterials. The radiation exposure indices are commonly estimated by the activity results of Ra-226, Th-232, and K-40 (Durašević et al. 2014; Kolo et al. 2016). The Ra_{eq} (Bq kg^{-1}) is related to the external and internal gamma dose due to Rn gas emission and its daughter's emanation. It was calculated according to Eq. (4) (Beretka and Mathew 1985):

$$\text{Radium equivalent activity, } \mathrm{Ra_{eq}} = C_{\mathrm{Ra}} + 1.43\, C_{\mathrm{Th}} + 0.077\, C_{\mathrm{K}} \leq 370 \quad (4)$$

where C_{Ra}, C_{Th}, and C_K are the specific activities of Ra-226, Th-232, and K-40 in Bq kg^{-1}, in the materials, respectively. The maximum value of Ra_{eq} in samples must be less than 370 Bq kg^{-1} to be within the safety threshold and to avoid radiation exposure (UNSCEAR 2000; Gupta et al. 2013). This index can be used to

Table 1 Values of natural radioactivity and normalized enrichment factor (EF) of determined radionuclides in the samples from Barapukuria coal mine (BCM) and Barapukuria bituminous coal-based thermal power plant (BTPS) in this work and compared with world average specific activity values for different geomaterials

Sample	U-238		Ra-226		Th-232		K-40		EF		
	[Bq kg^{-1}]	[±]	[Bq kg^{-1}]	[±]	[Bq kg^{-1}]	[±]	[Bq kg^{-1}]	[±]	U-238	Ra-226	Th-232
Bulk and core coal samples from Barapukuria											
RC 1	56.4	22.6	26.4	3.3	56.5	1.9	25.90	5.0	–	–	–
RC 2	32.3	18.1	21.8	3.5	46.8	1.3	24.90	4.3	–	–	–
RC 3	63.0	19.1	25.1	2.3	16.4	1.5	13.90	3.9	–	–	–
RC 4	103.7	20.4	63.4	3.5	70.4	0.6	349.3	8.1	–	–	–
RC 5	94.3	17.7	48.9	1.3	86.8	3	420.3	8.7	–	–	–
RC 6	78.0	19.1	56.7	6.3	82.1	1.1	544.0	9.8	–	–	–
RC 7	59.7	25.6	63.5	6	95.8	1.1	442.4	9.6	–	–	–
$RC_{average}$	69.6	24.3	43.7	18.7	65.0	27.4	260.1	230.3	–	–	–
Min	32.3	–	21.8	–	16.4	–	13.9	–	–	–	–
Max	103.7	–	63.5	–	95.8	–	544.0	–	–	–	–
PC	44.9	13.4	27.6	2.3	45.5	1.1	38.2	5	–	–	–
$Coal_{average}$	66.5	24.2	41.7	18.2	62.5	26.3	232.4	227.2	–	–	–
Min	32.3	–	21.8	–	16.4	–	13.9	–	–	–	–
Max	103.7	–	63.5	–	95.8	–	544.0	–	–	–	–
CCRs from BTPS											
FA 1	203.9	43	165.5	18.4	231.2	1.5	260.2	13.2	2.74	3.55	3.30
FA 2	329.5	29.8	175.4	13.9	263.7	0.8	277.8	8.9	4.14	3.52	3.53
$FA_{average}$	266.7	88.8	170.5	7.0	247.5	23.0	269.0	12.4	3.44	3.53	3.41
BA 1	190.2	17	130.9	11.1	188.1	3.5	183.9	5.3	3.61	3.97	3.80
BA 2	149.3	24.5	111.9	5.3	157.7	0.8	187.0	9.9	2.79	3.34	3.13
$BA_{average}$	169.8	28.9	121.4	13.4	172.9	21.5	185.5	2.2	3.20	3.65	3.47
PA	158.8	36.4	119.0	9.4	167.7	1.6	253.4	13.1	2.19	2.62	2.46
$CCRs_{average}$	206.3	72.4	140.5	28.4	201.7	44.7	232.5	43.8	3.1	3.4	3.2
Min	149.3	–	111.9	–	157.7	–	183.9	–	2.2	2.6	2.5
Max	329.5	–	175.4	–	263.7	–	277.8	–	4.1	4.0	3.8
PR: $(FA/BA)_{average}$	1.57	–	1.40	–	1.43	–	1.45	–	–	–	–
PR: $(FA/PA)_{average}$	1.68	–	1.43	–	1.48	–	1.06	–	–	–	–
$WC_{average}$[a]	35 (16-110)	–	35 (17–60)	–	30 (11–64)	–	400 (140–850)	–	–	–	–
$WFA_{average}$[b]	200	–	240	–	70	–	265	–	–	–	–
$WS_{average}$[c]	24 (8–110)	–	30 (7–180)	–	37 (4–78)	–	440 (0.2–1200)	–	–	–	–
$CC_{average}$[d]	36	–	33	–	44	–	850	–	–	–	–
$BS_{average}$[e]	102.9	–	63.6	–	103.4	–	494.2	–	–	–	–
$(Coal/WC)_{average}$	1.90	–	1.19	–	2.08	–	0.58	–	–	–	–

Table 1 (continued)

