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Radioactivity of soil at proposed Rooppur Nuclear Power Plant site in Bangladesh

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► ***Original article***

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ABSTRACT

Background: The concentrations and distributions of natural and anthropogenic radioactive materials in soils of the site of Rooppur Nuclear Power Plant (RNPP) were investigated with the aim of evaluating the environmental radioactivity and radiation hazard. ***Materials and Methods***: Soil samples were collected from 40 locations in and around the proposed site of Rooppur Power Plant and determined the activity level by using a gamma-ray spectrometry. ***Results***: The concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in soil samples, radium equivalent activity (Ra_{eq}) for soil, absorbed dose rates, external hazard (H_{ex}) values were determined. No artificial radioactivity (^{137}Cs) was found in these samples. ***Conclusion***: This present study provides a baseline of radioactivity and radiation levels in the proposed nuclear power plant site area at Rooppur. It will be utilized to correlate the radioactivity measured after operation of the reactor.

Keywords: *Gamma spectrometry, radioactivity, soil, Rooppur nuclear power pant (RNPP).*

INTRODUCTION

In modern times, economic growth of a country is measured in terms of its capacity for production of electricity. All countries seek to ensure a supply of electricity that is affordable, reliable, and secure for sustaining modern lives. In developing countries like Bangladesh the demand for electricity is growing and the actual production of electricity is expanding day-by-day. In order to improve the electric power generation capacity and sustain their economic growth, long-term plan is needed. From this view, the government of Bangladesh has attached firm commitment to implement the Rooppur NPP as the first Nuclear Power Plant (NPP) in Bangladesh. Bangladesh has domestically and internationally committed to the peaceful applications of ionizing radiation in order to achieve socio-economic development in the country. Environmental radiation and radioactivity base line study has become important nationally in every country. Such investigations can be useful for both the assessment of public dose rates and the performance of epidemiological studies, as well as to keep reference-data records, to ascertain possible changes in the environmental radioactivity due to nuclear, industrial and other human activities.

Radiation is a natural part of the environment in which we live [1-4]. All people receive exposure from natural origin (e.g., ^{238}U, ^{232}Th, ^{226}Ra, ^{222}Rn, ^{210}Pb, ^{40}K etc.), as well as anthropogenic ones (^{137}Cs, ^{134}Cs, ^{131}I, ^{90}Sr, ^{85}Kr, ^{239}Pu etc.) radionuclides in soil, water, air and food. The natural occurrence of ^{238}U and ^{232}Th in the earth's crust is a source of external exposure due to gamma-rays. The ^{238}U decay series consists of 15 members in total including ^{226}Ra and the series ends with stable ^{206}Pb which is a stable element. After ^{226}Ra, the series emits eight alpha and six beta particles along with many gamma-rays. There are 12 members in the decay series of ^{232}Th and it decays into ^{208}Pb after emitting seven alpha and five beta particles

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along with many gamma-rays. In addition, among the naturally occurring potassium isotopes, ^{40}K is unstable, and decays by the emission of 1,460 keV gamma-rays and is also a source of gamma-rays. ^{137}Cs in the environment comes from a variety of sources. The largest single source is a from atmospheric nuclear weapons tests in the 1950s and 1960s, which dispersed and deposited ^{137}Cs world-wide. However, much of the ^{137}Cs from the testing has now been decayed. Nuclear reactor accident, such the Chernobyl accident (1986) in the Ukraine, releases ^{137}Cs to the environment [5,6]. Radioactivity in soil and water system may ultimately find its way to human through food chain and by direct content with the ecosystem. So, the knowledge of the distribution pathways of both artificial and natural radionuclides is essential in maintaining some surveys of control of prevailing radiation and radioactive contamination levels [7].

Bangladesh Atomic Energy Commission (BAEC) has taken a comprehensive programme on measurement of radiation level at the site of the RNPP. There are no other nuclear/radioactive installations around the NPP site. The installation of radioactivity monitoring data will be utilized to compare pre and post installation data and to take safety measures if there is any abnormal rise of radiation level due to operation power reactors. Thus this will ensure safety of the population and the environment. The aim of this study is to determine natural (^{226}Ra, ^{232}Th, ^{40}K) and artificial (^{137}Cs) radioactivity levels in soils from various areas of RNPP site, and also to investigate the radium equivalent activity (Ra_{eq}), the absorbed dose rate, and external and internal radiation hazard indices. The result of the monitoring study will be required as regulatory pre-requisites for the installation /operation of RNPP in future.

