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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

radiation exposure are mainly terrestrial, cosmogenic, anthropogenic (e.g. technogenic, nuclear test). External γ radiation originates from soil which contains the great number of decay products of Th and U series and K-40 [2]. These radionuclides are dispersed in soil only after different slow natural processes. The level of γ radiation is directly related with the specific activity of radionuclides in soil [3]. Additionally, a significant quantity of natural radionuclides can be liberated with fly-ash into surrounding environment due to the operation of coal-burning power plants [4]. However, there is an uncertainty to how much the radiological influence and radioactivity impact on human health in the operation of a coal-burning plant. Thus, it is necessary to determine the level of the natural radioactivity and its influence on environment and human health around the coal power plant.

The coal fuel is combustible solid, considered as the primary nonrenewable energy and contributes about 40.8 % of world's electricity [5]. Inherently, coal contains naturally occurring radioactive materials (NORMs) (e.g. U-238) along with other trace metals as impurities [6]. The coal-based thermal power plants (CTPs) are considered one of the major non-nuclear sources of technogenic radionuclide pollutants [7], which are of most concern due to their radiotoxicity. CTPs produce large proportion of ash residuals which are highly enriched with terrestrial NORMs. Ash residuals then escape and are distributed into the atmosphere and biosphere, dispersed over a wide range of distance due to atmospheric convection [8] and settled on the soil surface [3]. Consequently, these residuals modify the natural background of radioactivity levels, total radioactive dose and chemical composition of the soils [9], where they can be adsorbed, retained and taken up by agricultural plants/crops [10] and affect the fragile ecosystem [cf. 11]. The major route for radioisotopes exposure to humans is through cultivated soil–crop–food pathways [cf. 1, 12], and finally leads to various potential acute and chronic diseases (e.g. cancer) [cf. 6, 13].

Investigations of natural radionuclides have received great attention throughout the world. The interest of studying the radiological impacts of CTPs began in the 1960s and elucidated that the coal combustion could represent a considerable addition to natural radiation doses [14]. Nowadays, many researchers have focused their attention on soil contamination due to the coal burning and revealed non-significant radioactive influence [6, 7, 15–17], whereas other studies have illustrated an evident increment of natural radionuclides concentration in soil around CTPs [cf. 1, 11, 18–28], largely due to the unusually high concentration of U-238 (and Ra-226) in feed coal.

Barapukuria coal-based thermal plant (BTP) has been in operation since 2005. It is the first and the only coal fueled subcritical type power plant in Bangladesh. Two units are operated (250 MW) since its inception, and currently a third unit is started, 275 MW (2017). Coal fuel type is principally Bituminous high volatile ranking coal (Permian Gondwana coal) and derived from local Barapukuria coal underground mine. Filtration system is electrostatic precipitator (ESP) with efficiency more than 99.5 %. Annual coal consumption is approximately 0.72 million tons (Mt) and ash production nearly 0.08 MT (~12–14 % ash produced such as fly-ash (FA), bottom ash (BA) of the total feed coal) [29]. With the rapid growth of number of coal power plants in the country, the radiation exposure from coal burning has become a great concern for environment. However, in Bangladesh, as yet no study has been systematically performed and properly addressed, considering radiological impact on soil environment around BTP so far. Thus, it is of great concern to examine the radiological impact on the soil environment around BTP and for the control of natural radioactivity.

The prime aims of the present work are focused on (1) to determine activity concentrations of NORMs in order to assess any influence on the soil background radioactivity level of BTP operation; (2) to study the soil mineralogy for evaluating the NORMs provenance, (3) to assess the soil contamination status with the NORMs; and (4) to estimate radium equivalent activity (Ra_{eq}), absorbed γ dose rates in air (D), the annual effective dose rate (E), external hazard index (H_{ex}) and excess lifetime cancer risk (ELCR) in order to evaluate any excess radiological risk for the immediate inhabitants.

2 Experimental

2.1 The study area

The area is an agricultural (cultivated) dominant farming area with double/triple cropped and moderate to densely populated (density: 823/km^2) land in northwest part of Bangladesh (Figure 1). The area is situated at the northern fringe of a Pleistocene terrace named the level Barind Tract (about 30 m above the mean sea level) and in humid subtropical region in Alluvial-fluvial floods plain system. The available data indicate that the prevailing dominant local wind directions from east to west (40 %) succeeded by west to east (25 %) and north-east (18 %) and rarely exceed the speed 8 m/s [30].