Sample	U-238		Ra-226		Th-232		K-40		EF		
	[Bq kg^{-1}]	[±]	[Bq kg^{-1}]	[±]	[Bq kg^{-1}]	[±]	[Bq kg^{-1}]	[±]	U-238	Ra-226	Th-232
$(CCRs/WFA)_{average}$	1.03	–	0.59	–	2.88	–	0.88	–	–	–	–
$(Coal/WS)_{average}$	2.77	–	1.42	–	1.69	–	0.53	–	–	–	–
$(CCRs/WS)_{average}$	8.60	–	4.68	–	5.45	–	0.53	–	–	–	–
$(CCRs/BS)_{average}$	2.01	–	2.21	–	1.95	–	0.47	–	–	–	–
CF: $(Coal/CC)_{average}$	1.85	–	1.26	–	1.42	–	0.27	–	–	–	–
CF: $(CCRs/CC)_{average}$	5.73	–	4.26	–	4.58	–	0.27	–	–	–	–

RC raw (bulk/core) coal; *PC* pulverized coal; *FA* fly ash; *BA* bottom ash; *PA* pond ash; *Min* minimum; *Max* maximum; *CCRs* coal combustion residues, mean of different ashes; *PR* partition ratio; *WC* world coal average; *WS* world soil average; *WFA* world fly ash; *CC* continental crust; *BS* Barapukuria soil; *CF* contamination factor or coefficient of pollution

[a] UNSCEAR 2010

[b] UNSCEAR 1982

[c] Bowen 1979

[d] Eisenbud and Gesell 1997

[e] Habib et al. 2018

estimate the level of radiation hazard associated with the natural radionuclides in the materials.

The external hazard index, H_{ex}, evaluates the external radiation exposure from Ra containing materials and the index must be less than unity to be within the safety threshold and to avoid radiation hazards to the respiratory organs (UNSCEAR 2000; Hasani et al. 2014). It was calculated according to Eq. (5):

$$\text{External hazard index}, H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \leq 1 \quad (5)$$

The absorbed gamma dose rate, D (nGy h^{-1}), for plant staff, miners, and the local population, for a uniform distribution of Ra-226, Th-232, and K-40 was computed based on UNSCEAR (2000) according to Eq. (6):

$$\text{Absorbed gamma dose rate}, D = 0.462\ C_{Ra} + 0.604\ C_{Th} + 0.0417\ C_K \quad (6)$$

The annual effective dose rate, E (mSv year^{-1}), represents the radiation in air received by occupational workers and members of the public staying around the power plant were estimated on the basis of UNSCEAR (2000) (Eq. (7)):

$$\text{External effective dose}, E = D \times 10{-}3 \times 1.23 \quad (7)$$

The excess lifetime cancer risk, ELCR (Sv^{-1}), caused by the annual effective dose due to external exposure was estimated using Expression (8) (ICRP 1990):

$$\text{Excess lifetime cancer risk}, \text{ELCR} = E \times \text{ALT} \times \text{RF} \quad (8)$$

where ALT is the average life time (70 years for Bangladeshi people) and RF is the risk factor based on the fatal cancer risk per Sievert and stochastic effects. The ICRP uses a value of 0.5×10^{-4} for the public exposure ICRP (1990).

Results and discussion

Activity concentrations of radionuclides in coal and associated residuals

The activity concentrations of the U-238, Ra-226, Th-232, and K-40 radionuclides detected in bulk/core coal

and associated combustion residuals (CCRs) sampled are presented in Table 1. The data shows a non-uniform distribution and a wide variation in activity in the measured samples. In the bulk/core coal the mean activity concentrations of U-238 vary from 32.3 to 103.7 with a mean value of 69.6 ± 24.3(Bq kg^{-1}). The mean activity concentrations of Ra-226 range from 21.8 to 63.5 with an average value of 43.7 ± 18.7 (Bq kg^{-1}), and for Th-232, the mean activity concentrations range from 16.4 to 95.8 with a mean value of 65.0 ± 27.4 (Bq kg^{-1}). For K-40, the value varies from 13.9 to 544.0 with a mean value of 260.1 ± 230.3 (Bq kg^{-1}). Thus, with the exception of K-40, the activity concentrations of radionuclides in bulk/core coal of this study are 1.19 and 2.08 times higher than the world average concentration values for coal adopted by UNSCEAR (2000) (Table 1). Noticeably, the radioactivity of K-40 in most of our samples is much lower than the world average values. However, in pulverized coal, the average activity level of U-238, Ra-226, Th-232, and K-40 are 44.9 ± 13.4, 27.6 ± 2.3, 45.5 ± 1.1, and 38.2 ± 5.0 Bq kg^{-1}, respectively. The mean activity concentrations of U-238, Ra-226, Th-232, and K-40 in Barapukuria coal samples are 66.5 ± 24.2, 41.7 ± 18.2, 62.5 ± 26.3, and 232.4 ± 227.2 Bq kg^{-1}, respectively (Table 1). The study reveals that the specific activity in pulverized coal is lower than the activity concentrations in bulk/core coal samples. The presumable reason is due to preparation, pulverization of the bulk coal samples by removing undesired radionuclides containing heavier minerals, incombustible materials, and other impurities.