MATERIALA AND METHODS

Sample collection

The NPP site is located on the east bank of the Ganges River at the village of Rooppur, the district of Pabna and approximately 170 km northwest of the city Dhaka, the capital of Bangladesh. The coordinates of the sites are latitude 24º4.35" North and longitude 89º2.80" East. Soil samples were collected from 40 locations around the proposed site of RNPP. The locations are shown in the figure 1. All the samples were collected at 0-5 cm depth from the soil-surface. Each of the samples weighed 1kg. The samples transferred in dried acetone-cleaned polyethylene bags with sample codes were transported to the laboratory.

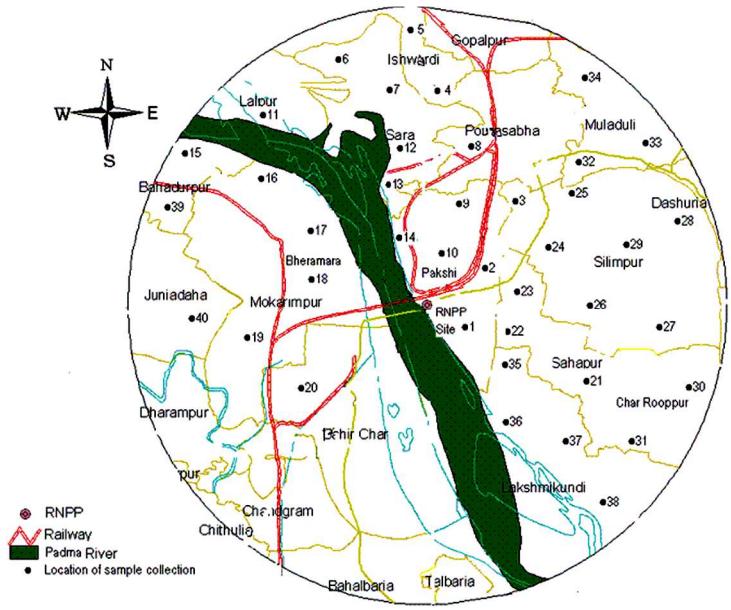

Figure 1. Location Map of Rooppur (Bangladesh) Nuclear Power Plant site.

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Sample preparation

At the lab, the collected samples were transferred from the polyethylene bags to the acetone-cleaned stainless steel bucket and dried in an oven at 100-105 ^{0}C until a constant weight was achieved. Each of the dried samples was ground to fine powder in an agate motor separately. The powdered soil samples were then sieved using a fine aperture mesh screen (mesh size 2 mm) in order to remove extraneous items like plant materials, roots, pebbles etc. and to obtain a fine-grained sample that would present a uniform matrix to the detector. The ground samples were stored in separate polyethylene packets as stock samples. Finally, each of the samples was transferred to cylindrical plastic-container. The containers were approximately of equal size and shape (i.e., diameter 6.5 cm and height 7.5 cm). The net weight of each sample was determined using a microbalance. The weight is different for each sample because of the different density of the sample material. The containers were then sealed tightly, rapped with thick vinyl tapes around their screw necks. The samples were stored for at least four weeks to reach secular equilibrium between the ^{238}U and ^{232}Th series and their respective progenies to get ready for measurements [8].