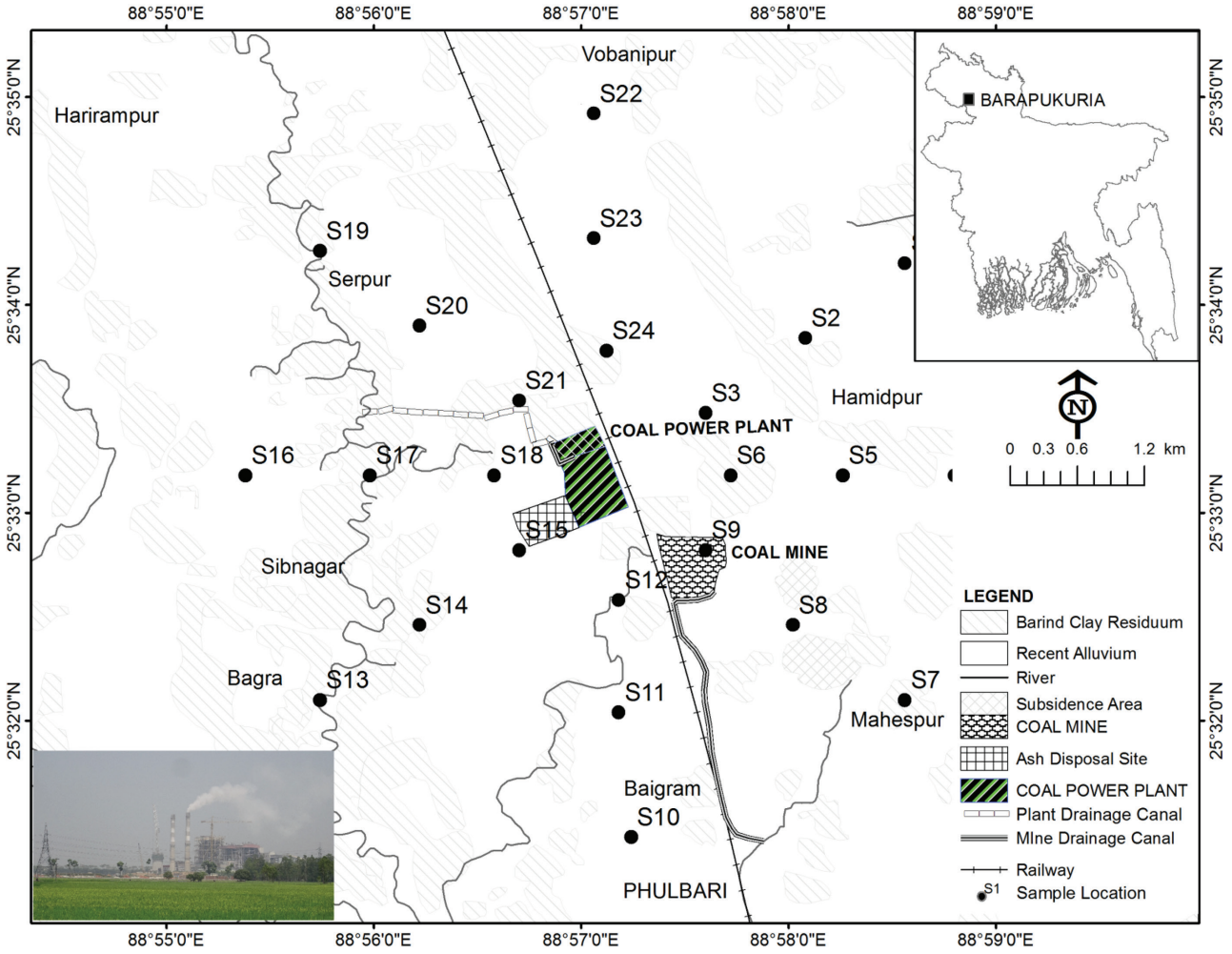

Figure 1: Map of Bangladesh showing sampling sites where the soil samples were taken and Barapukuria coal-based powerplant (BTP) area.

2.1.1 Geological characteristics of the area

Tectonically, the area (Barapukuria coal basin) lies within the Rangpur Saddle and 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 [31, 32] (Figure 1). 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 [32]. The Pleistocene sediments is underlined by the Plio-Pleistocene Dupi Tila formation [33]. 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 [34].

2.2 Sampling and preparation procedure

A total number of 24 soil samples from the vicinity of the BTP were collected carefully, up to 10 cm depth. Each site was logged by a global positioning system (Figure 1) during the dry month of April, 2017. The samples were taken following systematic random sampling protocol (IAEA 2004) [35], 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), feed coal (FC) (n = 3), fly-ash (FA) (n = 3) 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 BTP, 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, weighed, and dried to remove the moisture content in a temperature-controlled furnace at 110 °C until constant weight prepared for analysis [6]. Proper care was taken to avoid the cross contamination during sampling, sample preparation and measurement.

2.3 Sample analysis

2.3.1 Measurement of soil 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 $mL \cdot g^{-1}$ by using pH meter and dichromate digestion based on Walkley–Black method, respectively [36, 37]. 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 following Bowles [38] protocol.

2.3.2 Estimation of NORMs

The samples were packed in a vessel U8, mass weighed and then hermetically sealed with black electrical tape to prevent the loss of the radionuclides in gaseous form radon (Rn) [39] and stored at least for 4 weeks to attain the secular equilibrium between the long half-life parent and the short half-life daughter radionuclide prior to being measured. This procedure was also followed by many researchers around the world [cf. 6].

The soil, feed coal (FC), and FA samples were investigated for the activity measurements of the radionuclides U-238, Ra-226, Th-232, and K-40 indirectly by means of γ ray spectrometry with a low background Germanium semiconductor detector, GEM 30-70, ORTEC in the Radioisotope Center (RI), Hiroshima University, Japan at 0 cm distance from the detector. The correction of sum effect which influences counting accuracy of multiple γ ray emitting radionuclides was considered in radioactivity calculation. Detection efficiency calibration of γ spectrometer was conducted by using the set of standard sources (MX033U8PP) that consist of radionuclides with known radioactivity emitting from low to high energy γ ray manufactured by Japan Radioisotope Association (JRA). The set of standard sources that were used with different thicknesses, which were 5, 10, 20, 30 and 50 mm to consider the geometry of a sample. In principle, the radioactivity measured by using the γ ray spectrometer with decay correction was compared to initial radioactivity of the standard source measured by JRA to obtain the detection or counting efficiency. The γ ray spectrometer counting efficiency for NORM was estimated by using curve fitting of energy and counting efficiency, and furthermore, counting efficiency and thickness of sample to consider the geometry of measurement.

The screening measurement was conducted to analyze whether the radionuclides that are important in NORM analysis can be detected in the sample. In screening process, the background, a coal, ash and soil samples were measured and compared. The background was measured in 3.82 days, coal sample about 60 g in 1.83 days, soil sample about 10 g in 4.95 days and FA sample about 80 g in 2.32 days. The measurement time depended on the number of γ energy emission counting that is statistically representative. Quoted uncertainties (the confidence level of 1σ) were calculated by error propagation calculation. The combined standard uncertainties included the efficiency calibration uncertainty and the statistical uncertainties of the recorded peaks.