According to Hower et al. (2016), syngenetic, diagenetic, and epigenetic processes are considered for the accumulation and enrichment of radioactive elements (along with the other trace elements) in coal. The carbonaceous substances of sedimentary rocks contain a significant portion of the total U budget (Chen et al. 2017a, b) and the greater fraction of that U accumulates in coals during the initial stages of the coalification process(es) and subsequent burial stage(s) (Zivotić et al. 2008; Orem and Finkelman 2014). In the U accumulation process(es) in coal, sorptive uptake of the organic fraction of coal plays an important role in which chemical adsorption is empowered by the formation of strong humate complexes (Douglas et al. 2011; Huang et al. 2012; Singh et al. 2015). However, Th concentrates in coal as a detrital mineral grain from the source region (e.g., Swaine 2014) and the formation of organic Th is unlikely (Finkelman et al. 2018). Potassium is generally associated with inorganic materials, e.g., clays, which are common in coal (Swaine 2014; Finkelman et al. 2018).

In fly ash, the evaluated mean (ranges) activity concentrations are as follows: U-238, 266.7 ± 88.8 (203.9–329.5); Ra-226, 170.5 ± 7.0 (165.5–175.4); Th-232, 247.5 ± 23.0 (231.2–263.7); and K-40, 269 ± 12.4 (260.2–277.8) Bq kg^{-1}, respectively (Table 1). The specific activity of the radionuclides in fly ash are significantly higher (between 5.44 and 7.04 times) than the respective activity level in pulverized coal (Table 1). Similarly, in bottom ash, the calculated mean (ranges) activities are as follows: U-238, 169.8 ± 28.9 (149.3–190.2); Ra-226, 121.4 ± 13.4 (111.9–130.9); Th-232, 172.9 ± 21.5 (157.7–188.1); and K-40, 185.5 ± 2.2 (183.9–187.0) Bq kg^{-1}, respectively (Table 1). In pond ash, the activities are the following: U-238, 158.8 ± 36.4; Ra-226, 119.0 ± 9.4; Th-232, 167.7 ± 1.6; and K-40, 253.4 ± 13.1 Bq kg^{-1}, respectively (Table 1). The mean activity concentrations of U-238, Ra-226, Th-232, and K-40 in CCRs from BTPS are 206.3 ± 72.4, 140.5 ± 28.4, 201.7 ± 44.7, and 232.5 ± 43.8 Bq kg^{-1}, respectively (Table 1). The highest average activity level of U-238 is found in fly ash (266.7 ± 23.1) followed by bottom ash (169.8 ± 12.3), pond ash (158.8 ± 9.4), bulk/core coal (69.6 ± 24.3), and pulverized coal (44.9 ± 2.3) (all units are in Bq kg^{-1}) (Table 1). The specific activities in CCRs are dramatically higher than the respective activity in Barapukuria feed coals. It is clearly shown that the radionuclides are generally enriched in the CCRs after burning leading to higher radioactivity. The relative specific activity contributions of radionuclides in the samples are in descending order fly ash > bottom ash > pond ash > bulk coal > pulverized coal (Table 1). The activity results show that the concentration of Ra-226 is less than that of Th-232 and U-238 in the samples examined.

In comparison with the typical world soil average radioactivity of U-238, Ra-226, and Th-232, the activities are 1.42 to 2.77 times larger in coals and the respective activities are 4.68 to 8.60 times more in CCRs of this study, except for K-40 (Table 1). The respective activities are 1.19 to 2.08 times higher in coals than typical world coal average, 1.95 to 2.21 times more in CCRs than Barapukuria soil average values, and 1.26 to 1.85 times more in coal and 4.26 to 5.73 times more in CCRs than continental crust average, respectively (Table 1). The mean Th-232 concentrations in the CCRs

Table 2 Comparison of average activity (Bq kg^{-1}) of natural radionuclides and radiological hazard indices in coal and associated residuals (CCRs) from Barapukuria such as radium equivalent activity, Ra_{eq} (Bq kg^{-1}); external hazard index, H_{ex}; external absorbed gamma dose rate, D (nGy h^{-1}); annual effective dose rate, E (mSv $year^{-1}$); and excess life time cancer risk, ELCR (Sv^{-1}) computed following formulas adopted by UNSCEAR 2000 and compared with others published similar investigations

Country	Sample	Radioactivity				Radiological indices					Reference
		U-238	Ra-226	Th-232	K-40	Ra_{eq}	H_{ex}	D	E	ELCR	
Bangladesh	Coal	69.63	54.3	92.39	241.0	153.1	0.41	70.0	0.09	3.0×10^{-4}	Present study
	FA	266.7	165.5	231.2	260.2	545.0	1.47	243.9	0.30	10.5×10^{-4}	
	BA	169.75	130.9	188.1	183.9	367.4	0.99	164.5	0.21	7.1×10^{-4}	
	PA	158.8	119.0	167.7	253.4	378.3	1.02	169.8	0.21	7.3×10^{-4}	
India	Coal	–	16.8	19.5	37.2	47.5	0.13	21.4	0.03	0.9×10^{-4}	Sahu et al. 2014
	FA	–	78.8	61.7	99.1	174.7	0.47	78.9	0.10	3.4×10^{-4}	
	BA	–	41.4	24.4	9.5	77.0	0.21	34.7	0.04	1.5×10^{-4}	
China	Coal	–	33	37.5	105.7	94.8	0.26	43.0	0.05	1.8×10^{-4}	Lu et al. 2012
	FA	–	69.5	79.3	233	200.8	0.54	91.1	0.11	3.9×10^{-4}	
	BA	–	59.5	61.8	222.6	165.0	0.45	75.1	0.09	3.2×10^{-4}	
Brazil	Coal	–	321	–	191	–	–	–	–	–	Flues et al. 2006
	FA	1424	1284	–	764	–	–	–	–	–	
Nigeria	Coal	–	8.18	6.97	27.38	20.3	0.05	9.2	0.01	0.4×10^{-4}	Kolo et al. 2016
USA	Coal	8.9	7.4	6.3	27	18.5	0.05	8.5	0.01	0.4×10^{-4}	Coles et al. 1978
	FA	70.3	85.1	62.9	299.7	198.1	0.54	90.8	0.11	3.9×10^{-4}	
Turkey	Coal	14.55	11.12	123.01	14.55	39.9	0.11	18.7	0.02	0.8×10^{-4}	Cevik et al. 2008
	FA	149.43	57.97	94.15	149.43	239.6	0.65	108.9	0.13	4.7×10^{-4}	
	BA	49.96	24.72	375.89	49.96	114.3	0.31	54.0	0.07	2.3×10^{-4}	