Measurement procedures

Since 98.5% of the radiological hazard of the Uranium series are caused by the Radium, and its daughter products and the contribution from the ^{238}U and the other daughter products of ^{226}Ra can generally be ignored [9]. The radioactivities of soil samples were measured using a low-level gamma counting system, a high resolution HPGe coaxial detector (EG & G ORTEC) coupled with a silena Emcaplus multichannel analyzer (MCA) and associated microprocessors. The effective volume of the detector was 83.469 cm^3 and energy resolution of the 1.33 MeV energy peak for ^{60} Co was found as 1.69 keV at full width half maximum (FWHM) with a relative efficiency of 19.6%. The detector was shielded by using requisite thickness of mild steel ring at the side and lead at the top for optimization of the background radiation level. The radioactivity concentration of ^{226}Ra was determined from γ-ray energies of its daughters ^{214}Pb (351.92 & 295.21 keV) and ^{214}Bi (609.31 & 1120.30 keV), while the ^{232}Th was determined from γ-ray energies of its daughters ^{212}Pb (238.63 keV), ^{208}Tl (583.14 keV) and ^{228} Ac (911.07 & 969.11 keV). The radioactivity concentrations of ^{40}K and ^{137}Cs were determined from their γ-ray energy of 1460.80 keV and 662 keV respectively. The efficiency calibration of the detector was performed by using standard sources Eu-152 mixed with 250 g of analytical grade Al_2O_3 at different energy such as 121.78 keV, 244.69keV, 344.27 keV, 411.11 keV, 443.91 keV, 778.89 keV, 963.38 keV, 1085.78 keV, 1112.01 keV & 1407.67 keV. The efficiency calibration curve thus obtained was employed in the different energy peaks covering the range up to 2000 keV to obtain the efficiency of the detector for the particular gamma ray energy of interest. The geometry of the counting samples was the same as that of the standard samples and the counting time for all the sample was 5000sec. In order to determine the background distribution in the environment around the detector, an empty sealed beaker was counted in the same manner and in the same geometry as the samples. The background spectra were used to correct the net peak area of gamma rays of measured isotopes.

Theoretical calculations

The activity concentrations: The activity concentrations of the radionuclides in the measured samples were computed using the following relation [10],

$$A_s\left(BqKg^{-1}\right)=\frac{C_a}{\varepsilon P_\gamma M_s} \qquad (1)$$

where C_a is the net gamma counting rate (counts per second), ε the detector efficiency of the specific γ-ray, P_γ the transition probability of gamma decay and M_s is the mass of the sample (kg).

Radium equivalent activity

For the purpose of comparing the radiological effect or activity of materials that contain ^{226}Ra, ^{232}Th and ^{40}K by a single quantity,

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which takes into account the radiation hazards due to these isotopes, a common index termed as the radium equivalent activity (Ra_{eq}) is used. This activity index provides a useful guideline in regulating the safety standards on radiation protection for the general public residing in the area under investigation. The Ra_{eq} index represents a weighted sum of activities of the above mentioned natural radionuclides .The index is given as [3]:

$$Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_K \quad (2)$$

where A_{Ra}, A_{Th} and A_k are the average activity concentration in the sample in Bqkg $^{-1}$ of ^{226}Ra, ^{232}Th and ^{40}K respectively.

The absorbed dose rate

The absorbed dose rate in air at average gonad height of one meter above the surface of ground due to the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K was estimated using formula given in UNSCEAR, 1988[3]:

$$D=[0.427A_{Ra} + 0.662\,A_{Th} + 0.0432\,A_k]\ nGyh^{-1} \quad (3)$$

where A_{Ra}, A_{Th} and A_k are the same meaning as in equation 2.

External and Internal Hazard indices

The soils are used for manufacturing earthen huts, bricks and pottery materials and, hence, the external radiation hazard index, H_{ex} and internal radiation hazard index, H_{in} were calculated using the following relations [8].

$$H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \quad (4)$$

$$H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \leq 1 \quad (5)$$

where A_{Ra}, A_{Th} and A_k are the same meaning as in equation 2.

