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Natural Radioactivity Levels and Radiological Health Implications around the Mining Area of Bangladesh

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Abstract: *The activity concentrations of natural radionuclides in 6 different samples collected from the Barapukuria coal mine of Bangladesh are presented. The samples were analyzed by gamma spectrometry with low background HPGe detector. The activity concentrations of ^{238}U, ^{232}Th and ^{40}K in all the measured samples varied from 1.35 to 348.19 Bq/kg, 7.97 to 770.81 Bq/kg and 19.28 to 936.65 Bq/kg, respectively. Also were determined the radium equivalent activity, external hazard index, absorbed gamma dose rates and the annual effective dose rates to evaluate the radiological hazards due to these natural radioactivity. The present results were compared with those of other countries and the world average. The mean activity concentrations and all hazard indices were found to be within the safely limits.*

Keywords: *Natural radioactivity, Mining area, Gamma spectrometry, Hazard indices.*

1. INTRODUCTION

Human beings are continually being exposed to ionizing radiation from natural sources. This exposurecan either be external or internal. The major contribution to external exposure comes from gamma-emitting radionuclides present in trace amounts in soil and sediment, mainly from ^{238}U, ^{232}Th and ^{40}K and their progeny. Internal exposures arise from the intake of terrestrial radionuclides by inhalation and ingestion. The dominant components of exposure to inhalation are the short-lived decay products of radon, while the doses from ingestion are due to ^{238}U and ^{232}Th decay series and ^{40}K present in food and water [1].

Coal mining itself gives rise to a potential exposure of natural radioactivity. Coal can be mined in either open pits or underground mines, and produces a significant amount of waste rock, and drainage water that can present with elevated levels of radioactivity. Population exposure could also occur when soil, sediment or rock from the mining sites are used as components of building materials. The health risk to human due to the radionuclides in tailing could increase significantly when these materials accumulate in farmlands [2, 3].

Bangladesh has a good amount of coal deposit in its territorial area. It has five discovered coal mines namely Barapukuria, Phulbari, Khalashpir, Jamalganj and Dighipara. But only the Barapukuria coal mine is currently under production. The coal extracted from this mine is used in power generation, burning in the brick fields and so on. The water originating from the coal mine spreads coal to the adjacent crop fields. Besides, through small rivers, the coal spreads to different districts and their vicinity. This coal mixed water used in various purposes in these regions may affect the coal mine workers, public and the environment [4, 5]. It is, therefore, worthy of investigating the radioactivity level of the natural radionuclides for the safety of the general public [6, 7].

In recent years, many experimental groups [8-14] have paid their attention in measuring the activity concentrations of natural radionuclides intailing enriched soil and sediment samples. The concentration of natural radionuclides in tailings varies significantly from one country to another and even from one place to another in the same country.The assessment of radioactivity levels in different samples in every mining site are essentialto control the environmental pollution. However, for Bangladesh, there is only one experimental data [15] available in literature regarding this matter.Therefore, the present study was aimed to conduct a thorough study on the radionuclide contents and radiological implications on the coal mine workers, public and environment due to the mining in Bangladesh.The present results have been compared with the available experimental data.

The rest of this paper has been organized as follows. Section 2 outlines the experimental procedure. In section 3, we have presented and compared our results systematically with the available reported data. Section 4 contains the conclusion on the present study.

2. MATERIALS AND METHODS

2.1 Study Area

The study area, Barapukuria coal mine, lies in the administrative district named Dinajpur in the Northern part of Bangladesh. Dinajpur is bounded by the districts Thakurgaon and Panchagarh in the north, Gaibandha and Joypurhat in the south, Nilphamari and Rangpur in the east and the state of India in the west. The Barapukuria coal mine is situated approximately between the latitude 25˚33'15" to 25˚34'15" N and longitude 88˚57' to 88˚59' E.

2.2 Sample Collection and Preparation

A total of 12 samples namely 3 coal, 1 soil,1 tailing, 5 water, 1 grass and 1 herb were collected from in and around the coal mine area. Three coal samples were collected from the three sides of the coal deposit which were marked as the sample code: C1, C2 and C3. Soil, tailing, grass and herb samples were marked as S, T, G and H, respectively. Among the water samples, 2 were mine water (marked as MW1 and MW2), 2 were drinking water (marked as DW1 and DW2) and rest one was surface water (marked as SW) from inside the mine.