RC raw (bulk/core) coal, *PC* pulverized coal, *FA* fly ash, *BA* bottom ash, *PA* pond ash, *CCRs* coal combustion residues

are therefore 2.88 times higher than those in the world fly ash (UNSCEAR 1982) (Table 1). From the comparison, it can be evidently seen that the obtained activity values are unusually higher in the investigated samples than in the typical world coal, in world soil, in Barapukuria soil, and in continental crust (except for K-40) (Table 1). Thus, the elevated specific activities are likely due to the presence of Th and U containing minerals such as monazite and zircon in the examined samples (e.g., Swaine 2014; Khan et al. 2017, 2018b; Finkelman et al. 2018).

In Table 2, a summary of the obtained activity concentrations in coals and CCRs samples of this study along with the literature data from similar investigations are tabulated. The obtained activity results for Ra-226 in the studied samples are significantly higher than those of the corresponding activity in coal and CCRs in Brazil (Flues et al. 2006), China (Lu et al. 2012), Greece (Karangelos et al. 2004), India (Sahu et al. 2014), and the USA (Coles et al. 1978) and are slightly lower than those in Nigeria (Kolo et al. 2016) and Poland (Bem et al. 2002).

Fractionation of radionuclides among the feed coals and CCRs

The calculated enrichment ratio, ER (ash/coal), suggests that all determined natural radionuclides are found to be enriched by a factor of 1.16 to 4.09 in fly ash (highest), by a factor of 0.08 to 2.91 in bottom ash, and by a factor of 1.09 to 2.86 in pond ash, respectively, as compared to feed coal. The specific activity of the NORMs in feed coal is 4.43 to 6.09 times lower than that in CCR samples (Table 3). The determined natural radionuclides are considerably enriched in CCRs and this enrichment ratio is the maximum for fly ash radionuclides. In feed coal, activity concentrations of primordial radionuclides are low, but the corresponding concentrations are considerably higher in CCRs in this study. The ER values for other countries calculated in earlier published works are compared with this work. The ER values in this study are higher than those of India (Sahu et al. 2014) and China (Lu et al. 2012), but lower than those for the USA (Coles et al. 1978), Spain (Mora et al. 2009), and Poland (Bem et al. 2002) (Table 3).

Table 3 Comparison of the enrichment ratio (ER) in the samples studied from Barapukuria with the literature data

Country		Enrichment ratio (ash/coal)				References
		U-238	Ra-226	Th-232	K-40	
Bangladesh	FA/coal	4.01	4.09	3.96	1.16	This study
	BA/coal	2.55	2.91	2.76	0.80	
	PA/coal	2.39	2.86	2.68	1.09	
	CCRs/coal	3.10	3.37	3.22	1.00	
India	FA/coal	–	4.7	3.2	2.7	Sahu et al. 2014
	BA/coal	–	2.5	1.3	0.3	
China	FA/coal	–	2.1	2.1	2.2	Lu et al. 2012
	BA/coal	–	1.8	1.6	2.1	
Turkey	FA/coal	–	10.3	5.2	0.8	Cevik et al. 2008
	BA/coal	–	3.4	2.2	3.1	
Greece	FA/coal	3.2	2.6	2.8	2.6	Karangelos et al. 2004
	BA/coal	2.2	1.9	2.3	2.3	
USA	FA/coal	7.9	11.5	10.0	11.1	Coles et al. 1978
	BA/coal	6.2	9.5	8.8	9.3	

PC pulverized coal, *FA* fly ash, *BA* bottom ash, *PA* pond ash, *CCRs* coal combustion residues

The normalized enrichment factor, EF (with respect to feed coal), values of U-238, Ra-226, and Th-232 are 3.44, 3.53, and 3.41 in fly ash; 3.2, 3.65, and 3.47 in bottom ash; 2.19, 2.62, and 2.46 in pond ash; and 3.1, 3.4, and 3.2 in CCRs, respectively, which are within the range of previous studies (Tables 4). The obtained EF values suggest the investigated CCRs samples are moderately enriched with the U and Th chain radionuclides (Usmani and Kumar 2017). It is observed that our estimated EF values are larger than that for China (Lu et al. 2012), the USA (Coles et al. 1978), Poland (Bem et al. 2002), Spain (Mora et al. 2009), and Greece (Karangelos et al. 2004). However, EF values for Indian bottom ash (Sahu et al. 2014) and Turkey fly ash (Cevik et al. 2008) are considerably higher than the present study (Table 4).