Activity concentration of NORM was calculated by considering net count, counting efficiency and emission rate of certain radionuclides and weight of sample, as in the following Eqs. (1) and (2):

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

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

where, A is Activity (Bq); AC, Activity concentration ($Bq \cdot kg^{-1}$); cps_{sample}, Counts per second of sample (s^{-1}); cps_{BG}, Counts per second of background (s^{-1}); ε, Counting efficiency of the Ge detector; I_{γ}, Intensity of γ ray; w, Sample weight (kg).

The radionuclides of concern in this measurement are long half-life radionuclides including Th-232, U-238, U-235 and Ra-226 and were estimated based on the activity concentration of γ rays of their progenies in samples, except for K-40 that can be measured directly. Activity concentrations of U-238 and Th-232 were determined indirectly by analyzing full-energy peaks emitted by their progenies. In Th-232 decay series, Ac-228, Tl-208, Pb-212, Bi-212 were used to estimate Th-232 [40]. In U-238 decay series, Pb-214 and Bi-214 were

used to estimate Ra-226. Activity of Ra-226 was calculated from average value of activity of Pb-214 and Bi-214 on four peaks. In U-235 decay series, U-235 which emits a γ ray at 185 keV is detected with overlapping 186 keV of Ra-226. The activity of U-238 was calculated by natural abundance ratio of U-235 and U-238. The abundance of U and Th in ppm, and K (%) were calculated from activity concentration by a factor of 12.35, 4.06, and 313, respectively [41].

2.3.3 Mineralogical study

2.3.3.1 Optical microscope

Dried bulk soil samples were grounded, homogenized, coned and quartered to attain representative samples for slide preparation. Firstly, some Canada balsam was placed on a glass slide and kept it on the heater. When it was warmed and transformed into solution, then samples were placed on the Canada balsam solution. Subsequently a glass cover was placed on this solution and was pressed the cover until the extra Canada balsam and bubble removed. After cooling the slide, it was cleaned using xylene and cotton and was ready for microscopic study. 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) [42].

2.3.3.2 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 [43]. The selected quarter samples were prepared as powder mounts approximately 1.0 g and packed into a steel cavity mount suitable for insertion into the X-ray diffractometer. The samples were compacted on the sample holder to obtain a uniform surface, required for this technique [44]. X-ray diffraction patterns were recorded using a diffractometer with Cu-Kα radiation and with a scan range (2θ) of 2–90°, step size (2θ) of 0.05° and counting time of 1 s 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). The analysis was performed at the Scientific Equipment Centre (SEC), Prince of Songkla University, Thailand.

2.4 Soil contamination indices

In order to determine the degree of contamination due to operation of the BTP, the contamination factor (CF) [45] and pollution load index (PLI) [46] were calculated. The CF is the ratio obtained by dividing the activity concentration of each radionuclide by the natural background activity concentration in soil. Based on their intensity, CFs may be classified into four categories: $C^i_f < 1$, low; $1 \le C^i_f \le 3$, moderate; $3 \le C^i_f \le 6$, considerable; and $C^i_f \le 6$, 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 suggests the baseline level of contaminants while more than unity refers to the gradual degradation of soil health [cf. 26, 47].

2.5 Radiological hazard parameters

The Radium equivalent activity, Ra_{eq} ($Bq \cdot kg^{-1}$), is related to the external and internal γ dose due to Rn and its daughter. To compare the combined radiological effect of Ra-226, Th-232 and K-40 in the materials, the Ra_{eq}, were calculated by the Eq. (3) [48]. The maximum value of Ra_{eq} in building construction materials must be less than 370 $Bq \cdot kg^{-1}$ for safe use.

$$\text{Radium equivalent activity, } Ra_{eq} = C_{Ra} + 1.43\, C_{Th} + 0.077\, C_K \le 370 \quad (3)$$

where, C_{Ra}, C_{Th} and C_K are the average specific activity of Ra-226, Th-232 and K-40 in $Bq \cdot kg^{-1}$, in the materials, respectively.

External hazard index, H_{ex}, evaluates external radiation exposure from Ra containing materials through the Eq. (4) and the index must be less than unity to be within the safety threshold to avoid radiation hazards to the respiratory system [49].

$$\text{External hazard index, } H_{ex} = C_{Ra}/370 + C_{Th}/259 + C_K/4810 \le 1. \quad (4)$$

The absorbed γ dose rate, D ($nGyh^{-1}$), for power plant staff, miners and the local population, for a uniform distribution of Ra-226, Th-232 and K-40 is computed by the Eq. (5) provided by UNSCEAR [2].

$$\text{Absorbed } \gamma \text{ dose rate, } D = 0.462\, C_{Ra} + 0.604\, C_{Th} + 0.0417\, C_K. \quad (5)$$

The annual effective dose rate, E ($mSv \cdot y^{-1}$), is in air received by adults. The indoor occupancy factor must be taken into account and evaluated by the Eq. (6) UNSCEAR [2].

$$\text{External effective dose, } E = D \times 10^{-3} \times 1.23. \quad (6)$$

The excess lifetime cancer risk (ELCR), caused by the annual effective dose due to external exposure is estimated using the following Eq. (7) [50]:

$$\text{Excess lifetime cancer risk, } ELCR = E \times ALT \times RF \quad (7)$$

where, ALT is the average life time (70 years) and RF is risk factor, fatal cancer risk per Sievert and for stochastic effects, ICRP uses values of 0.5×10^{-4} for the public exposure ICRP [50]. This risk represents the number of extra cancers expected in a given number of people exposed to a carcinogen at a given dose.

