Potential Impact of Bituminous Coal-based Subcritical Thermal Power Plant on the Soil Resources of Barapukuria Area, Dinajpur, Bangladesh

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A Thesis Submitted in Fulfillment of the Requirements for the Degree of Doctor of Philosophy in Sustainable Energy Management
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ABSTRACT

Coal-fly-ash is one of the major byproducts of coal-based power-plant in which ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K radionuclides are drastically enriched compared to those of feed coals. To determine the level of radioactivity concentrations and to estimate the associated radiological risk, coal, combustion residuals (CCRs) and soils from the Barapukuria coal-based thermal power-plant surroundings were analyzed by gamma-ray spectrometer with high-purity germanium (HPGe) detector. Along with the radioactivity concentrations, soil minerals were also studied by X-ray diffractometer to assess the mineralogical source of radionuclides. The results reveal that the mean radioactivity concentrations ($Bq.kg^{-1}$) in feed coal samples are 66.5 ± 24.2, 41.7 ± 18.2, 62.5 ± 26.3 and 232.4 ± 227.2 for ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K, respectively while in CCRs they are 206.3 ± 72.4, 140.5 ± 28.4, 201.7 ± 44.7 and 232.5 ± 43.8, respectively. 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 $nGy.h^{-1}$ for the absorbed gamma dose rate; 0.09 and 0.25 $mSv.y^{-1}$ 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.

Mean radioactivity concentrations (in $Bq.kg^{-1}$) in soil samples for ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K are 102.9 ± 41.4, 63.6 ± 7.4, 103.4 ± 13.9 and 494.2 ± 107.5, respectively which are comparatively higher than the typical world mean value. Elevated levels of radioactivity are likely due to the presence of illite, kaolinite, monazite, rutile and zircon minerals in the soil sample rather than technogenic contributions from the power-plant. Furthermore, mean soil contamination factor (CF) are close to unity and mean pollution load index (PLI) is below unity while the average radium equivalent activity (Ra_{eq} in $Bq.kg^{-1}$), external hazard index (H_{ex}), absorbed gamma dose rate (D in $nGy.h^{-1}$), annual effective dose rate (E in $mSv.y^{-1}$) and excess lifetime cancer risk (ELCR) are 249.5 ± 21.7, 0.67 ± 0.06, 114.2 ± 9.4, 0.20 ± 0.02, 4.9×10^{-4} ± 0.4×10^{-4}, respectively, which are within the permissible limit. Thus, in terms of radioactivity concentrations and associated environmental and radiological indices, the effect of the power-plant on soil is insignificant.

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF ABBREVIATIONS

Abbreviation	Term
APCD	Air Pollution Control Devices
BA	Bottom Ash
BCM	Barapukuria Coal Mine
BS	Barapukuria Soil
BTPS	Barapukuria Coal-based Thermal Power Station
CC	Continental Crust
CCRs	Coal Combustion Residues
CF	Contamination Factor
CPS	Counts Per Second
CTPs	Coal-based Thermal Power Plants
EDXRF	Energy Dispersive X-Ray Fluorescence Spectrometer
EF	Enrichment Factor
ESP	Electrostatic Precipitator
FA	Fly Ash
FC	Feed Coal
GSB	Geological Survey of Bangladesh
HPGe	High Purity Germanium
IDW	Inverse Distance Weighting
IEA	International Energy Agency
INAA	Instrumental Neutron Activation Analysis
n	Sample size
NORMs	Naturally Occurring Radioactive Materials
PA	Pond Ash
PC	Pulverized Coal
PM	Particulate Matter
PSMP	Power System Master Plan
UCC	Upper Continental Crust
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation
XRD	X-ray diffraction
yrs	Years
RC	Raw coal

LIST OF PUBLICATIONS

The thesis is based on the papers listed below, which are referred to in the text by the indicated Roman numerals.

1. Published ISI Journal Paper

1) **Habib, M. A**., et al., 2019. Assessment of natural radioactivity in coals and coal combustion residues from a coal-based thermoelectric plant in Bangladesh: Implications for radiological health hazards. Environmental Monitoring & Assessment, 191 (1), p.27. **(ISI, IF: 1.804)**.
2) **Habib, M. A**., et al., 2018. Distribution of naturally occurring radionuclides in soil around a coal-based power plant and their potential radiological risk assessment. Radiochimica Acta (in press) **(ISI, IF: 1.202)**.

Author contribution:

Paper I: The author contributed significantly to the planning and conduct of the experiments and made additional contributions to the quantification and evaluation of the data and the writing of the manuscript.

Paper II: The author was heavily involved in the planning and conduct of the experiments and the interpretation of their results and made a major contribution to the writing of the manuscript.

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CHAPTER 1

INTRODUCTION

1.1 Background of the research

Radiation and the associated potential threat to human health has become a matter of enormous public concern throughout the world, even though it is an inevitable aspect of environmental samples (e.g., soil) (Finkelman and Greb, 2008; Finkelman and Stracher, 2011; Siegel and Bryan, 2014; Laraia, 2015). The sources of continuous radiation exposure are mainly terrestrial, cosmogenic, anthropogenic (e.g., technogenic, nuclear test) (Hu et al., 2010; Atwood, 2013; El-Mekawy et al., 2015). Inherently, coals, coal combustion residuals (CCRs) and soils commonly contains naturally occurring radioactive materials (NORMs) pollutants such as U and Th actinide series and their progenies, along with other radionuclide, e.g., ^{40}K (Arbuzov et al., 2011; Orem and Finkelman, 2014; Chen et al., 2011, 2017; Habib et al., 2018 in press, 2019 and the reference cited there-in). The level of gamma radiation is directly related to the activity concentrations of radionuclides (Siegel and Bryan, 2014). Considering the natural abundances of common radionuclides (here, ^{226}Ra, ^{232}Th, ^{238}U and ^{40}K), assessment of radiological health risk due to environmental radioactivity is more useful than simply studying the volumes of potentially hazardous wastes (Vuković et al., 1996; Dragović et al., 2008, 2013). Hence, obtaining the information regarding the level of radioactivity concentrations of natural radionuclides in coal and coal combustion residuals (CCRs, a collective term for fly- and bottom-ash, pond ash used in present study) is essential (Sajwan et al., 2011; Lauer at al., 2017) for evaluating the radiological risks. Their presence (radionuclides) modifies the composition of geomaterials and elevates the level of natural background radiation through alpha and beta energy, gamma-rays and spontaneous gaseous radon (^{222}Rn) release and the total dose rate to which all living things are exposed (Amin et al., 2013).

Coal is a very complex sedimentary rock and is the main natural solid fuel source for power generation (Depoi et al., 2008). It is the most ubiquitous, and readily combustible solid fuel containing more than 70% carbonaceous organic constituents by volume (e.g., Mick, 2010; Ozden et al., 2018; Zaman et al., 2018). Due to price stability, cost efficiency and it's use intensively used in many developed and developing countries including Bangladesh is currently popular and indispensable (e.g., Mishra, 2004; Bhangare et al., 2014). Thus, coal fuel is become as the primary nonrenewable energy contributing more than 40% of the world electricity (IEA, 2017) and this is expected to be increased gradually (Amin et al., 2013). Moreover, coal contributes about 28% of the world energy supply mix (IEA, 2017). It is a

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NORMs consist of materials, usually coal combustion residues enriched with radioactive elements found in the environment, such as ^{238}U, ^{232}Th and ^{40}K and any of their decay products, such as ^{226}Ra.
Radionuclide: An atom with an unstable nucleus which, to become more stable, emits energy in the form of rays or high-speed particles.
Activity concentration (Bq.kg^{-1}): Radionuclide activity per unit dry mass of material.

mixture of (in)combustible compounds, an organic fraction (macerals) and an inorganic fraction (minerals) (Goodarzi, 2006; Orem and Finkelman, 2014; Habib et al., 2018 in press, 2019) which was usually stratified, and was originated from the accumulation of plant debris through physical and chemical alteration, burial and compaction of partially decomposed vegetation over the geologic time; the accumulation of plant debris and their coalification (Orem and Finkelman, 2014). Geologically, coal carries minute amounts of NORMs (e.g., ^{226}Ra, ^{232}Th, ^{238}U and ^{40}K) along with small amounts of other trace metals as impurities and PAHs (Khandekar et al., 1999; Bragatoet al., 2012; Verma et al., 2015; Hower et al., 2016; Sengupta and Agrahari, 2017).

CCRs are, the noncombustible major residuals generated by CTPs, considered as one of the major non-nuclear sources of technogenic radionuclides pollutants, which are of most concern due to their radiotoxicity (Coles et al., 1978; Mishra, 2004; Papaefthymiou et al., 2007, 2013; Hower et al., 2016). Previous study reported that ~ 5-20% of CCRs of the mass of feed coal are mainly produced due to incineration in the CTPs which consist of 85-95% fly ash and 5-15% bottom ash along with few slag (Shaheen et al., 2014; Yao et al., 2015). Fly ash is a light form of coal ash residues that floats into the exhaust chimney; bottom ash is the heavier portion of ash that settles on the ground in the boiler; slag is the melted materials; and pond ash is the mixture of different type of ashes dumped to the disposal pond (Ahmaruzzaman, 2010; Hower, 2012; Bartoňová, 2015; Hower et al., 2017a). It is geochemically complex and highly enriched with extremely heterogeneous constituents, composed of (in)organic pollutants (Liu et al., 2008; Verma et al., 2015; Hower et al., 2017a, b). It contains potentially hazardous toxic elements, radiotoxic radionuclides (Turhan et al., 2018; Hower et al., 2016; Sahu et al., 2009, 2014, 2017, Wang et al., 2016a, b), unburned carbon, rock-fragment, and amorphous glass, nano-particles/minerals (Hower, 2012; Dai et al., 2014; Zhao et al., 2017; Jambhulkar et al., 2018). CCRs are originated mostly from the inorganic constituents of feed-coal (Koukouzas et al., 2006; Islam et al., 2011; Lu et al., 2012; Hower et al., 2012, 2017a, b; Fungaro et al., 2013). The multistage processes of combustion causing enormous changes of (in)organic constituents in feed-coal and simultaneously produce large volume of CCRs with physical-chemical-mineralogical transformation, elemental speciation, and isotopic fractionation of radionuclides in CCRs (Meij and Te Winkel, 2009; Sajwan et al., 2011; Michalik et al., 2013; Ozden et al., 2018). It is identified as a major potential anthropogenic source of radionuclides releasing to the environment (Mahur et al., 2013). The concentration of most radionuclides in CCRs are enriched with a several order of magnitude due to combustion by reducing the coal volume about 85% (Basu et al., 2009; Peppas et al., 2010; Hower et al., 2016; Turhan et al., 2018).

Gamma ray: The electromagnetic radiation, consists of sharp lines of discrete wavelengths, arising from the radioactive decay of atomic nuclei. Due to very short wavelength γ-rays are highly energetic and hence possess a very high penetrating property.

These are consistently disposed to the ash ponds (disposal site) with a high environmental risk as it is highly enriched in radiotoxic radionuclides and environmentally sensitive heavy metals (Mishra, 2004; Querol et al., 2011; Bhangare et al., 2014). Pollutants are released from CTPs in different form and stages of the processing system i.e. before (coal storage, processing), during combustion (flue gas) and after combustion processes and CCRs deposition, transportation and utilization to the ambient environments as in the form of gaseous phase, solid and liquid discharge (Dai et al., 2007, 2008, 2012, 2014; Karamanis et al., 2009; Mahur et al., 2013; Dragović et al., 2013; Saikia et al., 2014, 2015, 2016).

CTPs arrest the most of coarse fly ash, however, approximately 1 to 3% finest portion of the total fly ash having radionuclides escaped to the atmosphere and biosphere despite the use of filtration systems and consequently dispersed over a wide range of distance due to the atmospheric convection (Papastefanou, 2010; Charro et al., 2013; Dragović et al., 2013; Tripathi et al., 2014). Most of which henceforth fall to earth and find the way to the soil and surface water leading to contaminate through mobilization, migration processes, and finally reach to human body through inhalation and ingestion and exposure to external radiation (radionuclides in CCRs → atmosphere/ash pond leachates → soil to crop/water to fish → human body) (Figure 1.1) (e.g., Querol et al., 2011; Dai et al., 2012; Ruhl et al., 2012; Al-Masri et al., 2015; Wang et al., 2016a; Sahu et al., 2017; Skoko et al., 2017; Finkelman et al., 2018a). It has been reported that the pollutants like Ra are potential hazards to the environment even at low levels (Blissett and Rowson, 2012; Skousen et al., 2013; Yao et al., 2015; Medunić et al., 2016; Kalia et al., 2017). These can even reach the sub-soil and natural drainage systems and contaminate the groundwater with radionuclides (Belivermis et al., 2010; Navas et al., 2011; Nenadović et al., 2012; Charro et al., 2013; Dragović et al., 2013; Noli and Tsamos, 2016; Islam et al., 2018). It may finally lead to the harmful effects on human health due to their higher radiation level and pose acute and chronic diseases to public health (e.g., cell damage, lung and bone cancer, respiratory illnesses) (e.g., Fergusson, 1990; Borm, 1997; Dai et al., 2008, 2012; Silva et al., 2009, 2011; Oliveira et al., 2014; Bukhari et al., 2015; Munawer, 2018; Campaner et al., 2018; and the references cited there-in).

Currently, Bangladesh suffers from a shortage of energy and many comprehensive initiatives have been taken to solve that deficiency 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, 8 coal power projects are being constructed in the country with capacity of 6543 MW (Ahamed et al., 2016; Zaman et al., 2018).