The collected solid samples were packed in dried polyethylene bags and water samples were in marinelli type beakers. The bags and beakers were then labeled with sample codes and sealed. The samples were dried using a temperature-controlled oven until a constant weight was achieved. The solid samples were ground to fine powder and sieved using a fine aperture mesh (mesh size 2 μm). Each sample was then transferred to a cylindrical plastic-container. All samples were stored for at least four weeks before counting for achieving the secular equilibrium between gaseous and non-gaseous decay products of naturally occurring radionuclide series.

2.3 Gamma Spectrometry System

Gamma spectroscopic measurements were performed by means of a coaxial ORTEC HPGe detector with are lative efficiency of 28.2% and an energy resolution of 1.67 keV FWHM at the 1332.5keV peak of ^{60}Co.Data acquisition, display and analysis of gamma-ray spectra were performed using a commercial software Silena EMCAPLUS supplied by Silena, Germany. The efficiencies of the spectra peaks for the energies of the radionuclides were determined using the monoenergetic gamma sources ^{137}Cs, ^{60}Co and ^{40}K. The spectrum acquisition for the reference material and those of samples were carried out for 5000s.

The activity concentration of each sample was determined by subtracting the background from the spectrum of the sample. The activities of natural radionuclides were obtained from the peaks of their respective daughter products: ^{214}Pb (295 keV), ^{214}Pb (352 keV), ^{214}Bi (609 keV) and ^{214}Bi (1120 keV) for ^{238}U series; ^{212}Pb (239 keV), ^{208}Tl (583 keV), ^{228}Ac (911 keV) and ^{228}Ac (969keV) for ^{232}Th series; and the single peak of 1465 keV for ^{40}K. The activity concentration of individual radionuclides was calculated from the following analytical expression [15]:

$$A\left(\frac{Bq}{kg}\right) = \frac{N}{\varepsilon_\gamma \times \rho_\gamma \times T_s \times M}. \ldots\ldots (1)$$

Here $A$is the specific activity in Bq/kg of each radionuclide in the sample, $N$is the net number of counts in the resulting photo-peak, $\mathcal{E}_\gamma$ is the detector efficiency of the specific gamma-ray, ρ_γ is the intensity at the corresponding gamma-ray energy, T_s is the sample counting time in seconds and $M$is the mass of the sample in kg. Error associated with every calculation was measured by standard deviation equation.

3. RESULTS AND DISCUSSIONS

Table 1 presents the measured dry weight activity concentrations of the main gamma emitting radionuclides of the ^{238}U series, ^{232}Th series and ^{40}K in 12samplesof Barapukuria coal mine.The activities for each sample and radioisotope show a wide range of values. The activity concentration of ^{238}U ranged from BDL to 348.19 Bq/kg in all the measured samples with the mean concentration of 109.68 ± 119.97 Bq/kg. For ^{232}Th,the concentrations ranged from 7.97 to 770.81 Bq/kg with the mean concentration of 226.25 ± 293.16 Bq/kg. ^{40}K activity concentrations ranged from BDL to 936.65 Bq/kg with a mean concentration of 264.32 ± 340.93 Bq/kg. The mean specific radioactivity in the studied 6 different samples shown in Fig. 1 varies from 5.62 Bq/kg(for water) to 348.19 Bq/kg(for herb), 15.48 Bq/kg(for water) to 770.81 Bq/kg(for tailing) and 48.22 Bq/kg(for water) to 936.65 Bq/kg(for tailing) for^{238}U, ^{232}Th and ^{40}K, respectively.

Table 1: Radioactivity concentrations of ^{238}U series, ^{232}Th series and ^{40}K in Bq/kg in 12 studied samples. BDL stands for below detectable limit.