2.6 Statistical analysis

For all gathered samples, basic descriptive statistical analysis was performed for radionuclides (variables) in soil samples of different sites using SPSS version 20. Normality test (Kolmogorov-Smirnov) was performed to narrate data frequency distribution. As the data were normally distributed, Pearson correlation matrix was selected for the mutual relationship and association analysis in order to identify the degree of association between pairs variables and describe the interdependency also. 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. It is widely applied for highlighting the spatial variability of interesting variables in the environmental samples of the study area [cf. 3, 26]. An interpolated map for each parameter was generated in the ArcGIS 10.3.

3 Results and discussion

3.1 Physicochemical characteristics of soil

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 [3]. In our study, pH values of the soil samples vary from 4.0 to 6.3 with a mean value of 5.4, which indicates the moderate to strong acidity of the soil samples. Correspondingly, SOM of the soil samples ranges from 0.6 % to 14.4 % with a mean value of 2.6 % (Table 1). According to the USDA scheme [54], particle-size distribution of the samples examined in this study (Table 1) shows that the soil texture spread out from the silt end-member to the loam end-member. 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 Radioactivity concentrations in soil, coal and ash samples

The basic descriptive statistics of radionuclides specific activity in 24 soil samples are summarized in Table 1. Activity of Ra-226 in the soils varied from 51.2 to 77.6 with a mean value of 63.6 ± 7.4, of U-238 from below detection limit to 192.4 with a mean value of 102.9 ± 41.4; of Th-232 from 71.5 to 126.1 with a mean value of 103.4 ± 13.9 and for K-40 from 210.5 to 763.3 with a mean value of 494.2 ± 107.5 (all units are in $Bq \cdot kg^{-1}$), respectively. The highest coefficient of variation of activity is for U (40.3 %) and the lowest for Ra (11.6 %) (Table 1). The specific activities obtained are about 1.3–3.5 times greater than the typical world average value for soil [2]. The measured activity concentrations in soils are within the permissible limit, i.e. 370 $Bq \cdot kg^{-1}$, 259 $Bq \cdot kg^{-1}$ and 925 $Bq \cdot kg^{-1}$ for Ra-226, Th-232 and K-40, respectively [53]. The relative contributions to the total activity in soil samples are in descending order K-40 > Th-232 > U-238 > Ra-226. The frequency distribution of determined activity of radioisotopes follows normal distribution (significant at the 0.05 level) (Table 1).

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

In Table 2, a summary of the measured specific activities of radioisotopes in soil samples of this study along with the literature data [2, 18–20, 55–61] from similar investigations are tabulated. Radioactivity concentrations of Barapukuria soil samples are considerably higher than those of other parts of the world [18–20, 24, 57, 58], except for China [23, 27].

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. In Bangladesh, Th and U

Table 1: Activity concentrations of radionuclides, abundance of radioactive elements in the soils from the surroundings of Barapukuria coal-based thermal plant (BTP) with their associated uncertainties, test of normality (K-S α), and physicochemical properties.

	U-238		Ra-226		Th-232		K-40		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	80.0	15.0
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
CV	40.3		11.6		13.5		21.8	–	24.7	13.5	21.8	9.6	134.8	65.2	12.0	49.1
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]	–	–	–	–	–
Europeaverage[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; CV[%], coefficient of variability; Min, minimum; Max, maximum; [±], 1σ variation due to counting uncertainties.

[a]K-S α, Normality test (Kolmogorof-Semirnov), [b]Present study, [c][2], [d][51], [e][52], [f][53].

content in soil samples range from 16.5 ppm to 19.5 ppm and 2.9 ppm to 3.8 ppm, respectively [62, 63] which are systematically lower than the Barapukuria soil. Along with the Th and U, K content in the Barapukuria soil samples are considerably higher than those of world average soil samples [51], upper continental crust [52] and in sedimentary rock [64] as presented in Table 1.

3.3 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 (BTP). This is confirmed

Table 2: Comparative study of activity concentrations (in Bq · kg^{-1}) of radionuclides among this study and other literature data for samples with similar investigation.

Reference	Type	U-238	Ra-226	Th-232	K-40
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 (14–156)	69.6 18–156	396 (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 (21–43)	–	350 (130–610)

SE, South-East; NW, North-West.
[a][55], [b][56], [c][57], [d][23], [e][27], [f][19, 58], [g][59], [h][18], [i][20], [j][24], [k][60], [l][2].

by employing the one-way ANOVA test (between groups and within groups), which point out that no remarkable differences occur to the total variance of all radioactivity with distance-direction variation (U-238: $F = 1.173$, $p = 0.329$; Ra-226: $F = 0.109$, $p = 0.897$; Th-232: $F = 1.448$, $p = 0.258$; K-40: $F = 0.382$, $p = 0.687$). It indicates that the measured activity values were statistically equal, which is invoking that the influence of the BTP operation is insignificant (Table 3).

A histogram was constructed to evaluate the asymmetry distribution of the activity concentrations of the soil samples (Figure 2). This graphic depiction allows a visualization of the dispersion results in samples, median (–), range of data variation, as well as comparison between different radionuclides. It is found in Figure 2 that the mean concentrations for U-238 and K-40 increase with the distance from the point source (power plant) similar to that of Turkish power plant [65]. It is should be noted here that the K content in Barapukuria soil samples is considerably higher than in coal and fly-ash (Table 1). However, the

Table 3: One-way ANOVA test results for radionuclides activity concentration difference in soil samples from Barapukuria.

	Sum of squares	df	Mean square	F	Sig.
U-238					
Between groups	3967.618	2	1983.809	1.173	0.329
Within groups	35509.347	21	1690.921		
Total	39476.965	23			
Ra-226					
Between groups	12.910	2	6.455	0.109	0.897
Within groups	1240.764	21	59.084		
Total	1253.674	23			
Th-232					
Between groups	539.607	2	269.803	1.448	0.258
Within groups	3914.139	21	186.388		
Total	4453.746	23			
K-40					
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.