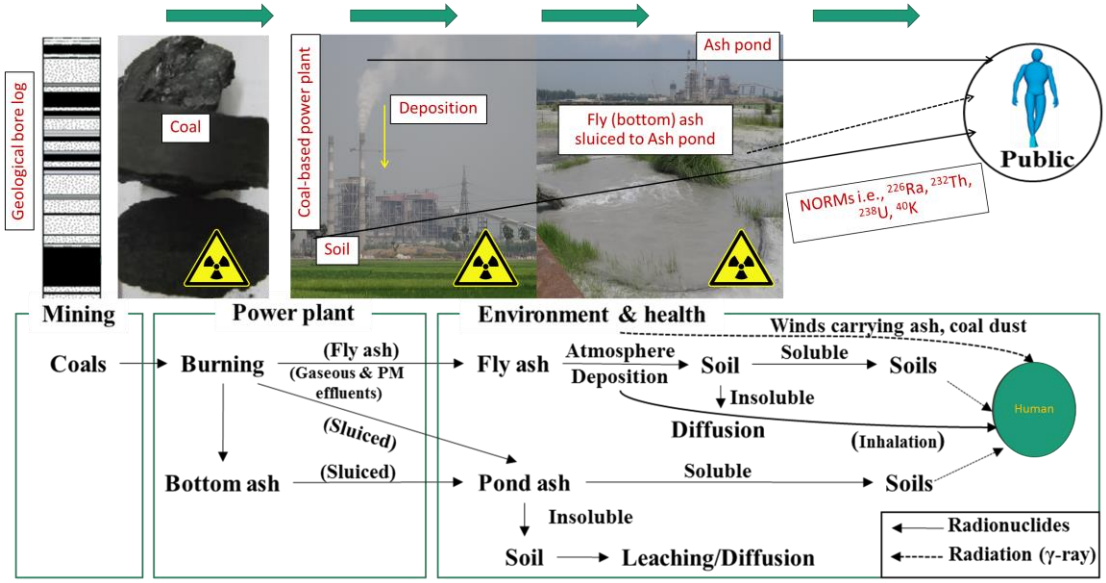

Figure 1. 1 The pathways of radionuclides migration through the different media from a coal power plant to soil.

Therefore, a significant portion of the total power production in coming years is expected to be derived from coal energy (Islam and Khan, 2017). The currently installed power generation capacity of the country is more than 12780 MW which is planned to increase up to 39000 MW by 2030, of which about 50% would be derived from local and imported coal (PSMP, 2010; Islam and Khan, 2017; Zaman et al., 2018). Presently, the first and only Barapukuria coal-based power station, BTPS (subcritical) with a capacity of 250 MW (2 X 125 MW), contributes only 3.75% of the total power while the largest portion of electricity (about 69.7%) is exclusively generated from local natural gas, which is being rapidly depleted (PSMP, 2010; Zaman et al., 2018). The BTPS has been operated since 2005. A third unit with 275 MW capacity has been started in 2017. According to the present development data, Bangladesh has five coal fields having reserve of about 3.33 billion tons (Bt) of high-volatile bituminous B ranked Permian Gondwana coal (Bostick et al., 1991; Norman, 1992; Bakr et al., 1996; Akhtar and Kosanke, 2000, Islam, 2002). The only Barapukuria coal basin (reserve 377 million tons, Mt) is in active commercial operation (Islam, 2008, 2009; Islam and Hayashi, 2008; Farhaduzzaman et al., 2012; Farhaduzzaman, 2013; Islam and Khan, 2017). About one million tons of coal is extracted from Barapukuria coalmine annually, of which 65% is combusted for power generation in the BTPS (Howladar, 2013; Howladar and Islam, 2016). Approximately 4000 tons (t) of feed coal is burnt per day in the boiler and simultaneously is generated nearly 480 t ash (Zaman et al., 2018). Every year about 0.08 Mt CCRs are produced, of which, 80% is estimated to be fly ash and the rest is bottom ash (Hashan et al., 2003; Howladar and Islam, 2016).

1.2 Statement of the problem

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 NORMs in coals, CCRs, and soils. Several investigations of natural radionuclide in coals, CCRs and soils have become very interesting topic and received great concern to the population around world and their unavoidable adverse effects on humans and environmental concerns were extensively studied (e.g., Frontasyeva et al., 2001; Peppas et al., 2010; Lu et al., 2012; Durašević et al., 2014; Lauer et al., 2017; Pak et al., 2018). Nowadays many researchers have focused their attention on soil contamination due to the coal burning. They have found non-significant radioactive influence on the soil environment (e.g., Rosner et al., 1984; Charro and Pena, 2013; Charro et al., 2013; Papaefthymiou et al., 2013; Habib et al., 2018 in press, and other reference cited there-in), whereas other studies have illustrated that an evident increment of natural radionuclides concentration in soil around CTPs (e.g., Flues et al., 2002; Papp et at., 2002; Bem et al., 2002; Mandal and Sengupta, 2006; Dai et al., 2007; Gür and Yaprak, 2010; Lu et al., 2012, 2013; Amin et al., 2013; Ćujić et al., 2015; Liu et al., 2015; Parial et al., 2016; Gören et al., 2017 and among others), largely due to the unusually high concentration of ^{238}U (and ^{226}Ra) in feed coal and associated CCRs. Hence there is a research debate among the international scientists and researchers. However, there is still an uncertainty to how much the radiological influence and radioactivity impact on human health due to the operation of coal-burning plant. Thus, it is necessary to determine level of the natural radioactivity and its influence on environment and human health around the CTP. Studies on CCRs from Barapukuria in terms of environment and human health potential risk of radionuclide issues are very scarce in the literature so far. Several studies on soil radionuclides found the radioactivity in soil is higher closer to the CTP within 1 km, and it becomes lessen to far (Dai et al., 2007). Mishra (2004) reported that the ash ponds are the source of highest radiation dose.

To have better understanding of the pollution levels and hazard risk to the human health and the environment were intensively studied and reported in literature considering following parameters and observations such as organic pollutants (e.g., polycyclic aromatic hydrocarbon), toxic heavy metals, rare earth elements and radioactive elements, nanominerals/particles (e.g., nano-quartz, anatase, pyrite) etc. in the coals, CCRs, soils, water, air, river sediments, agricultural plants, human body specimen samples from CTPs surrounding area employing different approaches and modes of advanced techniques such as gamma spectrometry with HPGe detector, ICP-OES, ICPMS, EDXRF, AAS, INAA, ore, optical and scanning microscopy, XRD (Taylor et al., 1998; Boyd, 2004; Dai et al., 2008, 2012; Finkelman

and Greb, 2008; Tian et al., 2008, 2013; Huggins and Goodarzi, 2009; Ribeiro et al., 2010, 2014, 2016; Weng et al., 2013; Oliveira et al., 2014; Saini et al., 2016; Dutta et al., 2017; Kalia et al., 2017; Rabha et al., 2018). These approach and techniques are commonly used around the world for environmental studies.

Previously several studies on the Permian Gondwana mined coal from Barapukuria coalfield 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, 2013; Hossain et al., 2014), and geochemical properties (Podder et al., 2004; Haider et al., 2011; Islam et al., 2011) applying EDXRF, LIBS and INAA techniques and CCRs characteristics and its potential uses (Howladar and Islam, 2016) have been carried out. Besides, several studies on Barapukuria soils suggested that it was intensively contaminated with trace elements released from coal matrix due to the coal mining activities (Bhuiyan et al., 2010a, b; Halim et al., 2015; Hossain et al., 2015; Zakir et al., 2017) employing EDXRF and AAS techniques for environmental studies. Despite the long and extensive history of previous studies on radionuclides in different parts of the world, the levels of radioactivity and radiation doses in coal and associated CCRs from BTPS and soils around it have not hitherto been investigated. However, to the best of our knowledge, there has been no systematic evaluation of the radiological characterization employing gamma spectrometry with HPGe detector of coal, CCRs and soils from Barapukuria to address environmental and health consequences. Hence, it is most important to give special attention to take care environment and protect occupational workers and local populations from adverse additional radiation exposure and monitor the trend of natural background composition and to further future research as reference work. Moreover, with the rapid growing a number of CTPs in the country, the radiation exposure from coal burning has become a great concern for environment. 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.

1.3 Research questions

This study is designed to provide answer to the following research questions:

- What are radiological characteristics of feed coals, and associated CCRs (e.g., fly ash, bottom ash, and pond ash)?
- What are radiological characteristics of soils around of BTPS?

- Is there any potential radiological threat to the population (both occupational and local residents) and have any adverse impact on the soil's quality due to coal burning in the BTPS?

1.4 Objectives of the research

The primary objectives of the present study are to determine radiological characteristics of coal, CCR and soil samples in order to elucidate their possible risks of exposure to the population and potential threat to the surrounding environments during mining, processing, conversion, combustion, and CCRs storage.

Thus, the specific objectives of this research are:

1. To determine the radiological characteristics of feed coals and associated CCRs from BTP.
2. To detect the activity concentrations of NORMs in soils from Barapukuria surroundings in order to assess any influence on the soil background radioactivity level of BTP operation.
3. To estimate the radiological hazard indices due to radioactivity from concentrations of radionuclides in the samples measured in order to evaluate any hazards risk for the immediate inhabitants (both public and occupational workers).

1.5 Research limitation

There is some limitation in the present study. The current research deals with the spatial variability of specific activity of NORMs in soils up to a depth of 10 cm within 9 km^2 merely around BTPS as well as feed coals, fly ash, bottom ash and pond ash samples. However, this study did not cover the temporal and vertical variation of activity concentration in soils and not consider radioactivity in (surface)groundwater, agricultural plants/crops, leachate/effluents and sediments. Encountered complications are to find for necessary background information and others essential data as this research is performed for the first time in the country in the coal power plant industry in the subject of environmental impacts and human health effect considering radiological characteristics.

Besides this introductory chapter, the thesis is structured of the following chapters:

Chapter 2 Materials and methods followed by Chapter 3 Results and discussion, Chapter 4 Conclusions.

CHAPTER 2

MATERIALS AND METHODOLOGY

2.1 Research framework

After project finalization, the research design is prepared for the coal, fly ash, bottom ash, pond ash and soil samples from Barapukuria coal industrial area, Bangladesh.

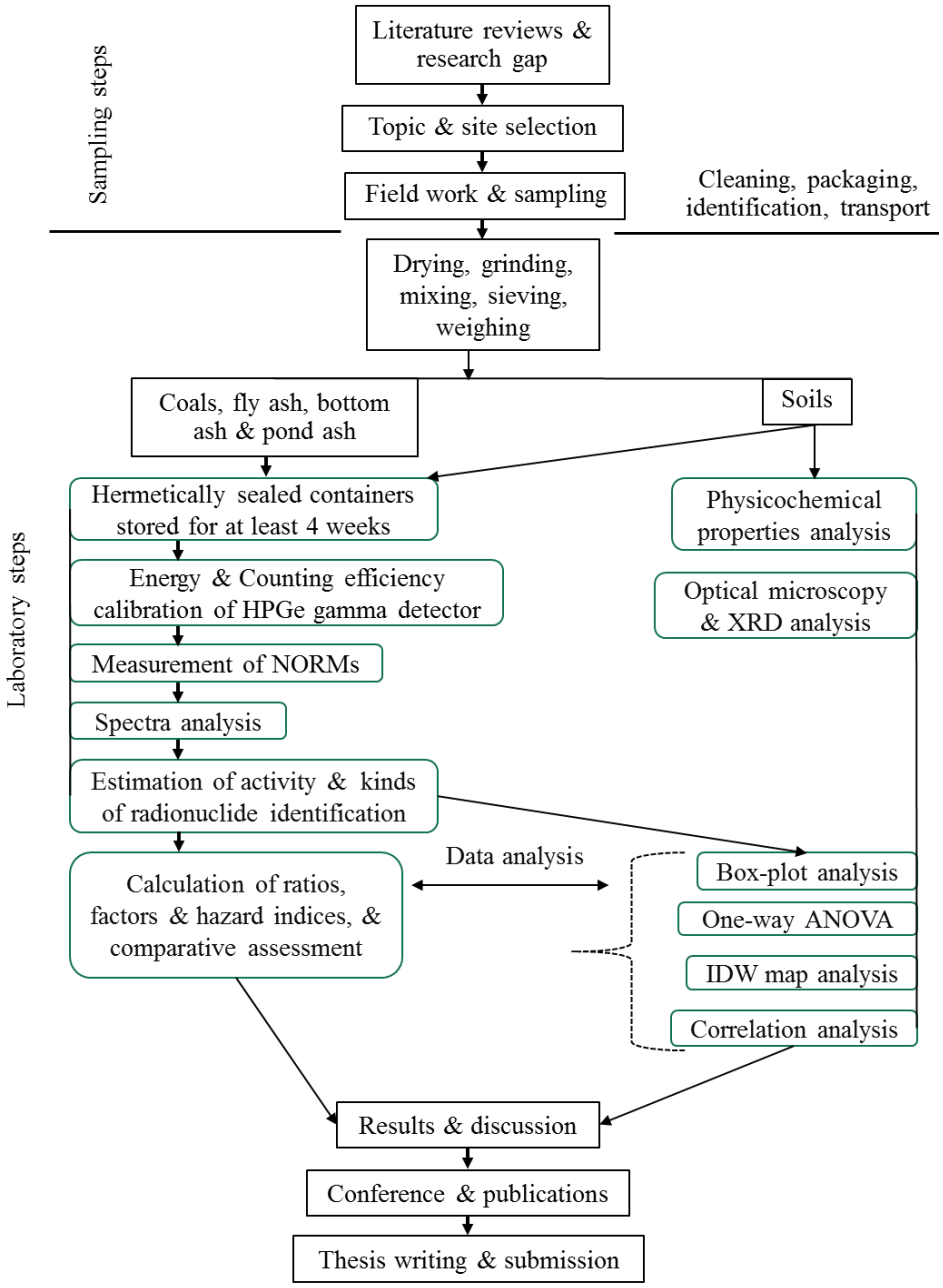

Figure 2. 1 A simplified methodological flow chart and outlines of the present research project.

The whole project is divided mainly into two parts including field work (sampling steps) and laboratory steps. End of the processes of sample collection and beginning of the experimental processes were started, and then the study concentrates for data analysis, publications and finally thesis preparation. Schematics of the evolution of the sample characteristics from the sampling site to the test laboratory (Figure 2.1).