RESULTS AND DISCUSSION

Activity concentrations of Natural Radionuclides

The calculated activity concentrations of radionuclides were obtained from gamma spectrometry measurements for 40 soil samples collected from different locations of the Rooppur NPP site, Bangladesh in table 1. Also shown are the derived radium equivalent activity, the absorbed dose rate, and external and internal radiation hazard indices due to activity concentration of the natural radionuclides measured. Three major natural gamma radiation sources were identified. Radium-226 was the precursor of most of the γ-emitting radionuclide identified in the Uranium decay series. Thorium-232 was likely the precursor of the isotopes identified in the Thorium decay series. Potassium-40 which accounts for most of the activity is the third source [9]. It should be noted that the values given in the tables 2 and 3 are not the representative values for each country but for the locations from where the samples have been collected. For ^{226}Ra activity concentration, each value was obtained from the 351.92 keV photo-peaks of ^{214}Pb and the 609.31 keV photo-peaks of ^{214}Bi. The results for soil ranged from 25.52±1.37 to 37.54± 2.28 Bqkg^{-1} with an average of 30.85 ± 3.42 Bqkg^{-1}. For the ^{232}Th activity concentration, each value was an average of three values obtained from 238.63keV photo-peaks of ^{212}Pb, 911.07 and 969.11 keV photo peaks of ^{228}Ac. The results for soil ranged from 32.89±1.66 to 53.37± 2.00 Bqkg^{-1} with an average of 40.88 ±5.17 Bqkg^{-1}. The activity concentration of ^{40}K was obtained from its photo-peak of 1460.80 keV. The results for soil ranged from 313.19±27.76 to 521.65± 28.35 Bqkg^{-1} with the average value of 390.10± 53.84 Bqkg^{-1}. The errors quoted represent the counting error for individual measurements and standard deviation for means. In the soil, in general, the activity concentration of ^{232}Th was found to be higher than that of ^{226}Ra, while that of ^{40}K markedly exceeds the values of both ^{226}Ra and ^{232}Th. Table 1 shows that the activity concentration of thorium is higher than radium, which is evident from the fact that the activity of thorium is 1.5 times higher than that of Uranium in earth's crust [11]. It is also observed that the measured activity concentration of ^{40}K exceeds markedly the values of both Radium and Thorium, as it is the most abundant radioactive element under consideration. Moreover the excessive use of the Potassium containing

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fertilizers in the area adjacent to the sampling sites may contribute to the higher values of ^{40}K activity. In the present study, the activity concentrations of ^{226}Ra and ^{232}Th in soil are comparable with the reported values in table 3, especially those at the different regions of Bangladesh [12-15]. The activity concentrations of ^{40}K are comparable with the other values reported in table 2. Moreover, our obtained values fall within the range of corresponding world values and other published results mentioned in table 2. The world average activity concentration [2,3,8] of ^{226}Ra is 40 Bqkg $^{-1}$ with ranges of 15 – 50 Bqkg $^{-1}$, ^{232}Th is 40 Bqkg $^{-1}$ with ranges of 7 – 50 Bqkg $^{-1}$ and ^{40}K is 580 Bqkg $^{-1}$ with ranges of 100 – 700 Bqkg $^{-1}$.

Table1. Radioactivity concentrations of radionuclides as well as the calculated Ra equivalent activity, absorbed dose rate, external hazard index and internal hazard index in soil samples across the site of RNPP.