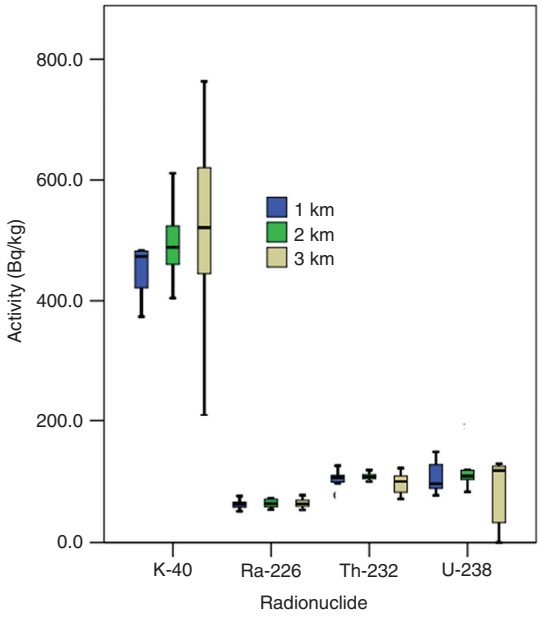

Figure 2: Histogram showing the distribution of the activity concentrations (Bq · kg^{-1}) of radionuclides and their variability in top soils from Barapukuria along with the mean and median (–), range of data variation.

mean concentrations of Ra-226 and Th-232 do not fluctuate with distance.

Figure 3 displays 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 at all sites except southeast and northwestern part of the area (Figure 3a). The Ra-226 spatial activity distribution map exhibits slightly higher value in the southeastern and north-central sides than other parts of the mapped area (Figure 3b). Thorium-232 distribution map shows higher activity all around the plant except in southwest part of the area (Figure 3c). Potassium-40 in soil has a wide variation around the power plant. Its activities are less around the plant but highest in the southwestern and north-central part than in the surrounding areas (Figure 3d). The spatial variability maps do not exhibit any particularly special distribution trend over the area. However, the obtained specific activity values in soils around BTP are virtually close to the mean natural background activity soil (taken

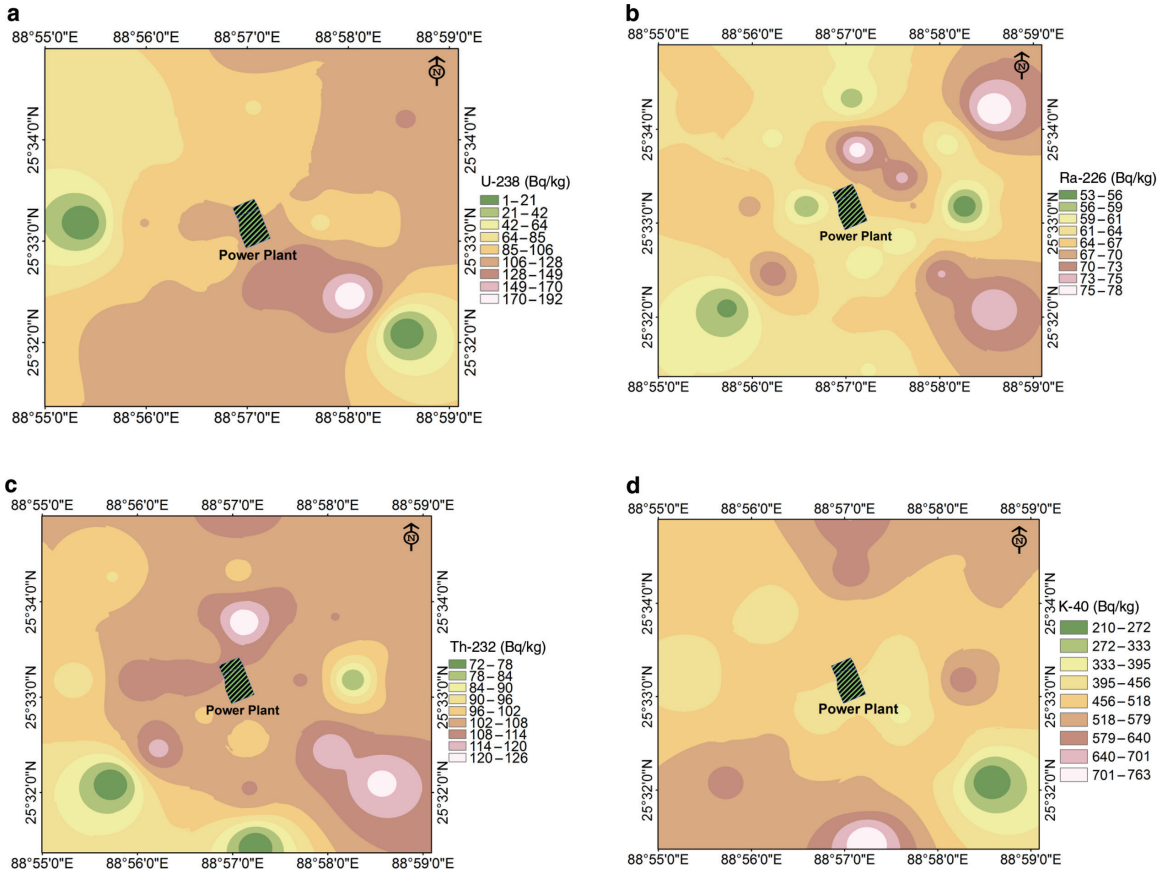

Figure 3: Inverse distance weighting (IDW) map for the spatial distribution of radionuclides activity in the soils from Barapukuria power plant vicinity.
(a) U-238, (b) Ra-226, (c) Th-232, and (d) K-40 activity concentrations in Bq · kg^{-1}.