2.2 Study area

BTPS and BCM are situated in a humid subtropical region in an agriculture dominant farming area with double/triple cropped in the northwestern part of Bangladesh (Figure 2.2). The population density is 823 peoples per km^2. The area is situated at the northern fringe of a Pleistocene terrace named the level Barind tract (about 30 m above from the mean sea level) and in humid subtropical region in alluvial-fluvial floodplain system. The mean annual rainfall is 1,800 to 2,000 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 north-east (18%). Wind speeds rarely exceed 8 $m.s^{-1}$. The major features of the BTPS and BCM are appeared in table C1 (Appendix C). Properties of the feed coals from Barapukuria, Bangladesh are presented in table C2 (Appendix C).

Tectonically, the area (Barapukuria coal basin) lies within the Rangpur Saddle and are surrounded by Himalayan Foredeep to the north, Bogra Shelf to the south, Indian Shield on the west and Shillong Massif on the east of Bengal basin (Bakr et al., 1996; Islam, 2008, 2009; Farhaduzzaman, 2013) (Figure 2.2). It is blanketed mainly with unconsolidated Holocene Tista Alluvial fan sediments and Pleistocene Level Barind clay, which were developed under fluvial-alluvial and rapidly prograding deltaic condition. The Pleistocene sediments is underlined by the Plio-Pleistocene Dupi Tila formation (Farhaduzzaman et al., 2012). Based on lithology, the sediments in the Barapukuria basin have been divided into four lithostratigraphic groups, namely Gondwana Group, DupiTila Formation, Barind Clay Formation, and Alluvium having the geological age presumably of Permian, Pliocene, Plio-Pleistocene and Holocene respectively (Islam et al., 2008). The area is drained by a number of rivers like Atrai, little Jamuna, Karatoya, Banglai, Jabuneswari, Kala, Kharkhari, Tillay, Chirnai etc. (distributaries of river Tista), flowing from north to south.

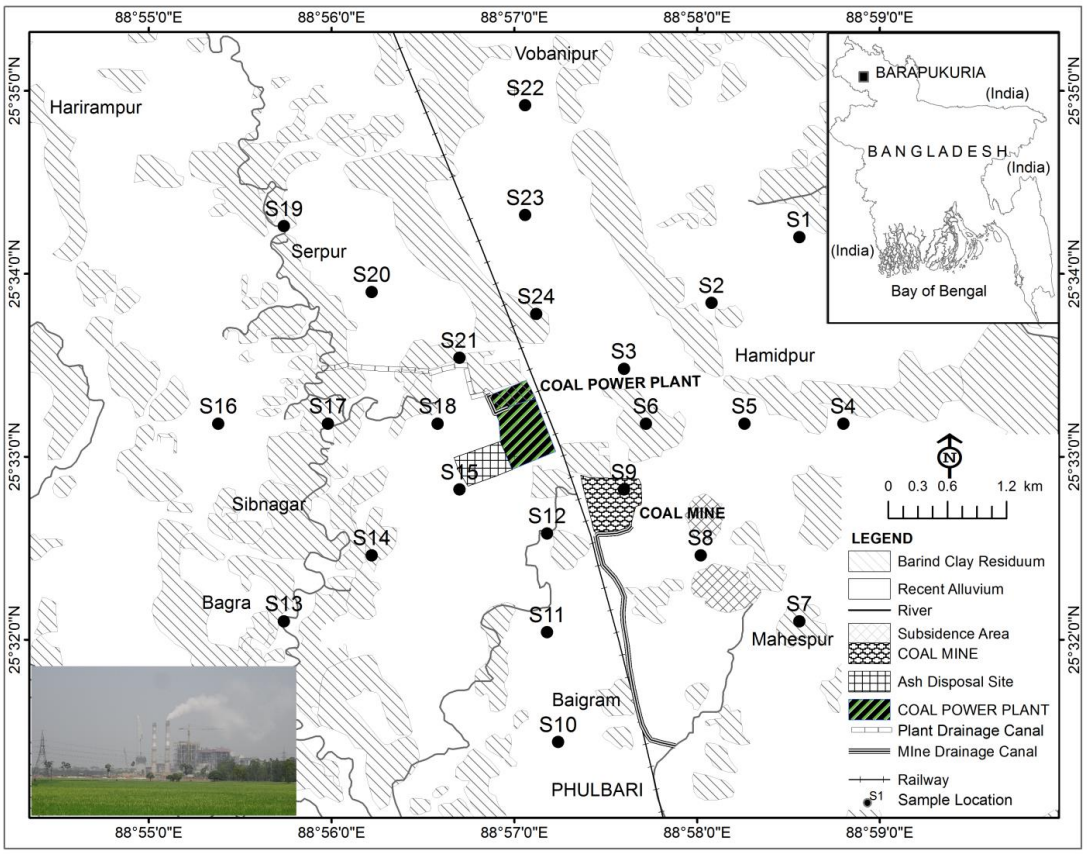

Figure 2. 2 Map showing sampling sites where the soil samples were taken and BTPS area.

2.3 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 (hour), however, the weight difference between pre and post-oven drying is negligible for both coal, ash samples, except for bottom ash samples. It is probably

due to the coal and fly ash samples naturally contain very low amount of moisture and is furthermore dried by air-drying in the winter season with very low humidity.

A total number of 24 soil samples from the vicinity of the BTPS were collected carefully, up to 10 cm (centimeter) depth. Each site was logged by a global positioning system (Figure 2.2) during the dry month of April, 2017. The samples were taken following systematic random sampling protocol, under 'dry soil' conditions, using a stainless-steel cylinder sampler and a plastic scoop. In addition, natural background soil samples (n = 3) (undisturbed soil from similar soil type at about 15 km far from the power-plant) were also collected. The sites were selected based on the morphological features of the terrain, topography, soil type, land use pattern, vegetation, the possible natural radionuclides contributions from various sources in addition to the expected atmospheric diffusion and ash disposal from the BTPS, prevailing wind-direction, surface water flow direction, accessibility also being taken into account. The samples were well mixed after removing extraneous materials such as roots, pebbles and plant materials and other impurities, were instantly stored in airtight clean zip-loc-polyethylene bags, labeled properly and transferred to the laboratory and kept at 4 °C until subsequent analysis. The samples were grounded into powder and homogenized, weight, and dried to remove the moisture content in a temperature-controlled furnace at 105 °C until constant weight prepared for analysis (Charro and Peña, 2012; Charro et al., 2013). Proper care was taken to avoid the cross contamination during sampling, sample preparation and measurement.

2.4 Measurement of soil's physicochemical properties

The soil pH and organic carbon (OC) of the soil samples were determined in distilled water in a solid–liquid (S/L) ratio of 1: 2.5 $ml.g^{-1}$ by using pH meter and dichromate digestion based on Walkley–Black method, respectively (Jackson and Barak, 1985). The soil organic matter (SOM) contents were estimated from OC content values multiplying by a factor of 1.724. Soil texture was carried out using traditional pipette method.

2.5 Measurement of NORMs in coals, CCRs and soil samples

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 and then 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 (^{238}U and ^{232}Th 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 in press, 2019).

The bulk/core coal, pulverized coal, fly ash, bottom ash, and pond ash samples were investigated for the activity measurements of the natural radionuclides ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K 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 was a cover on the detector and bottom of U8 vessel between sample and detector. Detection efficiency calibration of the gamma-ray spectrometer was conducted using the set of standard sources (MX033U8PP) those 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 (millimeter) to consider the geometry of the samples. In principle, the radioactivity measured by 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 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 those are important in NORMs 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 (gram) in 1.83 days, one soil sample about 10 g in 4.95 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):

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

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

where, A is the activity (Bq, becquerel); 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 ^{232}Th, ^{238}U, ^{235}U and ^{226}Ra, were estimated based on the activity concentration of gamma-rays of their progenies in the samples, with the exception of ^{40}K, which can be measured directly. The activity concentrations of ^{238}U and ^{232}Th were determined indirectly by analyzing the full-energy peaks emitted by their progenies. In the ^{232}Th decay series, ^{228}Ac, ^{208}Tl, ^{212}Pb, ^{212}Bi were used to estimate the ^{232}Th. In the ^{238}U decay series, ^{214}Pb and ^{214}Bi were employed to estimate the ^{226}Ra (L'Annunziata, 2003). The activity of ^{226}Ra was estimated from the average value of the activity of ^{214}Pb and ^{214}Bi on four peaks. In the ^{235}U decay series, ^{235}U, which emits gamma-ray at 185 keV was detected with an overlapping emission at 186 keV from ^{226}Ra. The activity of ^{238}U was calculated based on the natural abundance ratio of ^{235}U and ^{238}U. This radiochemical analysis technique is described elsewhere (Habib et al., 2018 in press, 2019).

2.6 Minerals in soil samples

2.6.1 Optical microscopy

Dried bulk soil samples were grounded, homogenized, coned and quartered to attain representative samples for slide preparation. Firstly, some Canada balsam were placed on a glass slide and kept it on the hitter. When it was warmed and transferred into solution, and then samples were placed on the Canada balsam solution. Subsequently a glass cover was placed on this solution and pressed the cover until the extra Canada balsam and bubble removed out. After cooling the slide, it was cleaned using xylene and cotton and ready for microscopic study (Mange and Maurer, 1992). The slides were studied at the Geological Survey of Bangladesh under optical microscope with transmitted light (plane and cross both view) (ZEISS Axio Scope.A1, Germany).

2.6.2 X-ray diffractometer (XRD)

The samples were dried on a hot plate then grinded in a silicon nitride ball-mill, homogenized, coned and quartered to attain representative samples. The selected quarter samples were prepared as powder mounts approximately 1 g and packed into a steel cavity mount suitable for insertion into the X-ray Diffractometer (Habib et al., 2019). The samples were compacted on the sample holder to obtain a uniform surface, required for this technique.

Activity (Bq): The number of radioactive transformations of energy per second that occur in a particular radionuclide.

X-ray diffraction patterns were recorded using a diffractometer with Cu-Kα radiation and with a scan range (2θ) of 2–90º, step size (2θ) of 0.05º and counting time of 1s per step. Machine Settings: Type of Radiation- CuKα; Filter – Nickel; Volt- 40 Kv and Amperes- 30 mA; Diffraction Speed- 3° per min (X-Pert MPD, Philips, the Netherlands).

2.7 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 (Usmani and Kumar, 2017).

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

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

where, $_X$ denotes the radionuclides, and A_x is the corresponding specific activity ($Bq.kg^{-1}$). The ^{40}K is used as a radio tracer since its concentration remains constant in the samples (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 3. 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) (Usmani and Kumar, 2017).

In order to determine the degree of contamination due to operation of BTPS, the Contamination factor (CF), Pollution load index (PLI) were calculated. The CF is the ratio obtained by dividing the activity concentration of each radionuclides by the natural background activity concentration in soil. Based on their intensity, CFs may be classified into four categories: $C^i_f < 1$, low; $1 \leq C^i_f \leq 3$, moderate; $3 \leq C^i_f \leq 6$, considerable; and $C^i_f \leq 6$, very strong contamination. For all soil samples, PLI were computed as the n^{th} root of the product of the n

CFs (n is the total number of contamination factors considered). Unity value of PLI suggest the baseline level of contaminants while more than unity referring to the gradual degradation of soil health (Habib et al., 2019).

2.8 Estimation of radiological hazards in coals and associated CCRs, and soils

Inherently, the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in the environmental substances (e.g., soil, coal, fly ash) are not uniform (Rachkova et al., 2010). 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 ^{226}Ra, ^{232}Th, and ^{40}K (Durašević et al., 2014). The Ra_{eq} ($Bq.kg^{-1}$) was calculated according to the following equation (4):

Radium equivalent activity, $Ra_{eq} = C_{Ra} + 1.43\ C_{Th} + 0.077\ C_K \leq 370$.....(4)

where, C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K 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; Amin et al., 2013). This index can be used to 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 the following equation (5):

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

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

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

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

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

Absorbed dose (Gy, grays): The amount of energy deposited by radiation in a unit mass of material, e.g., tissue.
Equivalent dose (Sv, sieverts): The absorbed dose multiplied by a radiation factor that takes into account the way different types of radiation cause biological harm in a tissue or organ.
Effective dose (Sv): The equivalent dose multiplied by organ factors that take into account the susceptibility to harm of different tissues and organs.

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

Excess lifetime cancer risk, ELCR = E × ALT × RF……...….(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.

2.9 Data analysis

For all gathered samples, basic descriptive statistical analysis and Pearson correlation matrix was performed using SPSS version 20. Analysis of variance (ANOVA) was also carried out to specify the existence or absence of significant differences between groups or more groups of observed parameters. The Inverse Distance Weighting (IDW) technique was employed to interpolate the value of a variable at unmeasured sites from observations of its values at nearby locations applying ArcGIS 10.3 (Habib et al., 2018 in press).

CHAPTER 3

RESULTS AND DISCUSSION

The part (3.1) has been published in Environmental Monitoring & Assessment Journal entitled "Assessment of natural radioactivity in coals and CCRs from a coal-based thermoelectric plant in Bangladesh: Implications for radiological health hazards" (Appendix A).

The part (3.2) has been published in Radiochimica Acta Journal entitled "Distribution of naturally occurring radionuclides in soil around a CTP and their potential radiological risk assessment" (Appendix B).

