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ACTIVITY CONCENTRATION OF ^{137}Cs AND NATURALLY OCCURRING RADIONUCLIDES IN SOIL AND WATER SITE OF GOAINGHAT AND JAINTAPUR AREA OF SYLHET, BANGLADESH

M. Shamsuzzaman[1], M. Nurul Alam[2], M. M. Rahman[2] and Molla M. S. Rahman[3*]

[1]*Reactor Operation and Maintenance Unit (ROMU), Atomic Energy Research Establishment, G.P.O. Box-3787, Dhaka-1000, Bangladesh*

[2]*Radiation Monitoring and Radioactive Waste Management Unit, Savar , Bangladesh*

[3]*Environmenta Science Discipline, Khulna University, Khulna-9208*

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Abstract: The concentration of anthropogenic ^{137}Cs and naturally occurring ^{226}Ra, ^{232}Th and ^{40}K radionuclides were measured in the undisturbed soil at 0-5 cm depth and water collected from the natural reservoir in the Terrene of *Goainghat* and *Jaintapur* area of Sylhet district of Bangladesh near the Indian border area by gamma ray spectrometry, with the aim of evaluating the environmental radiation hazard. The activity concentration of ^{137}Cs was observed in all the soil and water samples, ranging from 5.22 4 - 31.74 and 1.1 - 2.2 Bqkg^{-1}respectively. The activity level of ^{137}Cs in this area was lower than that of Cox′ s Bazar (27.38-45.51 Bq kg^{-1}) and higher than that of Chittagong (2.66 Bq kg^{-1}). Values of the natural radionuclides present in the samples were greatly influenced by the geomorphological conditions in the area. The average activity of ^{226}Ra, ^{232}Th and ^{40}K in soil were 57.5, 167.6 and 1202.3 Bq kg^{-1} respectively. The average activities of ^{232}Th and ^{40}K in the soil samples of this area were higher than the world average value of 25.0 and 350.0 Bq kg^{-1} respectively. The average γ-ray dose rate due to naturally occurring radionuclides in soil samples was found 180 nGy h^{-1}.

Key words: Activity concentration, radionuclide, environmental radiation hazard, Bangladesh

Introduction

The soil and plants of all over the world were contaminated with radioactivity from the Chernobyl nuclear power plant accident in Ukraine on the 26th April, 1986. The radioactivity then passed on to milk, fruits and vegetables (IAEA, 1989). Such an accident could be catastrophic in a densely populated country like Bangladesh. The previous environmental radioactivity studies revealed that no part of Bangladesh was contaminated by the Chernobyl accident or by any other anthropogenic nuclear activities. As a part of routine environmental radioactivity study samples were collected from the Goainghat and Jaintapur area of Sylhet district of Bangladesh close to the border of Indian Province Meghalaya and Assam (IAEA, 1991). The soil samples of this location were found contaminated by artificial ^{137}Cs radionuclides.

Radioactivity in soil and water system may ultimately finds its way to humans through food chain and by direct or indirect contact with the ecosystem, so the knowledge of the distribution pattern of both anthropogenic and natural radionuclides is essential in maintaining some sense of control

* **Corresponding author**: <mmsr68@yahoo.com>

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of prevailing radiation levels (IAEA, 1989). Soil and water is contaminated fast after the nuclear explosion or other events such as atmospheric pollutions and radioactive fallout. Measurement of natural and fallout radioactivities in soil and water give information on natural sources, cumulative deposition of fission products from nuclear device testing and nuclear accidents.

All rocks, soils and minerals contain naturally occurring radionuclides such as ^{226}Ra, ^{232}Th, ^{40}K and their progency (Ibrahiem *et al*., 1995; Schotzig U. *et al*., 1983; NEA-OECD, 1979). Radium radionuclides are of radiological importance to human; water and aquatic lives and plants contain radium radionuclides taken up from soil. Qualitative and quantitative knowledge of the radioactivity in soil and water ecosystems is important since it concerns with radionuclides liable to cause radiation protection problems under extreme conditions. The fission product ^{137}Cs was measured to assess their contents and the distribution pattern in soil and water ecosystem (Cox and Fankhauser, 1984).

The main objective of the present work is to provide information on natural and artificial radioactive isotopes and environmental pollution by radionuclides which may be useful in the assessment of human radiation exposure from natural environment and monitoring program on environmental radioactivity and radiation aimed at minimizing radiation exposure to population.