Table 4: Mutual correlation matrix of radionuclides and soil properties of Barapukuria.

	pH	OM	Sand	Silt	Clay	U-238	Ra-226	Th-232	K-40
pH	1								
OM	0.361	1							
Sand	0.089	−0.329	1						
Silt	−0.215	0.184	−0.385	1					
Clay	0.176	0.000	−0.188	−0.834[b]	1				
U-238	−0.031	−0.502[a]	0.291	−0.653[b]	0.521[b]	1			
Ra-226	0.450[a]	0.316	0.140	−0.242	0.174	−0.097	1		
Th-232	0.376	0.316	−0.049	−0.208	0.250	0.035	0.623[b]	1	
K-40	−0.278	−0.631[b]	0.351	−0.238	0.044	0.431[a]	−0.375	−0.558[b]	1

[a]Correlation is significant at the 0.05 level (two-tailed).
[b]Correlation is significant at the 0.01 level (two-tailed).

at 15 km away from BTP from similar undisturbed soil type) which is concomitant with those of previous literature studies [3, 6, 7, 16, 26, 66, 67].

3.4 Correlation study

In the areas of highly homogeneous lithology, strong correlations among the parameters (e. g. radionuclides) are quite common [68]. So, to draw the provenance of radionuclides and their relationship with the soil physicochemical properties, a Pearson correlation matrix is tabulated in Table 4. U-238 shows positive correlation with K-40 ($r = 0.431$, $\alpha \leq 0.05$), 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 [69]. Then from the clay, U and K are transferred to the soil solution and are available for migration and uptake by the crops/vegetation [70]. Similar, to U-238 and K-40, Th-232 and Ra-226 are also significantly correlated ($r = 0.623$, $\alpha \leq 0.01$) as that of Belivermis et al. [71]. K-40 is inversely correlated with SOM ($r = -0.631$, $\alpha \leq 0.01$) which reveals inorganic mineralogical (e.g. illite) affiliation of K [70] instead of organic association [69]. Similar to the SOM, Th-232 also shows inverse correlation with K-40 ($r = -0.558$, $\alpha \leq 0.01$) which indicates their different geochemical behavior [cf. 69, 70, 72]. 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 [73]. The soil pH was found to be correlated with Ra-226 ($r = 0.45$, $\alpha \leq 0.05$), which indicates that solubility and mobility of Ra increases with increasing soil acidity [7].

On the contrary, non-significant correlations were also found between radionuclides and soils physicochemical properties which can be deciphered over an area of different mineral suites/rock types, differences in geochemical behaviors of radionuclides and attributed to a variety of physicochemical–biogeological properties of soil [68] and/or external inputs (such as flying coal dust, fly-ash) operating in soil environment [71].

3.5 Soil mineralogy and radionuclide provenance

Uranium and Th concentrations in soil are closely related to the parent bedrock of the soil and crystal structure of the associated minerals [74]. So, to evaluate the radionuclide mineralogical provenance Barapukuria soil samples were studied. The major minerals in Barapukuria soils were found to be kaolinite, illite, quartz, Fe-oxides minerals (Figure 4a), and the accessory heavy minerals were also found, e.g. monazite, rutile, biotite, zircon, kyanite, garnet, and tourmaline mineral assemblages (Figure 4b) [42]. Aftabuzzaman et al. [75] demonstrated that kaolinite and illite in Barapukuria soil are volumetrically the most abundant and common minerals, while Alam et al. [32] reported that such type of soil contains dominant proportion of illite (source of K-40) and kaolinite is a minor fraction in the total clay content. Nevertheless, it is well established that clays (e.g. illite, kaolinite) are the main geochemical carriers, concentrator and great repository of radionuclides (such as U-238) in soils [cf. 7, 13, 72, 73], which most likely govern the soil radioactivity. However, U is associated with accessory minerals, such as zircon [76] while the carrier of Th is mainly monazite [cf. 63, 77] which is very resistant to weathering. Along with monazite, a significant portion of Th is also partitioned into