3.1 Radiological characteristics of coals and CCRs from Barapukuria (Paper I)

3.1.1 Activity concentrations of radionuclides in coal and associated CCRs

The activity concentrations of the ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K radionuclides detected in bulk/core coal and associated CCRs sampled are presented in Table 3. 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 ^{238}U vary from 32.3 to 103.7 with a mean value of 69.6 ± 24.3 ($Bq.kg^{-1}$). The mean activity concentrations of ^{226}Ra, range from 21.8 to 63.5 with an average value of 43.7 ± 18.7 ($Bq.kg^{-1}$), and for ^{232}Th the mean activity concentrations range from 16.4 to 95.8 with a mean value of 65.0 ± 27.4 ($Bq.kg^{-1}$). For ^{40}K, 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 ^{40}K, 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 3. 1). Noticeably the radioactivity of ^{40}K in most of our samples is much lower than the world average values. However, in pulverized coal, the average activity level of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K 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 ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K 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 3. 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 (Hasani et al., 2014; Chen et al., 2011, 2017) and the greater fraction of that U accumulates in coals during the initial stages of the coalification process(es) and subsequent burial stage(s) (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 (Bustin et al., 1985; Suárez-Ruiz and Crelling, 2008; Huang et al., 2012; Zhang et al., 2016). However, Th concentrates in coal as a detrital mineral grain from the source region (e.g., Swaine and Goodarzi, 1995) and the formation of organic Th is unlikely (Finkelman et al., 2019). Potassium is generally associated with inorganic materials e.g., clays, which are common in coal (Finkelman et al., 2018b).

In fly ash, the evaluated means (ranges) activity concentrations are ^{238}U: 266.7 ± 88.8 (203.9-329.5), ^{226}Ra: 170.5 ± 7.0 (165.5-175.4), ^{232}Th: 247.5 ± 23.0 (231.2-263.7), and ^{40}K: 269 ± 12.4 (260.2-277.8) Bq.kg^{-1}, respectively (Table 3. 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 3. 1). Similarly, in bottom ash, the calculated means (ranges) activities are ^{238}U: 169.8 ± 28.9 (149.3-190.2), ^{226}Ra: 121.4 ± 13.4 (111.9-130.9), ^{232}Th: 172.9 ± 21.5 (157.7-188.1), and ^{40}K: 185.5 ± 2.2 (183.9-187.0) Bq.kg^{-1}, respectively (Table 3. 1). In pond ash, the activities are ^{238}U: 158.8 ± 36.4, ^{226}Ra: 119.0 ± 9.4, ^{232}Th: 167.7 ± 1.6 and ^{40}K: 253.4 ± 13.1 Bq.kg^{-1}, respectively (Table 3. 1). The mean activity concentrations of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K 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 3. 1). The highest average activity level of ^{238}U 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 3. 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 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 3. 1). The activity results show that the concentration of ^{226}Ra is less than that of ^{232}Th and ^{238}U in the samples examined.

In comparison with the typical world soil average radioactivity of ^{238}U, ^{226}Ra, and ^{232}Th, 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 ^{40}K (Table 3. 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 in coal and

4.26 to 5.73 times more in CCRs than crustal average concentrations, respectively (Table 3. 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 ^{40}K) (Table 3. 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 and Goodarzi, 1995; Finkelman et al., 2018b). Thorium concentrates in coal as a detrital mineral grain from the source region (e.g., Swaine and Goodarzi, 1995) and the formation of organic Th is unlikely. Potassium is generally associated with inorganic materials e.g., clays, which are common in coal (Swaine and Goodarzi, 1995; Finkelman et al., 2018b).

In Table 3.2, 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 ^{226}Ra 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), and are slightly lower than those in Poland (Bem et al., 2002).

3.1.2 Fractionation of radionuclides among the feed coals and CCRs

The calculated enrichment ratio, ER (ash/coal) suggest 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 coal is 3.10 to 3.37 times lower than that in CCRs samples (Table 3.3). 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 China (Lu et al., 2012) (Table 3.3).

The normalized enrichment factor, EF (with respect to feed coal) values of ^{238}U, ^{226}Ra, and ^{232}Th 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 (Table 3.4). 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), Poland (Bem et al., 2002), Greece (Karangelos et al., 2004) (Table 3.4).

During combustion, most of the organic constitutes (OM) in coal matrix oxidizes leading to the enrichment of natural radionuclides in different fractions in CCRs matrix compare to the feed coal (Bhangare et al., 2014). Most of the incombustible constituents containing radionuclides in coal are partly released from the coal matrix in the finest particulate form. 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) (Papastefanou, 2010; Hasani et al., 2014) and hence, inorganic fraction controls the radioactivity in CCRs (Cevik et al., 2007; Lauer et al., 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 3.4).

The partition ratios, PR (fly ash/bottom ash) of ^{238}U, ^{226}Ra, ^{232}Th, and ^{40}K 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 3. 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) and tend to be retained in bottom ash mineral matter (Bem et al., 2002; Papastefanou, 2010; Li et al., 2012; Hasani et al., 2014; Al-Masri et al., 2015). 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 (Zhou et al., 2012). Additionally, ^{238}U and ^{226}Ra show a preference for adsorption on very fine to ultrafine ash fractions (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).

Table 3.1 Values of natural radioactivity and normalized enrichment factor (EF) of determined radionuclides in the samples from BCM and BTPS in this work and compared with world average specific activity values for different geomaterials

Sample	^{238}U		^{226}Ra		^{232}Th		^{40}K		EF		
	[Bq.kg^{-1}]	[±]	[Bq.kg^{-1}]	[±]	[Bq.kg^{-1}]	[±]	[Bq.kg^{-1}]	[±]	^{238}U	^{226}Ra	^{232}Th
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	-	-	-	-
$(CCRs/WFA)_{average}$	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 in press.

Table 3.2 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 ($mSv.y^{-1}$), 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
		^{238}U	^{226}Ra	^{232}Th	^{40}K	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.

Table 3.3 Comparison of the enrichment ratio (ER) in the samples studied from Barapukuria with the literature data.

Country		Enrichment ratio (ash/coal)				Reference
		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.

Table 3.4 Comparison of the normalized enrichment factor (EF) in the samples analyzed from Barapukuria in previously published work.

Country	Sample	Enrichment factor			Reference
		^{238}U	^{226}Ra	^{232}Th	
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

In contrast, Th exhibits no different partitioning behavior during burning and it mostly occurs in inorganic part both in coals and associated CCRs (Swaine and Goodarzi, 1995). The ^{238}U, being more soluble in water and having a solubility nature compared to ^{232}Th, 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 (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 (Hasani et al., 2014; Lauer et al., 2017).

3.1.3 Radiological hazard assessment

The calculated hazard index values due to the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K radionuclides in the bulk/core coal, pulverized coal, fly ash, bottom ash, and pond ash samples from BCM and BTPS appear in Table 3.5. The average relative contributions of ^{226}Ra, ^{232}Th and ^{40}K 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 (Figure 3.1). The relative radium equivalent activity contributions of radionuclides in the samples are in descending order ^{232}Th > ^{226}Ra > ^{40}K, where Th and Ra mutually contribute more than 90% (Figure 3.1).

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 3.5; Figure 3.2a).

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 (Figure 3.2a). The investigated CCRs contain 2 to 3 times more natural radionuclides than pulverized coal which is almost similar with Turhan et al. (2018). 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 CTPs (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 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 pulverized coal, 0.30 for fly ash, 0.20 for bottom ash and 0.21 for pond ash, (all values are in $mSv.y^{-1}$ and the permissible limit is 0.07 $mSv.y^{-1}$). 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) (Figure 3.2b).

Compare to the results of other studies (Table 3. 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.

Table 3.5 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 ($mSv.y^{-1}$), 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	$\leq 2.9\times10^{-4}$

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. Radiation indices were computed from their respective reported activities following formulas adopted by UNSCEAR 2000.
[a]UNSCEAR, 2010; [b]UNSCEAR, 1982; [c]Bowen, 1979; [d]Eisenbud and Gesell, 1997; [e]UNSCEAR, 2000.

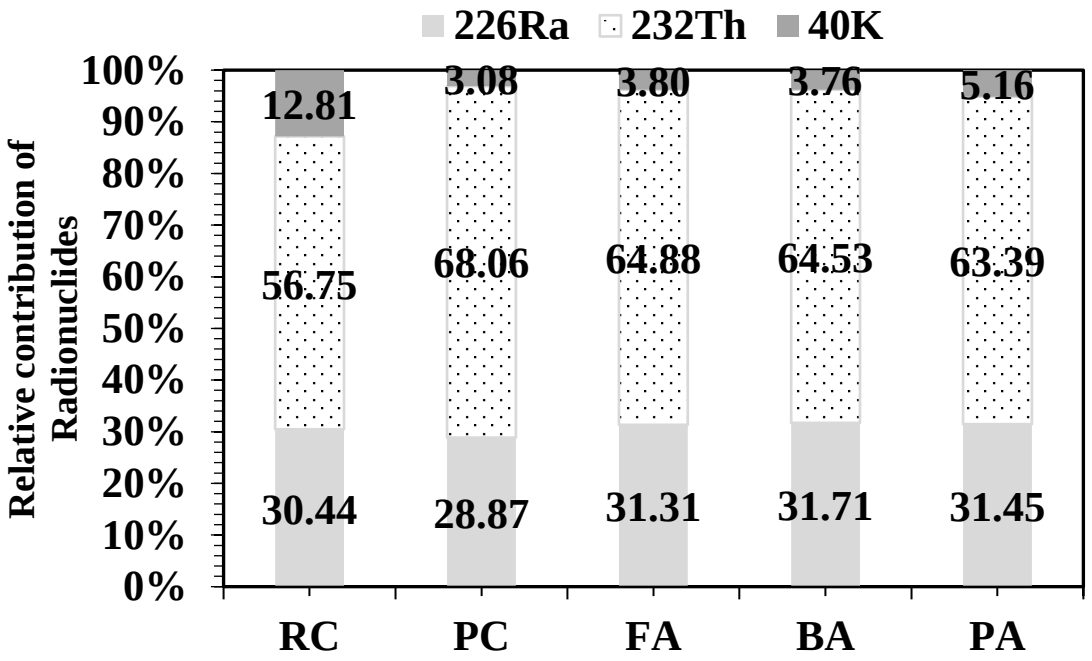

Figure 3.1 Relative ra equivalent activity contribution of radionuclides in coal and ash from Barapukuria. RC: raw (bulk/core) coal, PC: pulverized coal, FA: fly ash, BA: bottom ash, PA: pond ash.

Figure 3.3 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), Poland (Bem et al., 2002), Turkey (Cevik et al., 2008), Greece (Karangelos et al., 2004). All radioactive progenies of ^{238}U and ^{232}Th 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 (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 (Amin et al., 2013; Bhangare et al., 2014; 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 UNSCEAR (2000) to avoid potential radiation hazards to the respiratory organs (Durašević et al., 2014).

Nevertheless, it is clear that the activity concentrations and corresponding hazard indices are generally higher in CCRs and pose a potential radiological risk to the environment, occupational workers, and the entire population around the BTPS. Hence, BTPS generated CCRs has 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 CCRs are not carefully disposed and managed.

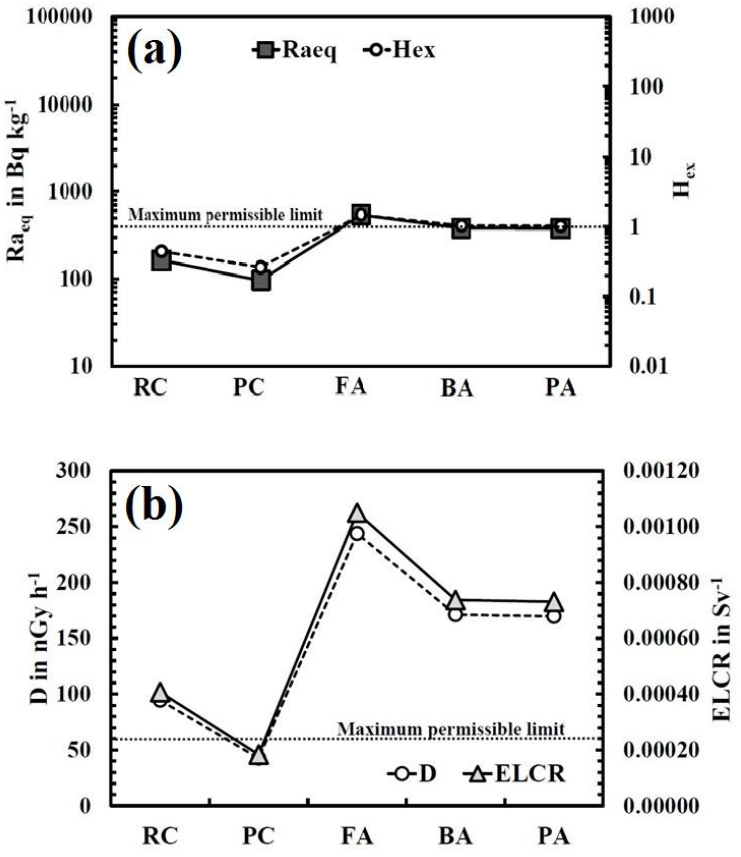

Figure 3.2 Hazardous indices: (a) radium equivalent activity, Ra_{eq} **(**Bq.kg^{-1}**)** and external hazard index, H_{ex}, (b) external absorbed gamma dose rate, D **(**nGy.h^{-1}**)** and excess life time cancer risk, ELCR **(**Sv^{-1}**)** due to ^{226}Ra, ^{232}Th and ^{40}K for the investigated coal and ash samples of this study. RC: raw (bulk/core) coal, PC: pulverized coal, FA: fly ash, BA: bottom ash, PA: pond ash.