No of Soil sample	Activity of Radionuclide (Bqkg^{-1})				Ra_{eq} (Bqkg $^{-1}$)	Dose Rate D (nGyh^{-1})	H_{ex}	H_{in}
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs				
RNPP-01	27.92±1.01	36.79±2.15	374.90±27.95	-	109.32	52.47	0.295	0.371
RNPP-02	30.14±2.24	36.56±2.17	330.59±32.93	-	107.80	51.35	0.291	0.373
RNPP-03	35.83±1.60	43.16±1.38	335.54±48.33	-	123.30	58.37	0.333	0.430
RNPP-04	31.71±2.10	39.73±2.15	396.93±30.88	-	119.00	56.99	0.322	0.407
RNPP-05	32.82±1.49	49.83±1.01	415.30±40.58	-	135.95	64.94	0.367	0.456
RNPP-06	29.05±1.31	38.86±1.46	374.39±30.08	-	113.36	54.30	0.311	0.38
RNPP-07	36.86±2.16	42.75±2.22	432.25±30.27	-	131.18	62.71	0.355	0.454
RNPP-08	28.05±1.59	35.98±1.29	376.12±33.35	-	108.38	52.04	0.293	0.369
RNPP-09	26.22±1.54	32.89±1.66	335.54±38.33	-	99.02	47.46	0.268	0.338
RNPP-10	34.41±1.45	46.13±2.29	394.45±32.36	-	130.65	62.27	0.353	0.446
RNPP-11	33.55±1.66	42.76±1.19	370.48±36.40	-	123.13	58.64	0.333	0.423
RNPP-12	36.91±1.77	44.32±1.45	354.14±38.57	-	127.466	60.40	0.345	0.444
RNPP-13	27.86±2.11	36.43±1.88	322.91±31.19	-	104.74	49.96	0.283	0.358
RNPP-14	29.87±2.19	43.14±1.95	401.07±29.68	-	122.35	58.64	0.331	0.411
RNPP-15	30.76±2.82	40.95±2.04	335.05±28.67	-	113.59	54.05	0.307	0.390
RNPP-16	30.13±2.62	39.94±1.31	464.88±38.14	-	122.95	59.39	0.332	0.414
RNPP-17	32.11±1.72	44.04±1.99	409.92±39.28	-	126.57	60.57	0.342	0.429
RNPP-18	25.52±1.37	36.34±1.84	389.29±29.55	-	107.38	51.77	0.291	0.359
RNPP-19	39.44±2.01	50.80±2.28	521.65±28.35	-	152.14	73.01	0.411	0.518
RNPP-20	29.32±1.69	40.89±1.44	399.10±32.55	-	118.43	56.83	0.320	0.399
RNPP-21	31.00±1.59	49.37±1.34	381.83±30.17	-	130.90	62.41	0.354	0.438
RNPP-22	30.74±2.64	40.31±2.10	361.28±30.39	-	116.15	55.42	0.314	0.397
RNPP-23	29.74±1.75	41.80±2.49	338.40±26.25	-	115.49	54.99	0.312	0.393
RNPP-24	32.29±1.41	49.35±1.27	396.70±35.36	-	133.31	63.60	0.360	0.448
RNPP-25	30.47±2.02	43.85±1.39	402.18±32.13	-	124.05	59.41	0.335	0.418
RNPP-26	28.87±1.76	34.56±1.93	313.19±27.76	-	102.33	48.74	0.277	0.355
RNPP-27	27.06±1.39	38.64±2.18	352.53±32.73	-	109.38	52.36	0.296	0.369
RNPP-28	29.24±1.94	36.33±1.56	415.30±30.58	-	113.09	54.48	0.306	0.385
RNPP-29	31.52±1.61	49.97±1.79	383.82±27.41	-	132.43	63.12	0.358	0.443
RNPP-30	26.99±2.01	33.45±2.07	396.83±29.26	-	105.30	50.81	0.286	0.358
RNPP-31	28.37±1.88	40.50±1.21	332.76±27.99	-	111.82	53.30	0.302	0.379
RNPP-32	31.26±1.51	42.74±3.55	436.49±30.10	-	125.89	60.50	0.340	0.425
RNPP-33	27.04±1.74	50.18±2.21	518.30±28.47	-	138.59	67.16	0.375	0.448
RNPP-34	33.76±2.10	46.62±1.95	438.30±21.29	-	134.08	64.21	0.362	0.454
RNPP-35	37.54±2.28	53.37±2.00	504.90±28.31	-	152.62	73.17	0.412	0.514
RNPP-36	30.26±1.79	45.17±2.66	494.85±28.19	-	132.85	64.20	0.359	0.441
RNPP-37	26.54±1.99	40.07±3.52	361.34±27.79	-	111.58	53.47	0.302	0.373
RNPP-38	33.92±1.75	38.12±2.05	350.66±30.26	-	115.35	54.86	0.311	0.403
RNPP-39	25.63±1.22	39.82±2.02	368.04±27.87	-	110.82	53.20	0.299	0.368
RNPP-40	33.25±1.67	38.79±2.48	321.65±28.51	-	113.40	53.77	0.306	0.396
Average	30.85	40.88	390.10		120.65	57.73	0.326	0.409
STD	3.42	5.17	53.84		12.66	6.10	0.034	0.042

(± values represent counting error for individual measurement and standard deviation for average. The maximum and minimum values are given in bold face).

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The fission product (^{137}Cs)

The manmade fission product residue ^{137}Cs, which would come down to this part of the earth from the atmosphere following the nuclear power plant accident at Chernobyl on 26 April 1986 and other previous test of nuclear devices around the world, was considered in this study to obtain an estimate of fallout in the Rooppur NPP site. But no detectable activity of ^{137}Cs was observed in the soil samples under study. It implies, therefore, that there is no activity due to fallout in the site of the Rooppur NPP, Bangladesh.