Table 4 Comparison of the normalized enrichment factor (EF) in the samples analyzed from Barapukuria in previously published work

Country	Sample	Enrichment factor			References
		U-238	Ra-226	Th-232	
Bangladesh	FA	3.44	3.53	3.41	This study
	BA	3.20	3.65	3.47	
	PA	2.19	2.62	2.46	
India	FA	–	1.84	1.52	Bhangare et al. 2014
	BA	–	3.50	2.30	
China	FA	–	0.96	0.96	Lu et al. 2012
	BA	–	0.86	0.78	
Australia	FA	–	0.78	0.78	Fardy et al. 1989
Greece	FA	1.20	1.10	1.06	Karangelos et al. 2004
	BA	0.95	0.82	0.99	
Hong Kong	FA	–	1.11	1.04	Tso and Leung 1996
	BA	–	1.07	0.95	
Spain	FA	–	1.48	1.39	Mora et al. 2009
	BA	–	1.36	1.24	

FA fly ash, *BA* bottom ash, *PA* pond ash, *CCRs* coal combustion

During combustion, most of the organic constitutes (OM) in coal matrix oxidizes leading to the enrichment of natural radionuclides in different fractions in CCR matrix compared to the feed coal (Bhangare et al. 2014). Consequently, the inorganic constituents (noncombustible part) containing radionuclides (non-volatile portion of radionuclides) in coals are concentrated in minerals in the remaining residue mass (e.g., CCRs) (Coles et al. 1978; Papastefanou 2010; Hasani et al. 2014), and hence, inorganic fraction controls the radioactivity in CCRs (Cevik et al. 2007; Lauer et al. 2015, 2017). EF represents the apparent enrichment phenomenon due to the loss of the organic substances and volatile constituents in coal matrix during incineration (Flues et al. 2007; Bhangare et al. 2014; Zhou et al. 2014) (Table 4).

The partition ratios, PR (fly ash/bottom ash), of U-238, Ra-226, Th-232, and K-40 are 1.57, 1.40, 1.43, and 1.45, respectively, whereas the respective PR (fly ash/pond ash) are 1.68, 1.43, 1.48, and 1.06, respectively (Table 1). The activity of the radionuclides measured in pond ash is very close to the activity measured in bottom ash in the present study. The inorganically bonded radionuclides are commonly non-volatile or have very low volatility (e.g., Th, a refractory element, Khan et al. 2015) and tend to be retained in bottom ash mineral matter (Bem et al. 2002; Dai et al. 2010; Papastefanou 2010; Bhangare et al. 2011; Li et al. 2012; Hasani et al. 2014). On the contrary, the organically bounded radionuclides are likely to be vaporized in the furnace and are subsequently condense totally or partially on the finest fraction, resulting to a higher content of volatile radionuclides in fly ash than in bottom ash (Menon et al. 2011; Zhou et al. 2012; Megalovasilis et al. 2013; Tang et al. 2013). Additionally, U-238 and Ra-226 show a preference for adsorption on very fine to ultrafine ash fractions (Coles et al. 1978; Papastefanou 2010). The U decay series show different volatility behavior inside the boiler owing to different physicochemical properties of its progeny (Hasani et al. 2014). In contrast, Th exhibits no different partitioning behavior during burning and it mostly occurs in inorganic part both in coals and associated CCRs (Swaine 2014). The U-238, being more soluble in water and having a solubility nature compared to Th-232, is normally expected to leach down from the surface of the disposal mounds into the deeper layers by the percolating rain water and subsurface run-off; however, Th remains adsorbed on the clay minerals at surface (Parial et al. 2016; Sengupta and Agrahari 2017).

The magnitude of the enrichment, partition, and transformation behaviors and fate of natural radionuclides mainly govern by several factors such as the nature (type and rank) of the feed coal, ash yield and mode of occurrence of radionuclides in feed coal, reactions between radioactive elements and minerals, combustion method and environment (temperature, device), ratio between solid phase and gas phase, and precipitation technique (Menon et al. 2011; Hasani et al. 2014; Lauer et al. 2015).

Radiological hazard assessment

The calculated hazard index values due to the activity concentrations of Ra-226, Th-232, and K-40 radionuclides in the bulk/core coal, pulverized coal, fly ash, bottom ash, and pond ash samples from BCM and BTPS appear in Table 5. The average relative contributions of Ra-226, Th-232, and K-40 activity in the Ra_{eq} budget for fly ash are 31.31, 64.88, and 3.80%; for bottom ash 31.71, 64.53, and 3.76%; and for pond ash 31.45, 63.39, and 5.16%, respectively (Fig. 2). The relative radium equivalent activity contributions of radionuclides in the samples are in descending order Th-232 > Ra-226 > K-40, where Th and Ra mutually contribute more than 90% (Fig. 2).

The average Ra_{eq} values are 161.4 for bulk/core coal, 95.6 for pulverized coal, 545 for fly ash, 367.4 for bottom ash, and 378.3 for pond ash (all values are in Bq kg^{-1}) respectively. Thus, the Ra_{eq} in CCRs is exceeded the threshold value (≤ 370 Bq kg^{-1}) (UNSCEAR 2000) (Table 5; Fig. 3a).