Materials and Method

Sample collection: The study area *Goainghat* and *Jaintapur* of Sylhet District are located at 91°50'-92°13' N and 24°56'-25°12' E .The soil samples were collected from 20 sites at a depth of 0-5cm with the help of pre-washed steel corer of 12 cm in diameter and 25cm height. Water samples were also collected from 20 sites of natural water reservoir corresponding to the location of soil sample sites at a distance of 500 m to 4 km from each other during January to October 2000.

***Sample preparation*:** The soil samples were oven dried at 110° C for 48 h, pulverized and passed through sieve (size 90 μm), weighed and then packed in cylindrical plastic containers (6.5 cm × 7.5 cm). They were then sealed tightly with caps, wrapped with thick vinyl tape around their screw necks and then stored for 4 weeks to allow secular equilibrium between ^{226}Ra, ^{232}Th and their daughter products. Water samples were collected with 5-liter plastic jars from the natural water reservoir. Each 5-litre water sample was boiled to reduce its volume to 500 ml and packed into 500 ml cylindrical containers, sealed tightly and wrapped with thick vinyl tapes around their screw necks. These samples were stored for 4 weeks (Ibrahiem *et. al*., 1995; Cox and Fankhauser, 1984; Schotzig, 1983).

***Measurement procedure*:** The γ-ray activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were analyzed using a p-type coaxial lead shielded high purity intrinsic germanium detector having relative efficiency of 30%, active volume 132 cm^3, resolution at FWHM of 1.85 keV for 1332 keV γ-ray of ^{60}Co coupled with PCA and other accessories. The calibration of peak efficiency of the detector was performed using IAEA reference samples ^{238}U (RGU-1), ^{232}Th (RGTh-1), ^{40}K (RGK-1) and ^{137}Cs (IAEA-152) (AQCS, IAEA, 1995). Since the chemical and packing densities of soil, water and standards are completely different so that the geometry effects of samples and standards (i.e., chemical composition, density, gamma ray self attenuation etc.) were considered during efficiency calibration for all the gamma rays. For the measurement of ^{226}Ra activity concentration, the γ-ray energies of 295.21 and 351.92 keV of ^{214}Pb, 609.31 and 1120.29 keV of ^{214}Bi were used. The activity concentration of ^{232}Th was determined at the γ-ray energies 338.4 and 969.11 keV of ^{228}Ac, 583.19 and 2614.53 keV of ^{208}Tl. The different gamma rays, which were emitted from the daughters of ^{226}Ra and ^{232}Th, used to quantify the parents, produced consistent results. The final activity concentration of ^{226}Ra and ^{232}Th were calculated taking the average of individual daughter products. The ^{40}K and ^{137}Cs radionuclides were measured from their respective γ-ray energies 1460 and 661.66 keV, respectively (IAEA, 1989; ICRP, 1983).The lower limits of detection of the

detector for 150 ml sample geometry are 0.07, 0.18, 0.03, 4.20 Bq kg^{-1} for ^{232}Th, ^{226}Ra, ^{137}Cs, ^{40}K respectively.

Results

Activity Concentration in soil and water of ^{137}Cs : The measured activity concentration of ^{137}Cs in soil and water of *Goainghat* and *Jaintapur* of Sylhet, Bangladesh is shown in Table 1.

Table 1: Activity concentration of ^{137}Cs in soil and water of Goainghat and Jaintapur of Sylhet, Bangladesh.