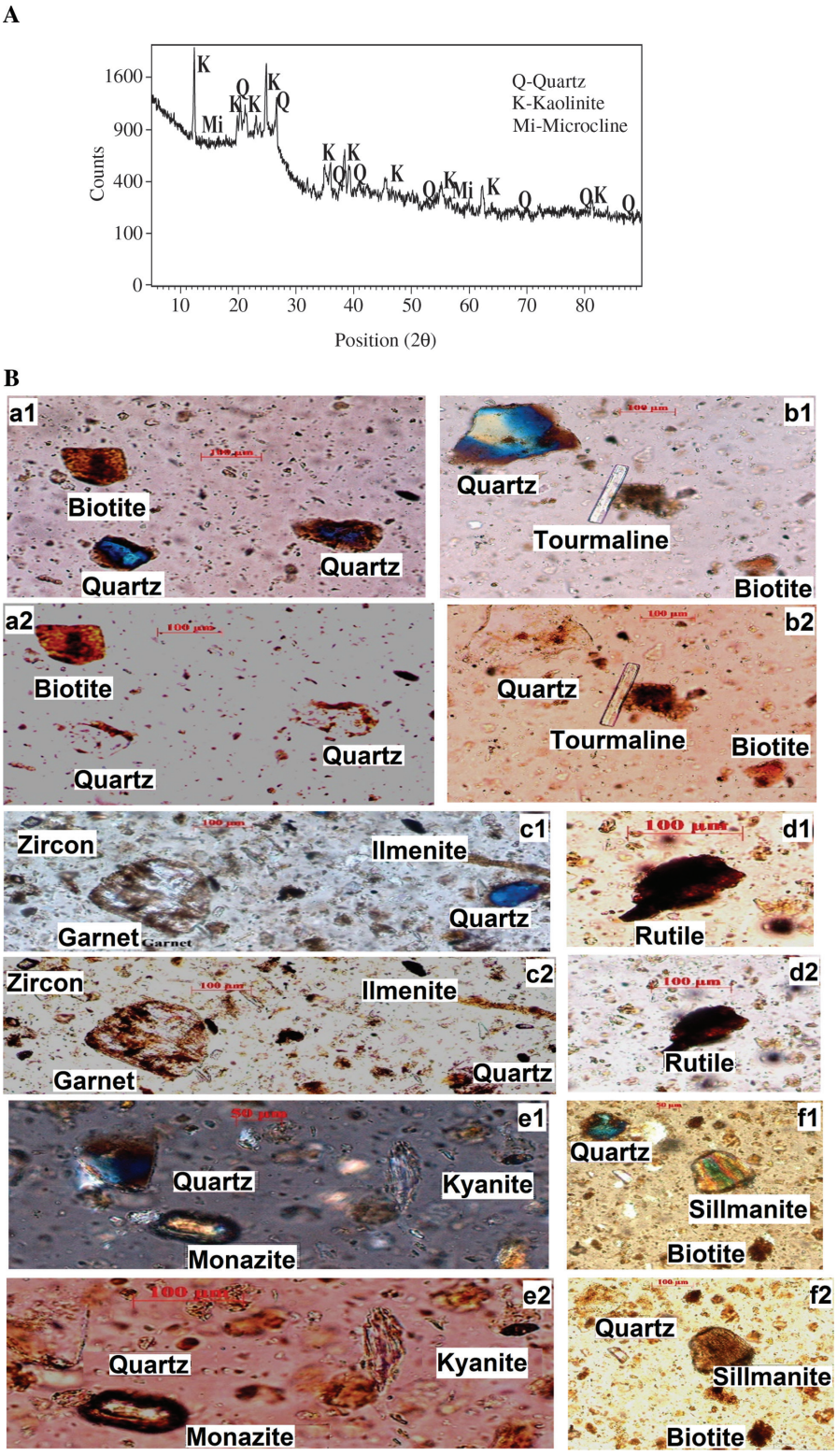

Figure 4: Mineralogical composition of the soil samples from around Barapukuria coal-buring power plant, investigated with XRD and optical microscope.
[A] XRD pattern showing the major indentified crystallite phases such as quartz, microcline, and kaolinite. [B] Photomicrographs of identified accessory minerals in soil samples from Barapukuria coal power plant. (a) biotite, quartz (sample S9); (b) quartz, tourmaline (sample S9); (c) zircon, garnet, ilmenite, quartz (sample S13); (d) rutile (sample S13); (e) monazite, kyanite, quartz (sample S19); and (f) quartz, sillimanite, biotite (sample S19). (All images are in transmitted light both cross and plane polarized view; a1 image indicating cross light view, whereas a2 image representing plane polarized view).

zircon, and clays [cf. 63, 78]. Thus, the elevated specific activity of Th-232in our sample is due to the high content of monazite [79]. Similarly, Alam et al. [80] and Khan et al. [77] found higher radioactivity in the beach soils due to the existence of heavy minerals in soil such as monazite, zircon, rutile, garnet, etc.

The estimated elemental concentrations of U and Th and determined minerals (monazite, zircon, biotite, clays) are matched to the granites mineralogical content and concentrations [70]. Previous studies demonstrated that Th and U rich granite serve as the source of Th and U to the soil [4, 78]. Moreover, Yang et al. [81] found higher radioactivity in soil originating from granite rocks in China. So, we can reasonably assume that the soil parent rocks originated from granitic rock source, which presumably control the soil radioactivity [cf. 74, 78].

Presumably, soil radionuclides concentration is principally determined by the soil minerals, local variability of radionuclides 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 possible enrichment of the soil samples with the radionuclides due to natural dispersion process rather than anthropogenic (technogenic) attribution.

3.6 Assessment of soil contamination level

In order to determine the degree of contamination due to the operation of BTP, 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 1. Calculated mean values for CFs for topsoil were –0.85, –1.04, –1.13, and –1.0 for K-40, Th-232, U-238, and Ra-226, 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 5). Similar to Ćujić et al. [26], we can reasonably infer from the estimated indices that the soils are uncontaminated with radionuclides, except for K-40.

The activity ratio reflects the relative depletion or enrichment of radioisotopes in the geo-environmental materials [6] which can be employed as indicator of the radioactive pollution of the soil samples. The world mean ratio of U-238/Th-232 in soil is close to unity [2]. The ratios between daughter and parent radionuclides is not unity, indicating disequilibrium within the U and Th decay subseries, and therefore the existence of contamination [82]. The value of the Ra-226/U-238 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 U-238/Th-232 and Th-232/Ra-226 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 near to the UCC [52] ratio values (Ra-226/U-238: 0.94, U-238/Th-232: 0.81, and Th-232/Ra-226: 1.3, respectively). The Th-232/Ra-226 ratio value in soil samples suggests that the original proportionality is preserved. The calculated ratios are analogous to the other indicated values in literature [82, 83].