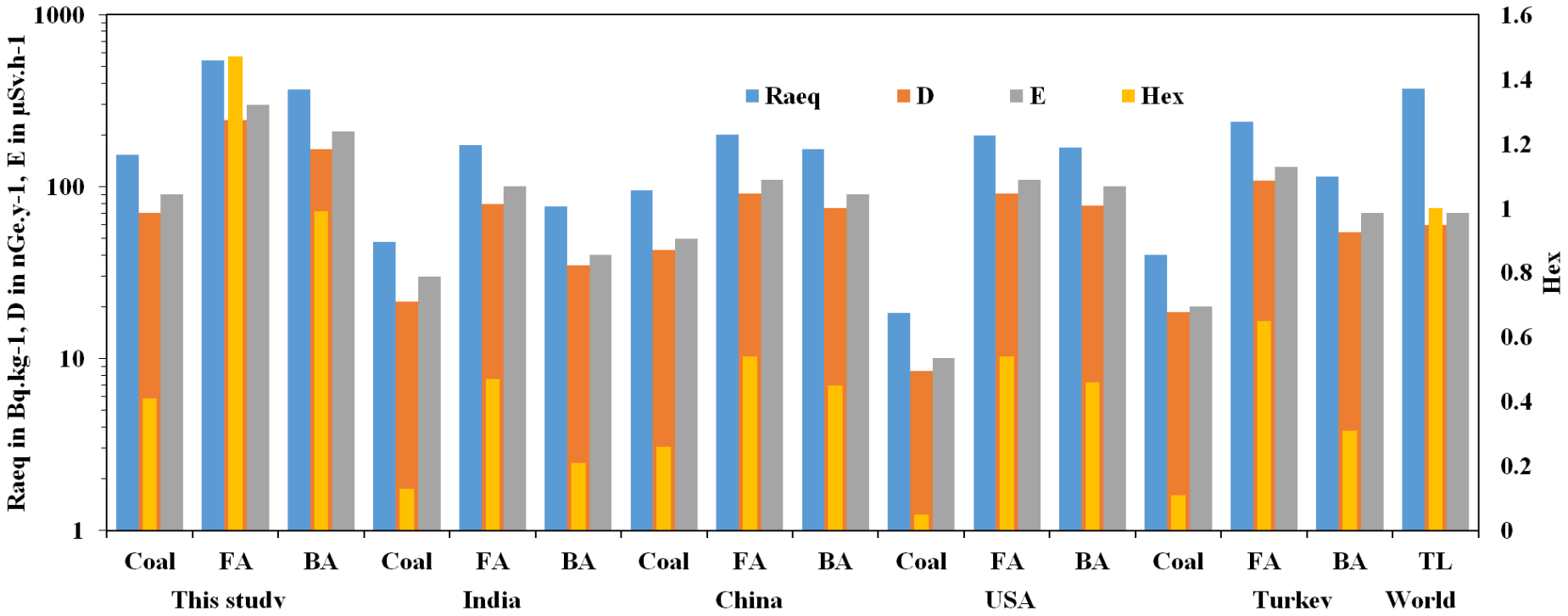

Figure 3.3 Evaluated radiological hazard parameters from this study compared with literature data. The data is taken from the following references: Bangladesh (present study), India (Sahu et al., 2014), China (Lu et al., 2012), USA (Coles et al., 1978), Poland (Bem et al., 2002), Turkey (Cevik et al., 2008), Spain (Mora et al., 2009), Greece (Karangelos et al., 2004). TL: threshold limit.

3.2 Radiological characteristics of soils from Barapukuria (Paper II)

3.2.1 Physicochemical characteristics of soil samples

Physicochemical parameters such as pH and soil organic matter (SOM) are of great importance due to their influences on the mobility, solubility and complexation of the radionuclides in the soils (Dragović et al., 2013). In our study, pH values of the soil samples vary from 4.0 to 6.3 with a mean value of 5.4, which indicate the moderate to strong acidity of the soil samples. Correspondingly, SOM of the soil samples range from 0.6% to 14.4% with a mean value of 2.6% (Table 3.6). Mean grain sized-fraction of sand, silt and clay are 7.8%, 75.1%, 17.2%, respectively invoking the uneven distributions of constituent fractions in the bulk soil. However, most samples belong to the silty loam or silt classes.

3.2.2 Radioactivity concentrations in soil samples

The basic descriptive statistics of radionuclides specific activity in 24 soil samples are summarized in Table 3.6. Activity of ^{226}Ra in the soils varied from 51.2-77.6 with mean value of 63.6 ± 7.4, ^{238}U from below detection limit to 192.4 with mean value of 102.9 ± 41.4; ^{232}Th from 71.5-126.1with mean value of 103.4 ± 13.9 and ^{40}K from 210.5-763.3 with mean value of 494.2 ± 107.5 (all units are in Bq.kg^{-1}), respectively. The highest coefficient of variation of activity is caused for U (40.3%) and the lowest for Ra (11.6%) (Table 3.6).

The obtained specific activity are about 1.3-3.5 times greater than the typical world average value for soil (UNSCEAR, 2000). The measured activity concentrations in soils are within the permissible limit, i.e., 370 Bq.kg^{-1}, 259 Bq.kg^{-1} and 925 Bq.kg^{-1} for ^{226}Ra, ^{232}Th and ^{40}K, respectively (Carini, 2001). The relative contribution to the total activity in soil samples are in descending order ^{40}K > ^{232}Th > ^{238}U > ^{226}Ra. The frequency distribution of determined radioisotopes activity follows normal distribution (significant at the 0.05 level) (Table 3.6).

Feed coal and fly-ash from BTPS were also studied and the mean radioactivity (Bq.kg^{-1}) are found to be 44.9 ± 13.4 for ^{238}U, 27.6 ± 2.3 for ^{226}Ra, 45.5 ± 1.1 for ^{232}Th, and 38.2 ± 5.0 for ^{40}K, and 329.5 ± 29.8 for ^{238}U, 175.4 ± 13.9 for ^{226}Ra, 263.7 ± 0.8 for ^{232}Th and 277.8 ± 8.9 for ^{40}K, respectively (Table 3.6). The specific activities in fly-ash are dramatically higher than the respective activity in feed coal. The activity of radionuclide is 2.0-3.2 times lower in the soils than FA from BTPS, except for ^{40}K.

In Table 3.7, a summary of the measured specific activities of radioisotopes in soil samples of this study along with the literature data from similar investigations are tabulated. Radioactivity concentrations of Barapukuria soil samples are considerably higher than those of

other parts of the world (UNSCEAR, 2000; Bem et al., 2002; Flues et al., 2002; Papp et al., 2002; Flues et al., 2007; Cevik et al., 2008), except for China (Lu et al., 2012; Liu et al., 2015).

In Barapukuria soil, the estimated average elemental abundances of K, U and Th are 1.6 ± 0.3 %, 9.1 ± 2.2 ppm and 25.5 ± 3.4 ppm, respectively. Along with the Th and U, K content in the Barapukuria soil samples are considerably higher than those of world average soil samples (Bowen, 1979) as presented in Table 3.6.

3.2.3 Spatial distributions of soil radionuclides

Statistically no significant spatial differences ($p \leq 0.05$) have been observed for specific activities in soil samples of different observation points having different distance and directions from the point source (BTPS). This is confirmed by the employing one-way ANOVA test (between groups and within groups), which point out that no remarkable differences to the total variance of all radioactivity with distance-direction variation (^{238}U : F = 1.173, p = 0.329; ^{226}Ra : F = 0.109, p = 0.897; ^{232}Th : F = 1.448, p = 0.258; ^{40}K : F = 0.382, p = 0.687). It indicates that the measured activity values were statistically equal, which is invoking the influence of the BTPS operation is insignificant (Table 3.8).

A box-plot was employed to evaluate the asymmetry distribution of the activity concentrations attained of the soil samples (Figure 3.4). This graphic depiction allows a visualization of the results dispersion in samples, median (–), range of data variation, as well as comparison between different radionuclides. It is worth to be noted here that K content in Barapukuria soil samples is considerably higher than those of K content in coal and fly-ash (Table 3.6). However, the mean concentrations of ^{226}Ra and ^{232}Th are not fluctuated with distance.

Figure 3.5 display irregular distribution pattern of radionuclides in soil over the area studied. Elevated concentration of U is found southeast direction of the plant. Three scattered elevated concentrations of Th and Ra are clearly found at three different observation sites. Uranium-238 concentration map exhibits higher in all sites except southeast and northwestern part of the area (Figure 3.5a).

The map of ^{226}Ra spatial activity distribution map exhibits slightly higher in the southeastern and north-central sides than other parts of the mapped area (Figure 3.5b). Thorium-232 distribution map shows higher activity in all around the plant except southwest part of the area (Figure 3.5c). Potassium-40 in soil revealed its wide variation around CTP. Its activities are less around plant but highest in the southwestern and north-central part than that of the surrounding areas (Figure 3.5d).

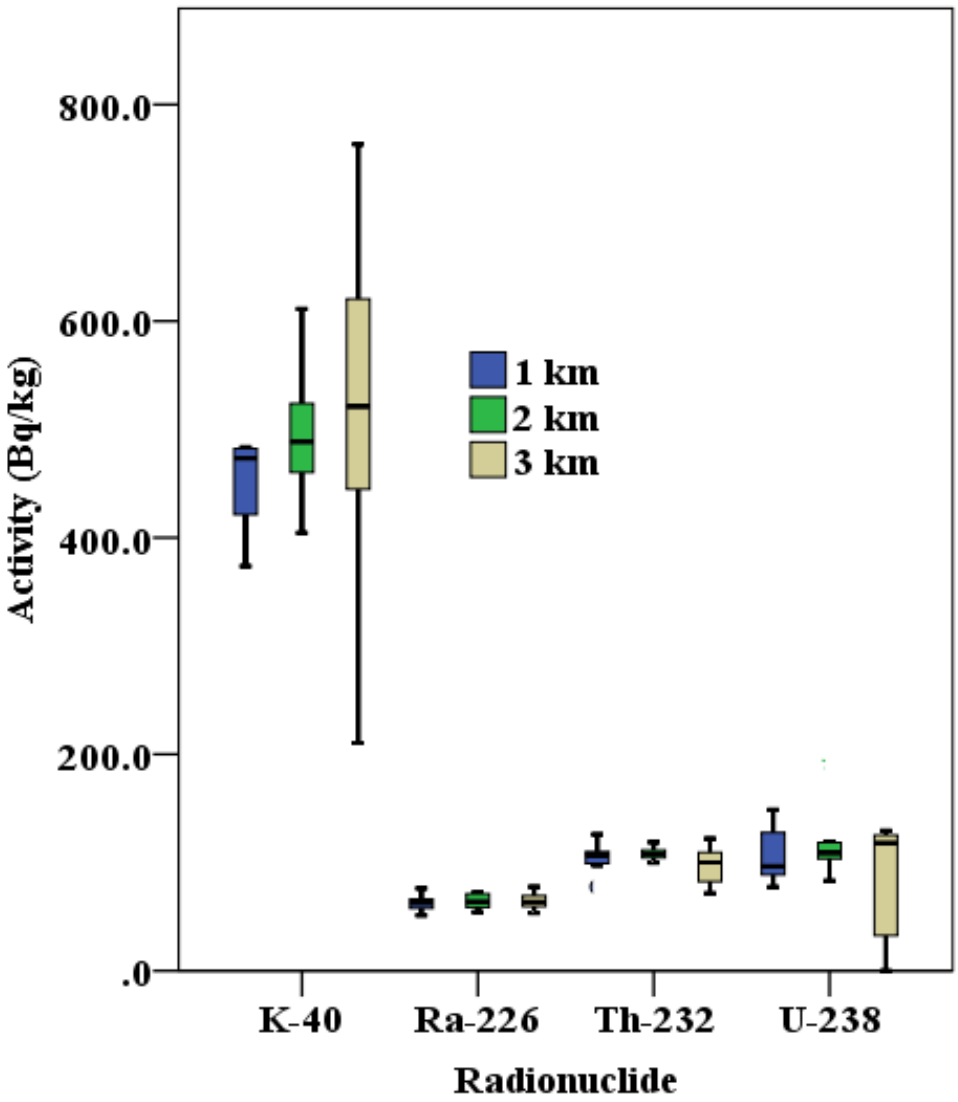

Figure 3.4 Histogram showing the distribution of the activity concentrations ($Bq.kg^{-1}$) of radionuclides and their variability in top soils from Barapukuria along with the mean and median (–), range of data variation.

The spatial variability maps do not exhibit any particularly special distribution trend over the area. However, the obtained specific activity values in soils around BTPS are virtually close to the mean natural background activity soil (taken from 15 km away from BTPS from similar undisturbed soil type) which is concomitant with those of previous literature studies (Vuković et al., 1996; Charro and Pena, 2013; Ćujić et al., 2015).

Table 3.6 Comparative study of activity concentrations ($Bq.kg^{-1}$) of radionuclides among this study and other literature data for samples with similar investigation.

Reference	Type	^{238}U	^{226}Ra	^{232}Th	^{40}K
This study					
	Fly ash	329.5	175.4	263.7	277.8
	Feed coal	44.9	27.6	45.5	38.2
	Soil	102.9	63.6	103.4	494.2
Literature data					
SE Bangladesh[a]	Soil	-	18	46	321
NW Bangladesh[b]	Soil	-	91	151	1958
India[c]	Fly ash	-	40-152	96-178	148-840
	Slag		44-156	74-215	373-633
	Feed coal		11-67	18-93	14-445
	Soil		37	69.6	396
			(14-156)	18-156	(11-707)
China[d]	Fly ash	-	69.5	79.3	233
	Slag		59.5	61.8	222.6
	Feed coal		33	37.5	105.7
China[e]	Soil		225	257	1571
Brazil[f]	Fly ash	-	1442-2718	43-95	471-1144
	Slag		1387-3621	45-92	422-525
	Feed coal		813-1251	22-40	200-450
	Soil		133	39	233
Turkey[g]	Fly ash	-	242	51	493
	Slag		313	51	307
	Feed coal		81	39	435
	Soil		33	36	379
Poland[h]	Fly ash	-	75-120	47-92	448-759
	Slag		32-91	28-80	307-608
	Feed coal		13-29	8-21	43-181
	Soil		9-23	9-20	221-435
Hungary[i]	Soil	-	129	25.5	329
Malaysia[j]	Soil	-	87	74	297
South Bangladesh[k]	Soil	-	42	81	833
Bangladesh[l]	Soil	-	34	-	350
			(21-43)		(130-610)

SE: south-east; NW: north-west.

[a]Rashed-Nizam et al., 2015; [b]Hamid et al., 2002, [c]Mishra, 2004, [d]Lu et al., 2012, [e]Liu et al., 2015, [f]Flues et al., 2002, 2007; [g]Cevik et al., 2008; [h]Bem et al., 2002; [i]Papp et al., 2002, [j]Amin et al., 2013; [k]Chowdhury et al., 2006, [l]UNSCEAR, 2000.