Sample ID	^{238}U series				^{232}Th series				^{40}K (1461keV)
	^{214}Pb (295keV)	^{214}Pb (351keV)	^{214}Bi (609keV)	^{214}Bi (1120keV)	^{212}Pb (238keV)	^{208}Tl (583keV	^{228}Ac (911keV)	^{228}Ac (969keV)	
C1	6.05	25.33	26.86	31.45	63.55	19.19	34.43	46.17	78.56
C2	BDL	8.27	6.04	BDL	28.54	8.10	15.14	67.89	56.77
C3	10.81	10.48	21.43	8.32	23.11	15.90	14.22	51.53	20.66
MW1	BDL	1.35	BDL	BDL	1.20	1.86	BDL	21.71	71.10
MW2	BDL	BDL	BDL	BDL	BDL	6.22	BDL	64.38	54.27
DW1	BDL	4.16	BDL	7.86	BDL	1.21	BDL	20.12	19.28
DW2	BDL	3.26	BDL	15.73	BDL	6.71	BDL	17.00	BDL
SW	BDL	BDL	BDL	BDL	BDL	2.50	BDL	13.45	BDL
S	61.75	40.78	36.01	27.02	93.53	24.95	80.16	18.25	287.75
T	348.58	553.23	253.63	237.32	908.41	253.31	811.10	1110.71	107.40
H	BDL	BDL	171.10	BDL	BDL	71.40	BDL	616.84	936.65
G	BDL	BDL	BDL	78.09	BDL	47.42	BDL	236.23	153.89
Average	109.68 ± 119.97				226.25 ± 293.16				264.32 ± 340.93

Table 2: Activity concentrations (Bq/kg) in soil, coal and tailing samples of different mining sites of the world.

Country	Coal			Soil			Tailing			Ref.
	^{238}U	^{232}Th	^{40}K	^{238}U	^{232}Th	^{40}K	^{238}U	^{232}Th	^{40}K	
Australia	27.75	40	81.5				51.50	48.1	114.7	[16]
Brazil	72	62	0				61.70	58.5	564	[17]
Egypt				17	18	320	78	33	337	[18]
Germany	32	21	225							[19]
Greece	253	25	0							[19]
Hungary	250	55	207							[19]
Poland	18	11	785							[19]
Romania	80	62	0							[19]
UK	13	13	184.5							[19]
USA	40	13	0	40	35	449				[20]
China				32	41	440	56.5	36.5	173.2	[21]
Japan				33	28	310	35.8	20.7	139.4	[22]
India				29	64	400	37	24.1	432.2	[23]
Iran				28	22	640				[19]
Denmark				17	19	460				[19]
Poland				26	21	410				[19]
Greece				25	21	360				[19]
Romania				32	38	490				[19]
Nigeria				14	19	896				[24]
Spain				32	33	470				[20]
World average	35	30	400	35	30	400	35	30	400	[20]
Present study	14	31	52	41	54	288	348	771	107	

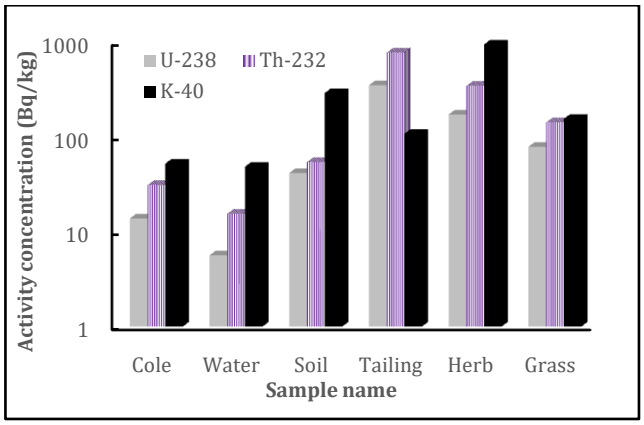

Fig -1: Mean specific activities of ^{238}U, ^{232}Th and ^{40}K measured in 6 different types of studied samples.

The present results for coal, soil and tailing are compared with the world average as well as the reported data available in literature and are depicted in Table 2. The comparison shows that the mean specific activities of natural radionuclides in the studied samples produce a reasonable agreement with those from other countries. However, the ^{238}U concentrations in coal samples are much higher in Greece and Hungary than the present values. For ^{232}Th concentrations, the lower values than the present results are found in Poland for coal and in Egypt, Denmark and Romania for soil samples. In case of ^{40}K concentrations for coal samples the present result is much lower than those reported by other countries. This variation in activity concentrations may be due to their radioactive mineral content and the geographical origins of the raw materials.