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 [52] and for

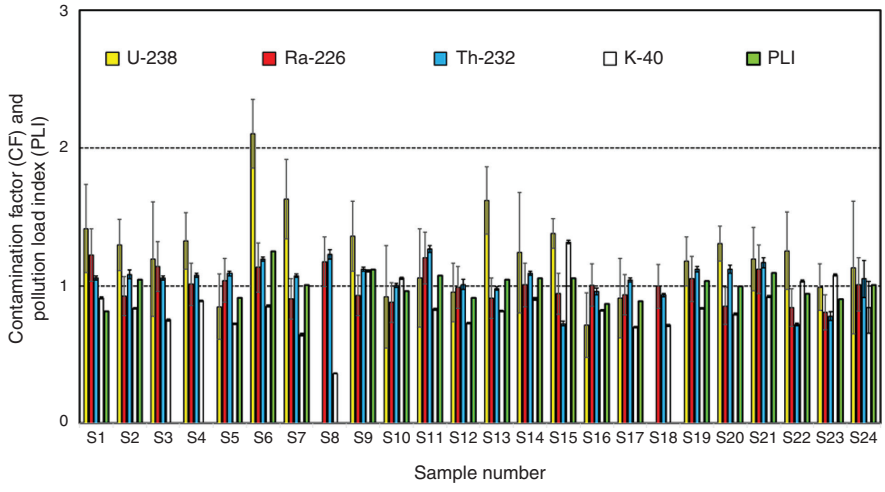

Figure 5: Computed contamination factors and the pollution load index (PLI) for radionuclides distribution in Barapukuria soils.

Table 5: Summary statistics of computed Ra equivalent activity, Ra_{eq} ($Bq \cdot kg^{-1}$), external hazard index, H_{ex}, absorbed γ dose rate, D ($nGy\ h^{-1}$); annual effective dose rate, E ($mSv \cdot y^{-1}$); 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 coal based power plants.

Radiological indices	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**[b]
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][2], [b][50],[c][56] ,[d][57], [e][27], [f][81], [g][61], [h][20], [i][7].

sedimentary rocks they are 4.14, 13571.43 and 0.00031, respectively [64]. The ratio K/U is highly variable and may suggest provenance from different sources. The obtained ratio value for Th/K is significantly higher compared to UCC and sedimentary rock but other two ratios are very close. 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 [84, 85], except for K [86].

3.7 Radiological hazard assessment

The summary statistics of radiation indices are presented in Table 5. The deduced values of radium equivalent activity, Ra_{eq}, range from 201.8 to 293.6 $Bq \cdot kg^{-1}$ with mean value of $249.5 \pm 21.7\ Bq \cdot kg^{-1}$, which are far below the internationally accepted values (370 $Bq \cdot kg^{-1}$, respectively) [2]. The values of hazard index, H_{ex}, vary from 0.54 to 0.79 and the mean value is 0.67 ± 0.06. Since these values are below the threshold limit of unity, soils of this study are quite safe from radiological harmful effect according to Krieger [49].

In our study, the corresponding average absorbed γ dose rate, D, is found to be 114.2 ± 9.4 with intervals of 94.0–133.6 $nGyh^{-1}$, which are about 1.9 foldss higher than the quoted global average value (60 $nGyh^{-1}$) for the public but within the world ranges (10–200 $nGyh^{-1}$) [87]. It is concomitant with the other reported values and average dose rates in different countries ranging from 57 to 203 $nGyh^{-1}$ [2, 7, 8, 20, 27, 56, 57, 59, 88–90].

The total annual effective dose, E, ranges from 0.17 to 0.24 with an average value of 0.20 ± 0.02, which is lower than the world average value 0.5 $mSv \cdot y^{-1}$ (recommended upper dose limit of 1.0 $mSv \cdot y^{-1}$ [50, 91]. The relative contributions to total external γ dose rate in air from Th-232, K-40 and Ra-226 are in the decreasing order 75.3 %, 23.6 %, and 1.0 %, respectively, where Th-232 is the major contributor in the studied area. Finally, the values of the excess lifetime cancer risk vary 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} [2]. 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 comes from K-40 (63.9 %), it can be seen that spatial variability of dose is mainly controlled by the concentrations

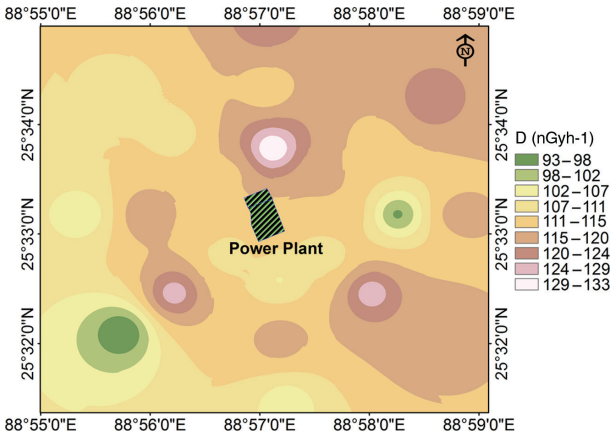

Figure 6: Estimated dose distribution in soil around the Barapukuria thermal power plant.

of Ra and Th (Figure 6). Hence the isolevel maps follow to a great extent the distribution pattern of these two radionuclides.

4 Conclusion

Systematic studies on the distribution of radionuclides and their associated health hazard have been evaluated around an area of a thermoelectric power plant. Depending upon the radioactivity concentrations, mineralogical study and evaluation of several environmental and radiological indices of this study, we can draw the following conclusions:

1. Radioactivity concentrations in soil around the Barapukuria Coal-based power plant are normally distributed and no spatial variations of NORMs have been observed.
2. A study of anthropogenic and geogenic contributors of radionuclides and an evaluation of environmental indices reveal that Coal-based power plant introduces insignificant effect on radioactivity concentrations of soils around the power plant.
3. Evaluation of radiological hazard indices, such as radium equivalent activity, external hazard index, absorbed γ dose rate, annual effective dose rate and excess lifetime cancer risk invoke for trivial radiological risk.

Thus, this study illustrates that in terms of radioactivity concentrations there is no additional pressure on the soil environment due to the operation of Coal-based power plant, nevertheless we also need to consider the duration of the plant's operational activity (~12 years), efficient efficiency of the filtration system [92], and local climatic and flood characteristics.

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