Table 3.7 Activity concentrations of radionuclides, abundance of radioactive elements in the soils from the surroundings of Barapukuria coal-based thermal plant (BTPS) with their associated uncertainties, test of normality (K-S α), and physicochemical properties.

	^{238}U		^{226}Ra		^{232}Th		^{40}K		U	Th	K	pH	OM	Sand	Silt	Clay
	[Bq.kg^{-1}]	[±]	[Bq.kg^{-1}]	[±]	[Bq.kg^{-1}]	[±]	[Bq.kg^{-1}]	[±]	[ppm]	[ppm]	[%]	[-]	[%]	[%]	[%]	[%]
This study																
S1	129.4	27.4	77.6	2.6	105.0	0.2	527.6	3.1	10.4	25.9	1.7	5.9	1.6	25.0	67.1	7.9
S2	118.5	14.5	58.7	1.8	107.9	2.9	483.5	1.7	9.6	26.6	1.5	5.3	1.0	8.0	66.4	25.6
S3	109.1	37.3	72.5	3.3	105.3	0.5	435.2	4.7	8.8	26.0	1.4	5.8	0.9	2.0	71.7	26.3
S4	121.2	16.3	64.2	1.6	107.1	0.9	515.0	1.9	9.8	26.4	1.6	5.0	0.7	7.3	67.4	25.3
S5	77.5	20.9	65.9	1.3	108.4	0.8	420.7	1.9	6.2	26.8	1.3	5.5	9.5	4.0	77.5	18.5
S6	192.4	17.2	71.9	2.3	118.8	0.0	494.5	2.9	15.5	29.3	1.6	5.9	1.6	6.0	52.0	42.0
S7	148.8	23.6	57.5	3.1	106.9	0.2	373.8	4.3	12.0	26.4	1.2	5.3	9.8	5.0	77.0	18.0
S8	bdl	14.3	74.6	1.6	122.2	2.9	210.5	1.9	-	30.2	0.7	6.3	14.4	1.0	82.0	17.0
S9	124.4	21.1	59.1	2.5	111.3	0.1	641.5	2.3	10.0	27.5	2.0	5.3	1.0	7.5	78.0	14.5
S10	84.2	33.3	55.8	3.6	99.8	0.2	611.1	2.8	6.8	24.6	2.0	5.6	0.9	10.0	74.4	15.6
S11	96.6	31.8	76.3	2.3	126.1	1.8	481.2	2.9	7.8	31.1	1.5	5.1	2.6	6.5	72.5	21.0
S12	86.9	18.1	62.8	1.9	100.5	3.3	422.1	2.2	7.0	24.8	1.3	4.9	2.2	5.0	91.0	4.0
S13	147.9	19.1	57.9	2.9	97.5	0.2	473.3	3.2	11.9	24.1	1.5	5.3	0.7	8.0	65.0	27.0
S14	113.3	38.9	63.9	3.0	108.5	0.1	523.8	3.8	9.1	26.8	1.7	6.0	0.6	13.0	73.2	13.8
S15	126.2	14.0	59.9	2.3	72.2	1.5	763.3	5.3	10.2	17.8	2.4	4.9	1.4	4.0	74.3	21.7
S16	65.3	20.7	63.8	1.4	95.4	2.2	476.7	1.8	5.3	23.6	1.5	4.0	2.8	10.0	82.0	8.0
S17	83.3	25.6	59.4	2.1	103.6	0.3	404.4	2.6	6.7	25.6	1.3	5.6	0.7	8.0	79.0	13.0
S18	bdl	-	63.4	2.9	92.8	0.3	412.9	3.4	-	22.9	1.3	5.4	0.9	2.0	89.0	9.0
S19	107.6	14.0	66.7	1.9	111.5	1.5	483.4	1.6	8.7	27.5	1.5	5.9	0.6	5.0	91.0	4.0
S20	119.3	6.8	54.1	1.8	111.4	2.5	460.4	1.7	9.6	27.5	1.5	4.60	1.10	5.00	80.00	15.00
S21	109.1	19.2	71.1	2.5	116.2	3.3	533.9	2.3	8.8	28.7	1.7	5.6	0.9	16.0	62.8	21.2
S22	114.6	24.2	53.3	3.1	71.5	0.2	599.2	2.6	9.2	17.7	1.9	5.2	1.9	10.0	77.0	13.0
S23	90.4	13.7	51.2	1.7	77.5	3.1	624.5	1.3	7.3	19.1	2.0	6.2	0.9	11.0	75.0	14.0
S24	103.4	43.3	64.1	7.2	104.5	13.4	488.6	1.6	9.2	25.8	1.6	5.4	4.0	7.7	75.0	17.3
Mean	**102.9**	**-**	**63.6**	**-**	**103.4**	**-**	**494.2**	**-**	**9.1**	**25.5**	**1.6**	**5.4**	**2.6**	**7.8**	**75.0**	**17.2**
SD	**41.4**	**-**	**7.4**	**-**	**13.9**	**-**	**107.5**	**-**	**2.2**	**3.4**	**0.3**	0.5	3.5	5.1	9.0	8.5
Min	**-**	**-**	**51.2**	**-**	**71.5**	**-**	**210.5**	**-**	**5.3**	**17.7**	**0.7**	**4.0**	**0.6**	**1.0**	**52.0**	**4.0**
Max	**192.4**	**-**	**77.6**	**-**	**126.1**	**-**	**763.3**	**-**	**15.5**	**31.1**	**2.4**	**6.3**	**14.4**	**25.0**	**91.0**	**42.0**
K-S α[a]	0.163	-	0.200	-	0.065	-	0.19	-	-	-	-	-	-	-	-	-
Background[b]	91.4	7.1	63.5	9.7	99.6	1.3	579.5	3.1	5.1	24.5	1.9	-	-	-	-	-
Fly ash[b]	329.5	29.8	175.4	13.9	263.7	0.8	277.8	8.9	26.6	65.1	0.89	-	-	-	-	-
Feed coal[b]	44.9	13.4	27.6	2.3	45.5	1.1	38.2	5.0	3.6	11.2	0.12	-	-	-	-	-
Literature data																
World soil	35[c] (16-110)		35[c] (17-60)		30[c] (11-64)		400[c] (140-850)		7.4[d]	2.8[d]	1.3[d]	-	-	-	-	-
Europe average[c]	46		-	-	31	-	465	-	-	-	-	-	-	-	-	-
UCC[e]	-	-	33	-	43	-	720	-	2.7	10.5	2.3	-	-	-	-	-
Permissible limits[f]			370		259		925		29.9	63.8	2.9					

OM: organic matter, bdl: below detection limit, SD: standard deviation, Min: minimum, Max: maximum. [±]: 1σ variation due to counting uncertainties. S1 Sample locations (refer location map in Figure 2.1). [a]K-S α, Normality test (Kolmogorof-Semirnov), [b]Present study, [c]UNSCEAR, 2000, [d]Bowen, 1979, [e]Rudnick and Gao, 2014, [f]Carini, 2001.

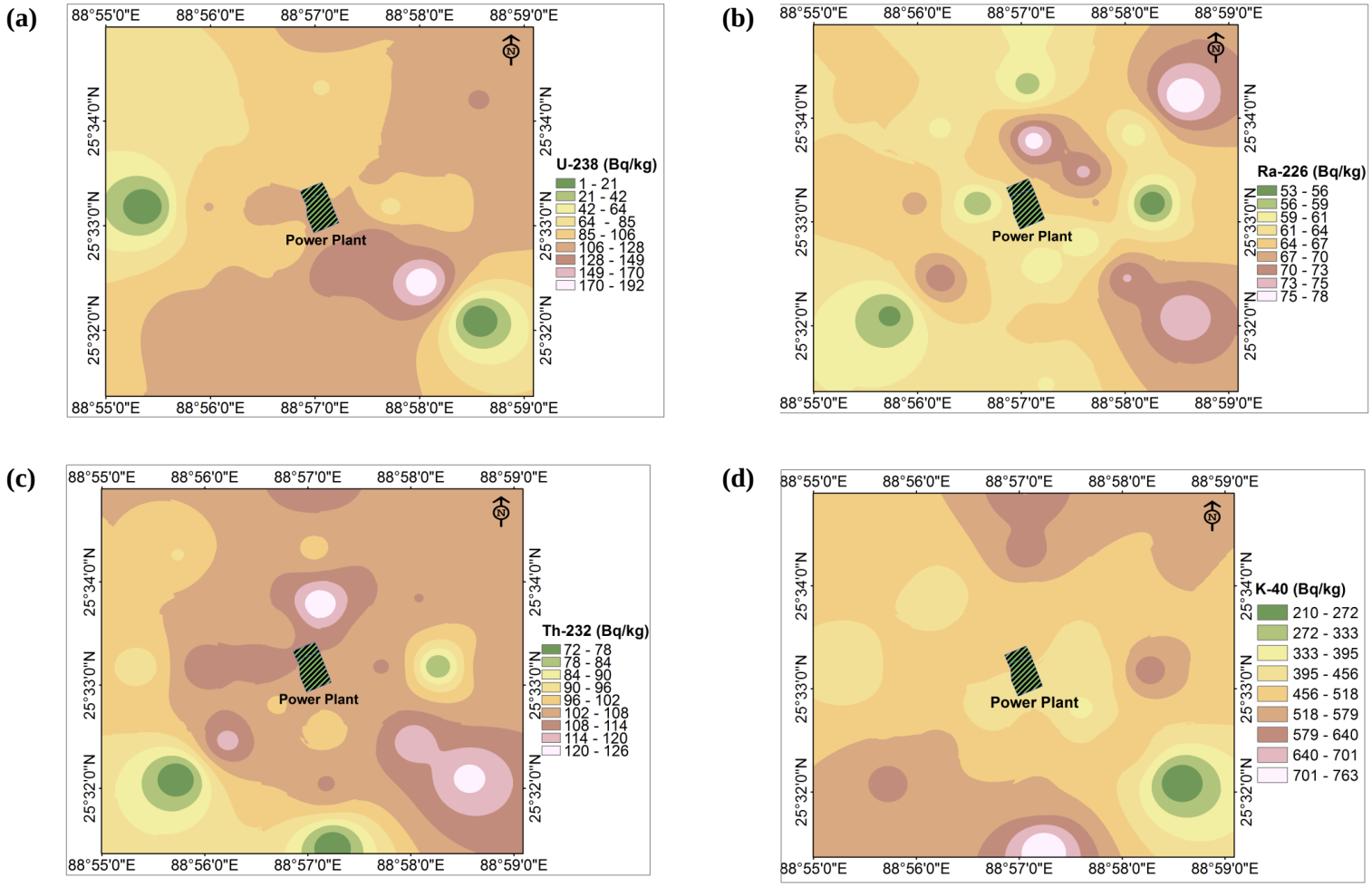

Figure 3.5 Inverse distance weighting (IDW) map for the spatial distribution of radionuclides activity in the soils from BTPS vicinity. (A) ^{238}U, (B) ^{226}Ra, (C) ^{232}Th, and (D) ^{40}K activity concentrations in $Bq.kg^{-1}$.

Table 3.8 One-way ANOVA test results for radionuclides activity concentration difference in soil samples from Barapukuria.

		Sum of squares	df	Mean square	F	Sig.
^{238}U	Between Groups	3967.618	2	1983.809	1.173	0.329
	Within Groups	35509.347	21	1690.921		
	Total	39476.965	23			
^{226}Ra	Between Groups	12.910	2	6.455	0.109	0.897
	Within Groups	1240.764	21	59.084		
	Total	1253.674	23			
^{232}Th	Between Groups	539.607	2	269.803	1.448	0.258
	Within Groups	3914.139	21	186.388		
	Total	4453.746	23			
40K	Between Groups	9321.741	2	4660.871	0.382	0.687
	Within Groups	256545.715	21	12216.463		
	Total	265867.456	23			

df: degree of freedom, F: statistics value (sum of squares/mean square), Sig.: significance.

3.2.4 Correlation study

In the areas of highly homogeneous lithology, strong correlations among the parameters (e.g., radionuclides) are quite common (Charro et al., 2013). So, to draw the provenance of radionuclides and their relationship with the soil physicochemical properties, a Pearson correlation matrix is tabulated in Table 3.9. ^{238}U shows positive correlation with ^{40}K ($r = 0.431$, $\alpha \leq 0.05$), which signify their common mineralogical affinity and/or similar source(es). Both U and K are released from parent minerals (e.g., clay minerals) as ions during weathering, and thereafter preferentially adsorbed by the clays (Charro et al., 2013). Then from the clay, U and K are transferred to the soil solution and are being available for migration and uptake by the crops/vegetation (Kabata-Pendias, 2011). Similarly, to the ^{238}U and ^{40}K; ^{232}Th and ^{226}Ra are also significantly correlated ($r = 0.623$, $\alpha \leq 0.01$). ^{40}K is inversely correlated with SOM ($r = -0.631$, $\alpha \leq 0.01$) which reveals inorganic mineralogical (e.g., illite) affiliation of K instead of organic association. Similar to the SOM, ^{232}Th also shows inverse correlation with ^{40}K ($r = -0.558$, $\alpha \leq 0.01$) which indicates their differential geochemical behavior. Uranium-238 is significantly correlated with clay sized fraction ($r = 0.521$, $\alpha \leq 0.01$), and inversely correlated with silt fraction ($r = -0.653$, $\alpha \leq 0.05$) and SOM ($r = -0.502$, $\alpha \leq 0.05$), which demonstrate the lithogenic origin of U and dominant association with finer fractions. The soil pH is found to be correlated with ^{226}Ra ($r = 0.45$, $\alpha \leq 0.05$), which indicates solubility and mobility of Ra increases with increasing soil acidity (Papaefthymiou et al., 2007, 2013).