In all kinds of the studied samples the present results of ^{40}K concentrations are less than the world average. However, the concentrations of both ^{238}U and ^{232}Th in tailing samples are found to be much higher in the present study than the world average. The possible reason for these higher values is that the tailings consist of a slurry of fine particles, ranging from the size of a grain of sand to a few micrometres that overlies an ore or mineral body and is displaced during mining without being processed. It is worth mentioning also that the river Padma flows, as the Ganges, through Bihar in India, where there is a uranium mine. The rivers Brahmaputra, Surma and Kushiyara flow through Assam in India, where there is a uranium deposit. Thus, it is likely that the traces of uranium and its decay products are carried with the water flow through these rivers, causing the uranium level to be somewhat higher in Bangladesh.

In order to evaluate the radiological hazards of the natural radioactivity several hazardindices are important. These indices includethe radium equivalent activity, the absorbed gamma dose rate in the indoor environment and the corresponding annual effective dose, the external and internal hazard indices,the alpha index (internal index), the gamma activity concentration (gamma index),annual gonadal dose equivalent, excess lifetime cancer risk, etc. Among the aforesaid indices we have estimated the radium equivalent activity $Ra_{eq,}$ absorbed gamma dose rate D, annual effective dose D_{eff} and external hazard index H_{ex}using the formulae given in ref. [25].Our estimated results are depicted in Table 3.

Table 3: Estimated various hazard indices associated with the radioactivity of the studied samples.

Sample ID	Ra_{eq} (Bq/kg)	D (nGy/h)	D_{eff} (μSv/y)	H_{ex}
C1	92.00	40.67	49.82	0.24
C2	48.78	21.41	26.22	0.13
C3	51.81	21.80	26.7	0.13
MW1	23.21	10.50	12.87	0.06
MW2	54.65	23.58	28.89	0.14
DW1	231.82	106.62	130.60	0.62
DW2	26.67	11.64	14.26	0.07
SW	11.40	4.81	5.90	0.03
S	194.96	88.76	108.73	0.52
T	1646.13	717.49	878.94	4.44
H	735.30	325.94	399.28	1.98
G	292.75	128.16	156.99	0.79
Safety limit	370	57	70	1

The radium equivalent activity takes into account the radiation hazards associated with each radionuclide. It is a widely used radiation hazard index that compares the specific activity of materials containing varying concentrations of ^{238}U, ^{232}Th and ^{40}K. As seen in Table 3, the Ra_{eq} values in the present work are within the accepted safety limit of 370 Bq/kg as recommended by the Organization for Economic Cooperation and Development (OECD). However, in tailing and herb samples the Ra_{eq} values are, respectively, 4.5 and 2 times greater than those of the safety limits.

The contribution of terrestrial gamma radiation to the absorbed dose *D* in air can be estimated by the determination of the exposure dose rate in air at 1 m above the ground surface. However, the absorbed dose rate does not directly provide the radiological risk to which an individual is exposed. The absorbed dose is, therefore, considered in terms of the annual effective

dose equivalent from outdoor terrestrial gamma radiation. Another important radiation hazard parameter is the external hazard index that reflects the external exposure to radiation dose. From Table 3 one can see that the present results of the absorbed dose, annual effective dose and external hazard index are below the safety limits for most of the samples except for tailing and herb.

4. CONCLUSIONS

The activity concentrations of natural radionuclides ^{238}U, ^{232}Th *and* ^{40}K have been measured in 6 different types of samples (coal, soil, water, tailing, herb and grass) collected from the Barapukuria coal mine of Bangladesh. The results show that the mean activity concentrations of the radionuclides in almost all samples (except tailing and herb) are smaller than the world average values. Radiological health hazard indices have also been estimated from the measured activity concentrations to assess the potential radiation hazards associated with these samples. Except for tailing and herb samples, the values of radium equivalent activity, external hazard index, absorbed gamma dose rate and annual effective dose are all found to be within their recommended safety limit and, therefore, do not pose any significant radiation hazards. But, the present study recommend to take proper management of tailings and other norm residues.

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