The average H_{ex} values are 0.44 for bulk/core coal, 0.26 for pulverized coal, 1.47 for fly ash, 0.99 for bottom ash, and 1.02 for pond ash, and it appears that the H_{ex} in fly ash is also 1.5 times higher than the threshold value (≤ 1) (UNSCEAR 2000) while the other samples are below the prescribed limit (Fig. 3a). The investigated CCRs contain 2 to 3 times more natural radionuclides than pulverized coal which is almost similar with Turhan et al. (2010). Thus, there is significant amount of radiological health risk to the ambient environment, occupational workers, and local inhabitants due to their harmful effects of ionizing radiation exposure staying around the power plant (Papastefanou 2010; Mahur et al. 2013; Laraia 2015; Hower et al. 2016). The radium equivalent activity (Ra_{eq}) and external hazards index (H_{ex}) values are closest to 370 Bq kg^{-1} and unity, respectively. The average

Table 5 Radiogenic hazard indices computed as radium equivalent activity, Ra_{eq} (Bq kg^{-1}); external hazard index, H_{ex}; external absorbed gamma dose rate, D (nGy h^{-1}); annual effective dose rate, E (mSv $year^{-1}$); and excess life time cancer risk, ELCR (Sv^{-1})

Sample	Ra_{eq}	H_{ex}	D	E	ELCR
Bulk and core coal samples from Barapukuria					
RC 1	142.3	0.38	66.2	0.08	2.8×10^{-4}
RC 2	90.6	0.24	40.2	0.05	1.7×10^{-4}
RC 3	49.6	0.13	22.4	0.03	1.0×10^{-4}
RC 4	191.0	0.52	87.5	0.11	3.8×10^{-4}
RC 5	205.4	0.55	94.0	0.12	4.0×10^{-4}
RC 6	216.0	0.58	99.8	0.12	4.3×10^{-4}
RC 7	234.6	0.63	107.3	0.13	4.6×10^{-4}
$RC_{average}$	161.4	0.44	73.9	0.09	3.2×10^{-4}
Min	49.62	0.13	22.4	0.03	1.0×10^{-4}
Max	234.56	0.63	107.3	0.13	4.6×10^{-4}
PC	95.61	0.26	42.7	0.05	1.8×10^{-4}
$Coal_{average}$	153.1	0.41	70.0	0.09	3.0×10^{-4}
Min	49.6	0.13	22.4	0.03	1.0×10^{-4}
Max	234.6	0.63	107.3	0.13	4.6×10^{-4}
CCRs from BTPS					
FA 1	516.2	1.39	231.1	0.28	9.9×10^{-4}
FA 2	573.9	1.55	256.6	0.32	11.0×10^{-4}
$FA_{average}$	545.0	1.47	243.9	0.30	10.5×10^{-4}
BA 1	351.8	0.95	157.6	0.19	6.8×10^{-4}
BA 2	382.9	1.03	171.4	0.21	7.4×10^{-4}
$BA_{average}$	367.4	0.99	164.5	0.2	7.1×10^{-4}
PA	378.3	1.02	169.8	0.21	7.3×10^{-4}
$CCRs_{average}$	446.8	1.21	200.1	0.25	8.6×10^{-4}
Min	351.8	0.95	157.6	0.19	6.8×10^{-4}
Max	573.9	1.55	256.6	0.32	11.0×10^{-4}
$WC_{average}$ [a]	108.70	0.29	51.4	0.06	2.2×10^{-4}
$WFA_{average}$ [b]	360.51	0.97	165.2	0.20	7.1×10^{-4}
$WS_{average}$ [c]	116.8	0.32	55.1	0.07	2.4×10^{-4}
$CC_{average}$ [d]	161.4	0.44	77.8	0.10	3.3×10^{-4}
Threshold limit[e]	≤370	≤1	60	0.07	2.9×10^{-4}

Radiation indices were computed from their respective reported activities following formulas adopted by UNSCEAR 2000

RC raw (bulk/core) coal; *PC* pulverized coal; *FA* fly ash; *BA* bottom ash; *PA* pond ash; *CCRs* coal combustion residues, mean of different ashes; *Min* minimum; *Max* maximum; *WC* world coal; *WS* world soil; *CC* continental crust

[a] UNSCEAR 2010

[b] UNSCEAR 1982

[c] Bowen 1979

[d] Eisenbud and Gesell 1997

[e] UNSCEAR 2000

absorbed dose (D) values are 73.9 for bulk/core coal, 42.7 for pulverized coal, 243.9 for fly ash, 164.5 for bottom ash, and 169.8 for pond ash (all units are in nGy h^{-1}). Thus, the D values for bulk/core coal, fly ash, bottom ash, and pond ash exceed the threshold limit (≤ 60 nGy h^{-1}) by a factor of 1.2, 4.0, 2.7, and 2.8 times, respectively (UNSCEAR 2000). The calculated mean effective doses (E) are 0.9 for bulk/core coal, 0.5 for