Table 3.9 Mutual correlation matrix of radionuclides and soil properties of Barapukuria.

	pH	OM	Sand	Silt	Clay	^{238}U	^{226}Ra	^{232}Th	^{40}K
pH	1								
OM	.361	1							
Sand	.089	-.329	1						
Silt	-.215	.184	-.385	1					
Clay	.176	.000	-.188	-.834**	1				
^{238}U	-.031	-.502*	.291	-.653**	.521**	1			
^{226}Ra	.450*	.316	.140	-.242	.174	-.097	1		
^{232}Th	.376	.316	-.049	-.208	.250	.035	.623**	1	
^{40}K	-.278	-.631**	.351	-.238	.044	.431*	-.375	-.558**	1

*Correlation is significant at the 0.05 level (2-tailed).
**Correlation is significant at the 0.01 level (2-tailed).

3.2.5 Soil mineralogy and radionuclide's source

Uranium and Th concentrations in soil are closely related to the parent bedrock of the soil and crystal structure of the associated minerals. So, to evaluate the radionuclide's mineralogical provenance Barapukuria soil samples have been studied. In the present study, the major minerals in Barapukuria soil's are found to be kaolinite, illite, quartz, Fe-oxides minerals (Figure 3.6A), and the accessory heavy minerals are also found, e.g., monazite, rutile, biotite, zircon, kyanite, garnet, and tourmaline mineral assemblages (Figure 3.6B) (Mange and Maurer, 1992). Aftabuzzaman et al. (2013) demonstrated that kaolinite and illite (source of ^{40}K) in Barapukuria soil are volumetrically the most abundant and common minerals. Nevertheless, it is well established that clays (e.g., illite, kaolinite) are the main geochemical carriers, concentrator and great repository of radionuclides (such as ^{238}U) in soils (Swaine and Goodarzi, 1995), which are most likely governing the soil radioactivity. However, U is associated with accessory minerals, such as zircon while the carrier of Th is mainly monazite which is very resistant to weathering. Along with monazite, a significant portion of Th is also partitioned into zircon, and clays (Swaine and Goodarzi, 1995). Thus, the elevated specific activity of ^{232}Th in our sample is due to the high content of monazite (Finkelman et al., 2018b).

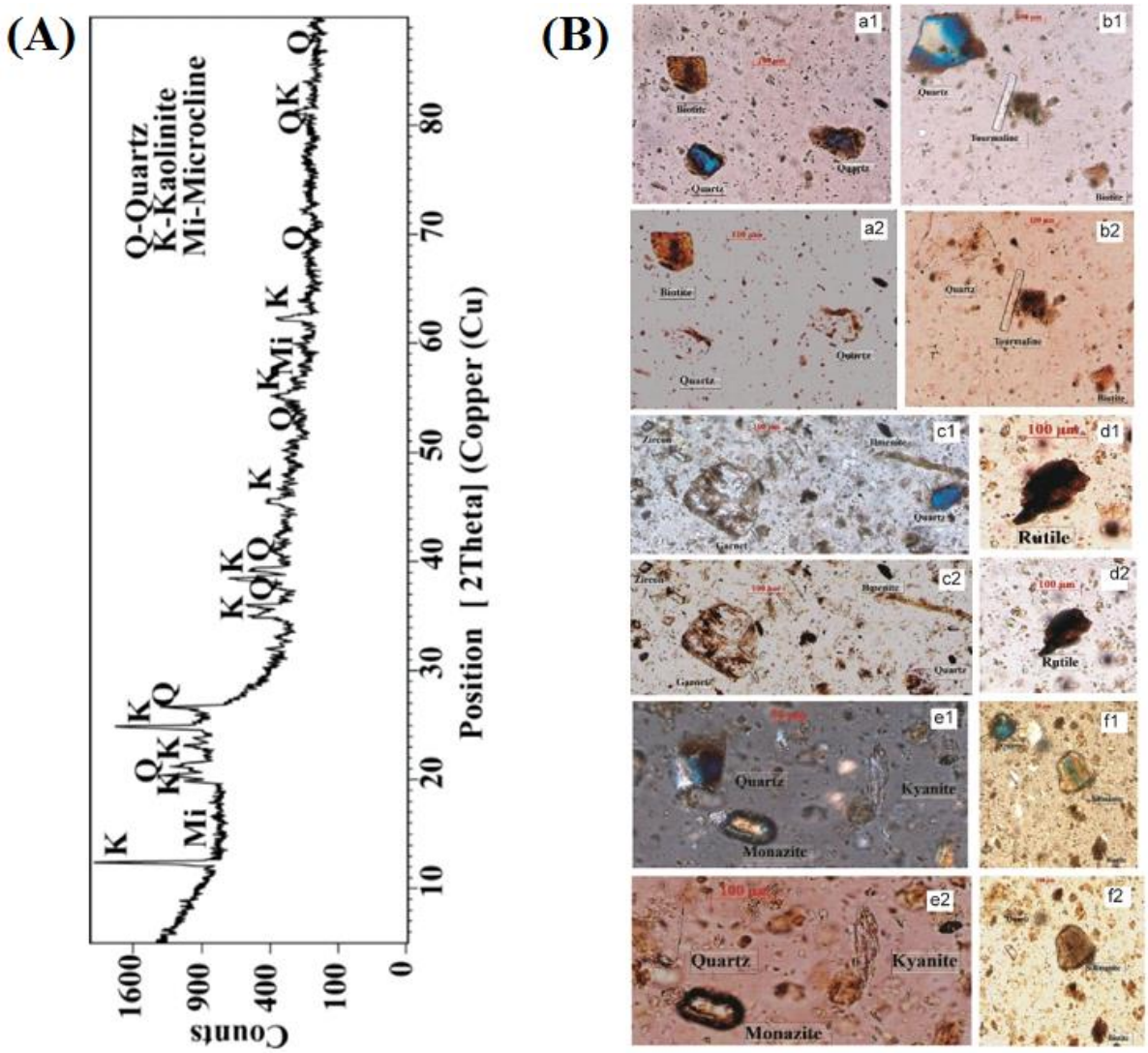

Figure 3.6 XRD pattern of soils (A) and photomicrographs of soil minerals (B) from Barapukuria. Images of (a) biotite, quartz in sample s9; (b) quartz, tourmaline and biotite in sample (s9); (c) zircon, garnet, ilmenite and quartz in sample s13; (d) rutile in sample s13; and (e) monazite, kyanite and quartz and biotite in sample s19. (all images are in plane, a1, and cross, a2, polarized transmitted light, respectively).

Presumably, soil radionuclides concentration principally determines by the soil minerals, local variability of radionuclide's distribution due to the variation of the soil properties and different geochemical behavior of radionuclides in the soil. It is inferred that the detected radioisotopes are most likely carried and governed by these identified minerals in the soil sample of this study. It could be ascribed to the fact that all examined soil samples might be enriched with the radionuclides from the results of the natural dispersion process rather than anthropogenic (technogenic) attribution. On the contrary, while authors reported that there is

no influence of radionuclides releasing from CTPs on the local environment rather it is governed by mineralogy in the soils, soil characteristics, geomorphology and terrain characteristics, radionuclides physicochemical behavior, soil type and practice and local climatic factors as well as technological feature of CTP (filtration efficiency), mode of CCRs disposal, means of way of ash management, wind speed and direction, duration of plant operation, coal quality that is combusted (Habib et al., 2018 in press).

3.2.6 Assessment of soil contamination level

In order to determine the degree of contamination due to the operation of BTPS, the contamination factor (CF) and pollution load index (PLI) were calculated. The mean background values for topsoil from undisturbed area of similar soil type (15 km away from the point source) are presented in Table 3.6. Calculated mean values for CFs for topsoil were -0.85, -1.04, -1.13, and -1.0 for ^{40}K, ^{232}Th, ^{238}U, and ^{226}Ra, respectively while the estimated PLI values are found to be 0.96 to 1.38 with an average value of 0.83 for all sampling points (Figure 3.7). Similar to Ćujić et al. (2015), we can reasonably infer from the estimated indices that the soils are uncontaminated with radionuclides, except for ^{40}K.

The activity ratio reflects the relative depletion or enrichment of radioisotopes in the geo-environmental materials which can be employed as indicator of the radioactive pollution of the soil samples (Charro et al., 2013). The world mean ratio of ^{238}U/^{232}Th in soil is close to unity (UNSCEAR, 2000). The ratios between daughter and parent radionuclide is not unity indicating disequilibrium within the U and Th decay subseries, and therefore the existence of contamination. The value of the ^{226}Ra / ^{238}U in our study is 0.60 ± 0.16 (0.37-0.98) which indicates a deviation from the radioactive equilibrium. The calculated mean value of ^{238}U / ^{232}Th and ^{232}Th / ^{226}Ra are 1.1 ± 0.3 (0.7-1.8) and 1.4 ± 0.5 (0.5-2.1) respectively. These computed ratio values are very nearly to the UCC (Rudnick and Gao, 2014) ratio values (^{226}Ra / ^{238}U: 0.94, ^{238}U / ^{232}Th: 0.81, and ^{232}Th / ^{226}Ra: 1.3, respectively). The ^{232}Th / ^{226}Ra ratio value in soil samples suggests that the original proportionality is preserved.

The computed mean mass ratio of Th/U is 3.7 in the soil samples of this study, while the ratio values of K/U and Th/K are 1,731.1 and 0.0017, respectively. These ratios for UCC are 3.89, 8592.6 and 0.00045 (Rudnick and Gao, 2014). The ratio K/U is highly variable and may suggest provenance from different sources. The obtained ratio value for Th/K is significantly higher compare to UCC and sedimentary rock but other two ratios are very closer. The Th/U ratio of this study (3.7) indicates that no significant fractionation during different

natural/anthropogenic processes of U and Th has occurred, except for K (Papaefthymiou et al., 2013).

3.2.7 Radiological hazard assessment

The summery statistics of radiation indices are presented in Table 3.10. The deduced values of radium equivalent activity, Ra_{eq}, range from 201.8 to 293.6 $Bq.kg^{-1}$ with a mean value of 249.5 ± 21.7 $Bq.kg^{-1}$, which are far below the internationally accepted values (370 $Bq.kg^{-1}$, respectively) (UNSCEAR, 2000). The values of hazard index, H_{ex} are varied from 0.54 to 0.79 and mean value 0.67 ± 0.06. Since these values are below than the threshold limit unity, soils of this study are quite safe from radiological harmful effect.

In our study, the corresponding average absorbed gamma dose rate, D are found to be 114.2 ± 9.4 with intervals of 94.0-133.6 $nGy.h^{-1}$, which are 1.9 folds higher than quoted global average value (60 $nGy.h^{-1}$) for the public but within the world ranges (10-200 $nGy.h^{-1}$) (UNSCEAR, 2000).

The total annual effective dose, E range from 0.17 to 0.24 with an average value of 0.20 ± 0.02, which is lower than world average value 0.5 $mSv.y^{-1}$ (recommended upper dose limit of 1.0 $mSv.y^{-1}$ (UNSCEAR, 2000). The relative contributions to total external gamma dose rate in air in the decreasing order from ^{232}Th, ^{40}K and ^{226}Ra are 75.3%, 23.6%, and 1.0%, respectively, where ^{232}Th is the major contributor in the studied area. Finally, the values of the excess lifetime cancer risk varied from 4.0×10^{-4} to 5.8×10^{-4} with an average of $4.9 \times 10^{-4} \pm 0.4 \times 10^{-4}$ which is higher than the world average value of 2.9×10^{-4} (UNSCEAR, 2000). All the assessed indices values are within the safety limits. In general, from the viewpoint of radiological protection, Barapukuria soils do not represent radiological threat to the surrounding areas and to the human health (both, staff and public).

Although the largest attribution to the radiation doses from ^{40}K (63.9%), it can be seen that spatial variability of dose is mainly controlled by the concentrations of Ra and Th (Figure 3.8). Hence the isolevel maps follow to a great extent the distribution pattern of these two radionuclides.

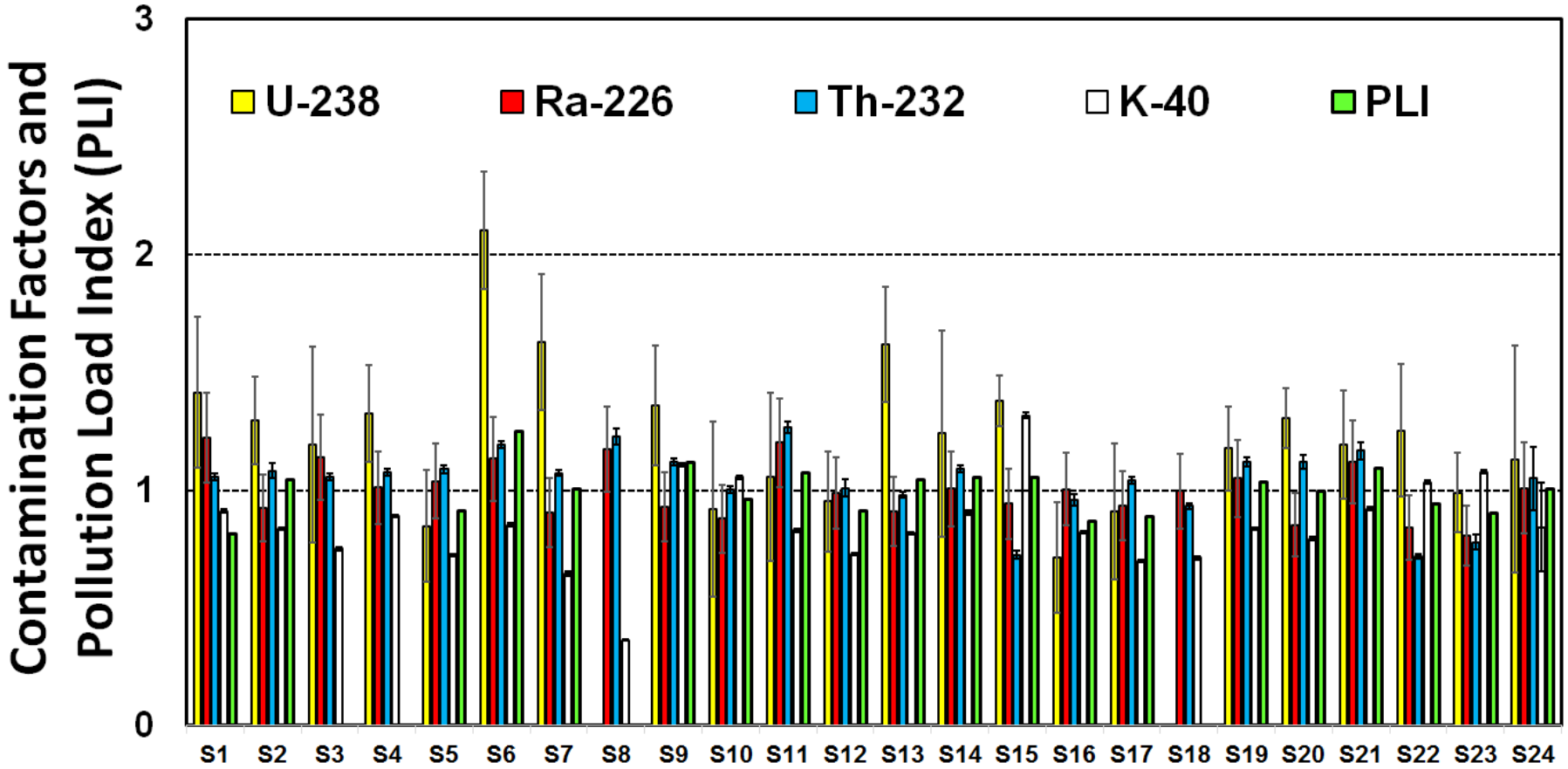

Figure 3.7 Computed contamination factors and the pollution load index (PLI) for radionuclides distribution in Barapukuria soils.