Radium equivalent activity, Ra_{eq}

Calculated data of Ra_{eq} is presented in column 6 of table 1. The Ra_{eq} for soil varied in the range 99.02 to 152.62 $Bqkg^{-1}$ with the average value of 120.65±12.66 $Bqkg^{-1}$. Those are far below the allowable limit (370 $Bqkg^{-1}$) as recommended by the IAEA[2,3,8].

The absorbed dose rate

In the present study the dose rate due to ^{226}Ra, ^{232}Th, and ^{40}K in soil samples varied from from 47.46 to 73.17 $nGyh^{-1}$ with an average of 57.73±6.10 $nGyh^{-1}$. These values are comparable with the world average [3] value of 55 $nGyh^{-1}$.

External and internal hazard index

The external hazard index (H_{ex}) due to natural gamma radiation was calculated using the equation (3) which is shown in column 8 of table 1. The H_{ex} value for soil in the area of interest ranged from 0.268 to 0.412 with the average value of 0.326±0.034. The calculated H_{ex} values for all samples should be lower than unity, which do not cause any harm to the population in all regions under investigation. All values of H_{ex} in the present work are less than unity. There is also a radiation hazard threat to respiratory organs due to the ^{222}Rn, decay product of ^{226}Ra, and its short-lived decay products. To account for this the maximum permissible concentration for radium must be reduced to half of the normal limit [8]. Considering this limit, the internal hazard index, H_{in} was calculated and is shown in the last column of table 1. The H_{in} for soil is found to range from 0.338 to 0.514 with the average value of 0.409±0.042.

Table 2 and table 3 show the comparative study of the present work with others of the world. However, slight variation in the radioactivity content in soil was observed with various locations worldwide mainly due to soil type, formation and transport process involved.

Table 2. Comparison of present study with other work.

Location	Activity in $Bqkg^{-1}$			
	^{226}Ra	^{232}Th	^{40}K	^{137}Cs
Dhaka, Bangladesh [23]	37.8	58.2	790.8	-
Chittagong, Bangladesh [12]	34.6	60	438	1.08
Jessore, Bangladesh [22]	48	53	481	-
Nine southern districts, Bangladesh [14]	42	81	833	2.08
Eastern sichuan province, China [20]	26	49	440	6
Peshwar, Pakistan [18]	65	84	646	-
Nigeria [17]	8.3	34.3	684	-
Nile Delta, Egypt [19]	17	-	316	-
Louisiana, USA [9]	43 - 95	50 - 190	43 -729	5-58
Worldwide average [1]	40(15-50)	40(7-50)	580(100-700)	-
RNPP site (Bangladesh) (Present study)	30.85	40.88	390.10	-

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Table 3. Comparative study of the present works with others of the world.

Country	Outdoor average absorbed dose rate in nGy h^{-1}	Range	Outdoor effective dose equivalent at 1.0m gonad height in μSvy^{-1}	Method of analysis
Romania [3]	81	32-210	99.34	Analysis of soil using gamma Spectrometry
Tripoli[16]	23	-	28.21	Analysis of soil using gamma Spectrometry
Nigeria [17]	128	5-186	156.90	Analysis of soil using gamma Spectrometry
Pakistan[18]	49	39-63	60	Analysis of soil using gamma Spectrometry
Egypt[19]	93	-	114.6	Analysis of soil using gamma Spectrometry
China[20]	60	50-72	-	Analysis of soil using gamma Spectrometry
Six districts, Bangladesh [21]	77	59-103	94.93	Analysis of soil using gamma Spectrometry
Nine districts, Bangladesh [14]	107	60-156	-	Analysis of soil using gamma Spectrometry
Worldwide average [4]	55	30-70		
RNPP site, Bangladesh (present study)	40	57.73	47.46 – 73.17	Analysis of soil using gamma Spectrometry

CONCLUSION

The radioactivity and radiation levels in soil samples in and around the proposed Rooppur NPP Site must be useful as baseline data for the proposed NPP site at Rooppur for the purpose of detecting any changes in radioactivity in future. Radiological impact on the environment after commissioning of the nuclear power plant and any other nuclear fallout could be assessed by using this baseline data. The present study also helps to assess the radium equivalent activity, the absorbed dose rate, and external and internal radiation hazard due to presence of radionuclides in the soil. The report was submitted to the RNPP Implementation Committee.

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Conflicts of interest: *none to declare.*

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