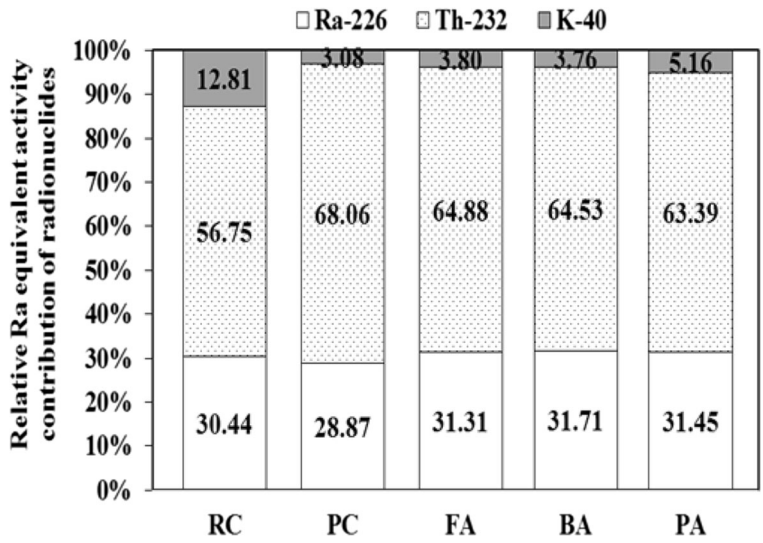

Fig. 2 Relative Ra equivalent activity contribution of radionuclides in coal and ash from Barapukuria. RC, raw (bulk/core) coal; PC, pulverized coal; FA, fly ash; BA, bottom ash; PA, pond ash

pulverized coal, 0.30 for fly ash, 0.20 for bottom ash, and 0.21 for pond ash (all values are in mSv year^{-1} and the permissible limit is 0.07 mSv year^{-1}). The estimated average values of the ELCR are 3.2×10^{-4} for bulk/core coal, 1.8×10^{-4} for pulverized coal, 10.5×10^{-4} for fly ash, 7.1×10^{-4} for bottom ash, and 7.3×10^{-4} for pond ash (all units are in Sv^{-1}) of which the values for fly ash, bottom ash, and pond ash are above the precautionary limit of 2.9×10^{-4} Sv^{-1} prescribed by UNSCEAR (2000) (Fig. 3b). Compared to the results of other studies (Table 2), it can be seen that the results of the present investigation found some indices to be in good agreement with their results while some indices are higher than those recorded in the literature.

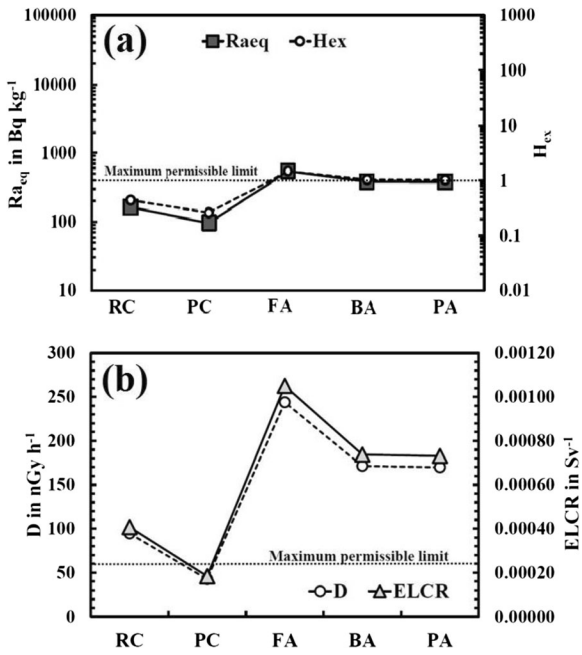

Fig. 3 Hazardous indices: **a** radium equivalent activity, Ra_{eq} (Bq kg^{-1}) and external hazard index, H_{ex}, **b** external absorbed gamma dose rate, D (nGy h^{-1}), and excess life time cancer risk, ELCR (Sv^{-1}), due to Ra-226, Th-232, and K-40 for the investigated coal and ash samples of this study. RC, raw (bulk/core) coal; PC, pulverized coal; FA, fly ash; BA, bottom ash; PA, pond ash

Figure 4 shows radiological hazard parameters from this study compared with literature data and similar results across some countries, including India (Sahu et al. 2014), China (Lu et al. 2012), the USA (Coles et al. 1978), Poland (Bem et al. 2002), Turkey (Cevik et al. 2008), Spain (Mora et al. 2009), and Greece (Karangelos et al. 2004). All radioactive progenies of U-238 and Th-232 parents contained in coals and CCRs emit harmful alpha and/or beta particles followed by gamma rays until their end-up to stable isotopes (Amin et al. 2013; Bhangare et al. 2014; Lauer et al. 2017). However, majority of the emitted such particles cannot come out from the material to the atmosphere due to their low penetration powers. Conversely, most of the gamma rays may easily penetrate the environmental materials (e.g., coals, CCRs) and enter into the local environment. Moreover, radionuclide may easily reach human body (Gupta et al. 2013; Megalovasilis et al. 2013; Hasani et al. 2014; Skoko et al. 2017) may continuously be exposed by gamma radiation and associated harmful health effects (e.g., cell damage or cell death, create cancer) can occur via extended period of exposure