Table 3.10 Summary statistics of computed ra equivalent activity, Ra_{eq} ($Bq.kg^{-1}$), external hazard index, H_{ex}, absorbed gamma dose rate, D ($nGy.h^{-1}$); annual effective dose rate, E ($mSv.y^{-1}$); excess lifetime cancer risk, ELCR (Sv^{-1}) in the soil, feed coal and fly ash samples of this study along with the literature data for soil sample around CTPs.

Radiological Index		Ra_{eq}	H_{ex}	D	E	ELCR
This study						
Soil						
	Mean	249.5	0.67	114.2	0.20	0.00049
	SD	21.7	0.06	9.4	0.02	0.00004
	RSD	8.7	8.7	8.3	8.26	8.3
	Median	250.7	0.68	114.6	0.20	0.00049
	Minimum	201.8	0.54	94.0	0.17	0.00040
	Maximum	293.6	0.79	133.6	0.24	0.00058
Background		250.5	0.68	115.3	0.21	0.00050
Coal		95.6	0.3	42.7	0.08	0.00018
Fly ash		573.9	1.6	256.6	0.46	0.00110
Recommended limits[a]		**≤370.0**	**≤1.0**	**60 (10-200)**	**0.50 (0.3-1.0)**	**0.00029**
Literature data						
NW Bangladesh[c]		426	1.18	203	0.24	-
India[d]		-	1.0	178.4	0.22	-
China[e]			0.49	86.6	0.11	-
China[f] (granite area)		266	0.84	124	0.15	-
Turkey[g]		138.8	0.38	68.65	0.08	-
Hungary[h]		-	-	89.2	-	-
Greece[i]		-	-	57	0.08	-

SD: standard deviation; RSD [%]: relative standard deviation; NW: north-west.
[a]UNSCEAR, 2000, [c]Hamid et al., 2002, [d]Mishra, 2004, [e]Liu et al., 2015, [f]Yang et al., 2005, [g]Cevik et al., 2007, [h]Papp et al., 2002, [i]Papaefthymiou et al., 2013.

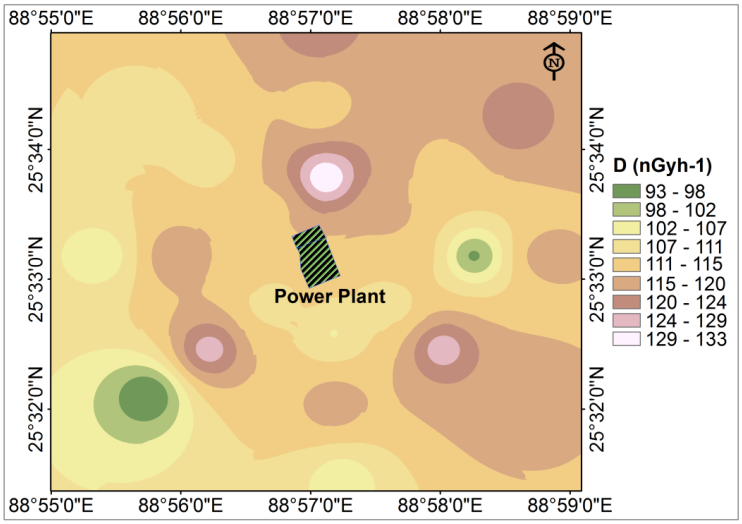

Figure 3.8 Estimated dose distribution in soil around BTPS.

CHAPTER 4

CONCLUSIONS

4.1 Major findings

Based on the activity concentrations of the radionuclides of the coals, CCRs and soil samples examined mineralogical study of soils and evaluation of several environmental and radiological indices of this study, we would like to draw the following conclusions:

For coal, the averages (ranges) of activities of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K 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 CCRs samples in this study are considerably higher than the world soil, Barapukuria soil, earth crust average values. The specific activity of ^{238}U, ^{226}Ra, and ^{232}Th in feed coal is 3.10 to 3.37 times lower than in CCRs 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, if no proper regulation or law are not considered. 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 ($mSv.y^{-1}$) 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} (UNSCEAR, 2000). The estimated various radiation exposure indices values indicate a potential risk of ionizing radiation exposure.

Radioactivity concentrations in soil around the Barapukuria Coal-based power plant are normally distributed and no spatial variations of NORMs have been observed. Both anthropogenic (feed coal and coal fly-ash) and geogenic (mineralogical study) contributors are considered to assess the natural radioactivity. Along with these (anthropogenic and geogenic contributors) experimental findings, environmental indices, such as contamination factor (CF), pollution load index (PLI), different activity and elemental ratios, and previous literature and

natural background soil radioactivity data have been implemented to evaluate the occurrence and source of radionuclides and the level of natural radioactivity of our studied area. No spatial variation of radioactivity around the power plant, study of anthropogenic and geogenic contributors of radionuclides and evaluation of environmental indices reveal that Coal-based power plant introduces insignificant effect on radioactivity concentrations of soils around the power plant. Evaluation of radiological hazard indices invoke for trivial radiological risk from soils power plant surroundings. Thus, this study illustrates that in terms of radioactivity concentrations there is no additional pressure on the soil environment due to the operation of power plant.

4.2 Recommendations

The following recommendations for possible future environmental impact investigations and human health hazards risk identification are made. Firstly, the findings of this research may use in assessing public radiation doses and in monitoring in the trend of environmental radioactivity as reference data for future academic research and also for implementing the new coal power plant projects. This information also could be applicable to the effective CCRs disposal practices and management approaches and also possible safe utilization. It can use to reduce the degree of potential environmental radiological threat and detrimental health hazard risk. The present studies can reinforce the environmental legislation and proper implementation of remedies.

4.3 Future research

The following issues could be considered for future research:

1. Pb, C, S, B, Sr isotopic characterization of coals and associated CCRs for environmental studies.
2. Leaching and toxicity characteristics of hazardous trace elements in coals and associated CCRs and mode of occurrence of elements in coals, CCRs and soils. Toxic heavy metals, organic pollutants (e.g., polycyclic aromatic hydrocarbon) in coals, fate and behavior of those elements during combustion and in other environmental samples from around power plant and coalfield.
3. Mineralogical and geochemical studies of coals and associated CCRs and its relationship with organic matter and relationship between organic matter and trace elements in coals. Molecular structural study of coals and CCRs.
4. Distribution of rare earth elements in coals and CCRs and potentiality assessment of rare earth elements and other precious elements extraction from CCRs residues.

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APPENDIX A

Journal Paper I (Published)

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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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APPENDIX B

Journal Paper II (Published)

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Distribution of naturally occurring radionuclides in soil around a coal-based power plant and their potential radiological risk assessment

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Abstract: Coal-fly-ash is one of the major byproducts of coal-based power plant in which naturally occurring radioactive materials (NORMs) are drastically enriched compared to those of feed coals. Thus, improper management of fly-ash may introduce additional radioactivity to the surrounding environment and cause radiological risk. So, in order to study the distribution of radionuclides in soil around a coal-based power plant and to evaluate their radiological risk, soil, coal and fly-ash samples were analyzed by using a HPGe detector for U-238, Ra-226, Th-232 and K-40 radioactivity concentrations. Furthermore, soil minerals were also studied by X-ray diffractometer to assess the mineralogical provenance of the radionuclides. Mean radioactivity concentrations (in $Bq \cdot kg^{-1}$) of U-238, Ra-226, Th-232 and K-40 in soil samples are 102.9 ± 41.4, 63.6 ± 7.4, 103.4 ± 13.9 and 494.2 ± 107.5, respectively which are comparatively higher than the typical world mean value. Elevated levels of radioactivity are likely due to the presence of illite, kaolinite, monazite, rutile and zircon minerals in the soil samples rather than technogenic contributions from the power plant. Furthermore, mean soil contamination factor (CF) are close to unity and mean pollution load index (PLI) is below unity while the average radium equivalent activity (Ra_{eq} in $Bq \cdot kg^{-1}$), external hazard index (H_{ex}), absorbed γ dose rate (D in $nGyh^{-1}$), annual effective dose rate (E in $mSv \cdot y^{-1}$) and excess lifetime cancer risk (ELCR in Sv^{-1}) are 249.5 ± 21.7, 0.67 ± 0.06, 114.2 ± 9.4, 0.20 ± 0.02, $4.9 \times 10^{-4} \pm 0.4 \times 10^{-4}$, respectively, which are within the permissible limit. Thus, in terms of radioactivity concentrations and associated environmental and radiological indices, the effect of the power plant is insignificant.

Keywords: Soil, radionuclides, X-ray diffractometer, HPGe detector for γ ray spectrometry, coal-based power plant.

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1 Introduction

Radiation and its potential hazard to the human health has become a serious civic concern throughout the world, even though it is an inevitable part of environmental materials (e.g. soil, water) [1]. The sources of continuous

APPENDIX C

Table C1 Salient features of Barapukuria Coal-fired thermal plant (BTPS) and Barapukuria coal field (BCM)

Barapukuria Coal-fired thermal plant (BTPS)	
Plant type	Subcritical
Stack (Chimney) height	100 m
Emission control device (ECD)	Electrostatic precipitators (ESP) (Efficiency ≥ 99 %)
Yearly coal consumption	0.72 million tons (Mt)
Yearly ash production	0.08 MT (~12 to 14 % ash produced of feed coal)
Unit	2 (2 x 125)
Capacity	250 Megawatt (MW)
Year of operation	2005 (first and the only coal-based thermal plant)
Ash pond	1 (capacity about 0.183 million tons)
Fuel source	Barapukuria underground coal mine (BCM)
New unit	275 MW (operated in 2017)
Barapukuria coal field (BCM)[a]	
Commercial extraction	2005
Coal extraction method	Underground mining
Number of coal seams	6
Depth range of coal seams	118-518 m (below surface)
Coal resource	377 Mt
Yearly coal production	1 Mt
Thickness range of coal seam VI	21.63-42.37 m
Coal type	High-volatile bituminous B rank
Age of coal	Permian
Depositional mode	Peat swamp flood basin (terrestrial origin)
Basin area	6. 68 km^2
Paleovegetation	Coniferous gymnosperms (herbaceous plants)

[a] Bakr et al., 1996.

Table C2 Properties of coal (seam-VI) from Barapukuria, Bangladesh (Bakr et al. 1996). All values are in %, except for calorific value (kcal). Average and ranges are in parenthesis. Abundances of major elemental oxides in analyzed coal ash are presented in wt.% in dry weight basis (Islam and Hayashi 2008)

Moisture	**Ash**	**VM**	**FC**	**Carbon**	**Hydrogen**	**Nitrogen**	**Oxygen**
4.11 (3.1-5.3)	16.37 (8.9-28.3)	30.27 (25.8-33.1)	53.36 (45.9-62.1)	83.16 (81.83-84.58)	5.1 (4.95-5.23)	1.63 (1.59-1.76)	9.52 (8.14-10.68)
PS	**SS**	**OS**	**TS**	**Vitrinite**	**Liptinite**	**Inertinite**	**MM**
0.24 (0.07-0.41)	0.02 (0.01-0.07)	0.23 (0.04-0.42)	0.59 (0.52-0.64)	35.6 (26.4-47.6)	6.41 (3.7-8.9)	57.99 (48.7-66.7)	10.3 (6.7-14.5)
VR	**CV**	**TOC**	**pH**	**SiO_2**	**Al_2O_3**	**Fe_2O_3**	**TiO_2**
0.73 (0.71-0.78)	6603.88 (5546-7202)	63 (50-76)	4.7	51.49 (47.99-59.59)	36.55 (30.65-40.09)	4.65 (0.95-9.54)	3.16 (2.75-3.48)
CaO	**K_2O**	**P_2O_5**	**SO_3**	**MnO**			
0.93 (0.5-1.24)	0.78 (0.59-0.99)	0.61 (0.30-0.93)	0.07 (0.02-0.16)	0.16 (0.05-0.32)			

VM: volatile matter; FC: fixed carbon; PS: pyrite sulphur; SS: sulphate sulphur; OS: organic sulphur; TS: total sulphur; VR: vitrinite reflectance; CV: calorific value; MM: mineral matter; TOC; total organic carbon**.**

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