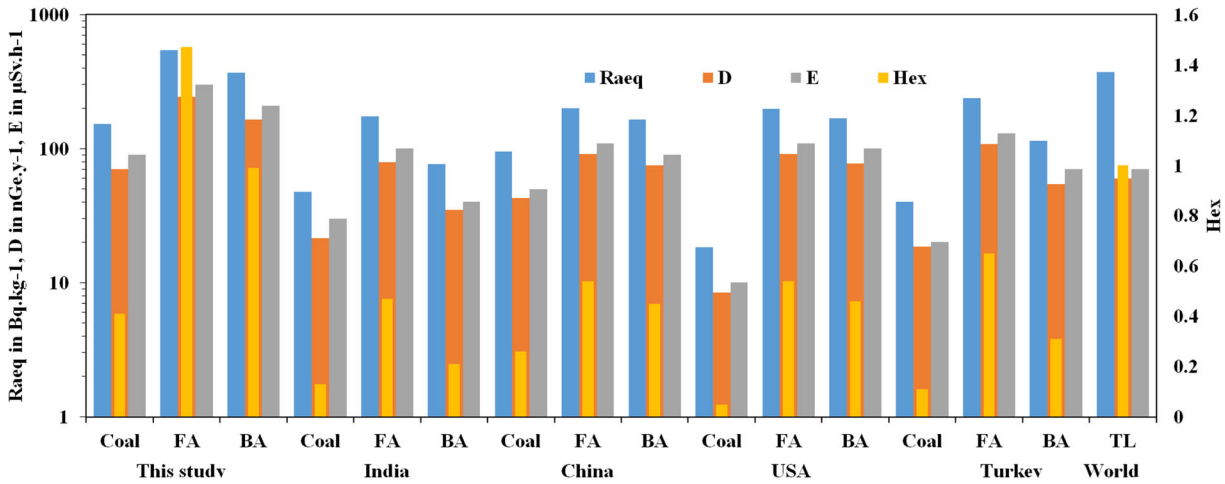

Fig. 4 Evaluated radiological hazard parameters from this study compared with literature data. The data is taken from the following references: Bangladesh (present study), India (Sahu et al. 2014), China (Lu et al. 2012), the USA (Coles et al. 1978), Poland (Bem et al. 2002), Turkey (Cevik et al. 2008), Spain (Mora et al. 2009), and Greece (Karangelos et al. 2004). TL, threshold limit

(Amin et al. 2013; Bhangare et al. 2014; Asaduzzaman et al. 2015; Munawer 2018). Thus, the radiation indices find great significance to understand the health hazards from gamma radiation exposures. For these reasons, radiological hazard parameters or risk factors are considered and evaluated for coals and associated CCRs materials in this research based on the proposed equations provided by Beretka and Mathew (1985), UNSCEAR (2000), and ICRP (1990) to avoid potential radiation hazards to the respiratory organs (Durašević et al. 2014; Kolo et al. 2016).

Nevertheless, it is clear that the activity concentrations and corresponding hazard indices are generally higher in the CCRs and pose a potential radiological risk to the environment, occupational workers, and the entire population around the BTPS. Hence, BTPS generated CCRs have a significant amount of radioactivity leading to higher radiation risk factors which could pose a serious threat to the environment and human health (both staff and public) if the CCRs are not carefully disposed and managed.

Conclusion

Based on the activity concentrations of the radionuclides of the coal and CCRs discovered in this study and on the calculated radiological health hazard indices, we would like to draw the following conclusions:

For coal, the averages (ranges) of activities of U-238, Ra-226, Th-232, and K-40 are found to be 66.5 (32.3 to 103.7), 41.7 (21.8 to 63.5), 62.5 (16.4 to 95.8), and 232.4 (13.9 to 544.0) Bq kg^{-1}, respectively, all exceed the worldwide mean values for coal. The respective values for CCRs are 206.3 (149.3 to 329.5), 140.5 (111.9 to 175.4), 201.7 (157.7 to 263.7), and 232.5 (183.9 to 277.8) Bq kg^{-1}, respectively. The activity concentrations in the CCRs samples in this study are considerably higher than the world soil, Barapukuria soil, and earth crust average values.

The specific activity of U-238, Ra-226, and Th-232 in feed coal is 3.10 to 3.37 times lower than in CCR samples. The respective normalized enrichment factors (with respect to pulverized coal) are 3.1, 3.4, and 3.2 in CCRs, respectively. The radioactivity of fly ash and bottom ash is partitioned and ratio ranges from 1.40 to 1.57. Higher level of radioactivity in CCRs than the world soil and the earth crust average activity indicate that these CCRs are highly contaminated and could pose radiological threat to the local environments.

The recorded averages (ranges) values for radium equivalent activity (Bq kg^{-1}), external hazard index, absorbed gamma dose rate (nGy h^{-1}), annual effective dose rate (mSv $year^{-1}$), and excess lifetime cancer risk (Sv^{-1}) are 153.1 (49.6 to 234.6), 0.41 (0.13 to 0.63), 70.0 (22.4 to 107.3), 0.09 (0.03 to 0.13), 3.0×10^{-4}(1.0×10^{-4} to 4.6×10^{-4}) for coals and 446.8 (351.8 to 573.9), 1.21 (0.95 to 1.55), 200.1 (157.6 to 256.6), 0.25 (0.19 to 0.32), and 8.6×10^{-4} (6.8×10^{-4} to 11.0×10^{-4}) for CCRs, respectively. The average value of ELCR is 2.34 to 3.81 times more than the permissible maximum limit of 2.9×10^{-4} Sv^{-1}. The estimated

various radiation exposure index values indicate a potential risk of ionizing radiation exposure.

This work is the first of its kind carried out relating to the coal industry in Bangladesh and it is hoped that it will encourage the responsible authority to adopt proper management of produced CCRs in order to protect the environment and to prevent radiological health hazards to the human health (both occupational and public) from potential adverse radiological impact living in the vicinity of BTPS.

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