Sample No.	Location	No. of sample	Activity of ^{137}Cs in soil (Bqkg^{1}) (October,2000)	Activity of ^{137}Cs in water (Bq L^{-1}) (October,2000)	Calculated activity of ^{137}Cs in soil (Bq kg^{-1})(1986)*
1	Dibirhaor	3	12.65 ± 0.93	2.21 ± 0.62	16.80
2	Dibirhaor	3	14.73 ± 0.86	2.11 ± 0.62	19.05
3	Dibirhaor	3	31.74 ± 0.89	1.72 ± 0.61	10.86
4	Gilatoil	3	8.76 ± 0.45	1.25 ± 0.44	28.09
5	Gilatoil	3	20.98 ± 0.68	1.34 ± 0.47	19.76
6	Goirisankar	3	15.56 ± 0.79	1.20 ± 0.42	20.75
7	Fhulbari	3	15.87 ± 0.75	1.15 ± 0.40	35.57
8	Fhulbari	3	26.53 ± 0.88	1.24 ± 0.36	22.44
9	Fatehpur	3	16.97 ± 0.79	1.35 ± 0.48	11.71
10	Fatehpur	3	9.58 ± 0.68	1.10 ± 0.39	9.74
11	Fatehpur	3	7.95 ± 0.78	1.20 ± 0.43	5.79
12	Fatehpur	3	5.22 ± 0.45	1.0 0 ± 0.35	11.15
13	Fatehpur	3	8.99 ± 0.71	1.25 ± 0.36	13.83
14	Goirisankar	3	10.91 ± 0.79	1.22 ± 0.43	13.69
15	Shreepur	3	10.98 ± 0.77	1.0 0 ± 0.34	16.80
16	Tamabil	3	12.89 ± 0.83	1.20 ± 0.42	17.22
17	Pas. Jaflong	3	13.35 ± 0.86	1.20 ± 0.44	15.63
18	Pas. aflong	3	14.41 ± 0.78	1.31 ± 0.27	17.71
19	Pas. Jaflong	3	13.87 ± 0.69	1.14 ± 0.32	16.31
20	Pas. Jaflong	3	14.18 ± 0.81	1.21 ± 0.41	18.89
	Average Value		14.31 ± 0.59	1.32 ±0.19	17.09

*Calculated initial activity of ^{137}Cs in soil at the time after the Chernobyl nuclear power plant accident of 1986 on the basis of activity at present.

Activity Concentration and derived hazard parameters of ^{226}Ra, ^{232}Th and ^{40}K: The range and mean activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K of soil and water as well as external dose rate and other radiation hazard parameters corresponding to the activity in soil samples are shown in Table 2.

Table 2: Activity concentration of naturally occurring radionuclides in soil and water and derived external dose and other hazard parameters.

No. of location = 20		
No. of sample = 60		
Radionuclide	Activity in soil (Bq kg^{-1})	
	Range	Mean
^{226}Ra	7.19 ± 0.86 - 199.09 ± 1.25	57.48 ± 2.19
^{232}Th	28.81 ± 3.28 - 443.17 ± 19.79	167.60 ± 7.26
^{40}K	484.15±3.05 – 2064.28 ± 42.69	1202.27 ± 18.11
^{232}Th/^{226}Ra	4.01 - 2.22	2.92 ± 1.71

Radiation hazard indices	Range	Mean
Radium equivalent activities, Req (Bq kg^{-1})	121.60 ± 4.18 – 799.02 ± 33.24	391.163 ± 14.11
Representative level index values, $I_{\gamma r}$ (Bq kg^{-1})	1.11 ± 0.02 – 5.91 ± 0.24	2.83 ± 0.11
External dose rate at 1 m above the ground surface,	14.13 ± 2.31 – 383.67 ± 28.75	180.03 ± 8.17

D, (nGy h^{-1})		
Radionuclide	Activity in water (Bq L^{-1})	
	Range	Mean
^{226}Ra	5.71 ± 1.19 – 56.38 ± 3.287	26.57 ± 1.60
^{232}Th	42.73 ± 0.99 – 67.12 ± 1.24	52.69 ± 1.08
^{40}K	78.31 ± 1.28 – 130.56 ± 2.41	53.27 ± 1.58

Discussion

The anthropogenic radionuclide ^{137}Cs, deposited in the soil of Bangladesh presumably due to the results of fallout of radioactivity from the atmosphere following the nuclear power plant accident at Chernobyl on 26 April 1986 and other previous atmospheric tests of nuclear devices in the subcontinent or washout from which there may be a high input of ^{137}Cs. The activity of ^{137}Cs observed in the soil of *Goainghat* and *Jaintapur* area of Sylhet ranged from 5.22 ± 0.45 to 31.74 ± 0.89 Bq $kg^{-1}$with an average value of 14.31 ± 1.16 Bq kg^{-1}. As compared with the ^{137}Cs levels in the soil samples of other area of Bangladesh (Alam *et al*., 1995; Mollah *et al*., 1986; Mian *et al*., 1985), the level of ^{137}Cs in the study area is higher. The activity concentration of ^{137}Cs at *Goainghat* and *Jaintapur* in October 2000 and April 1986, on the basis of present activity, were calculated and are shown in Table 1. The absorbed dose rate in air (one meter above the ground surface) due to the radionuclides ^{226}Ra, ^{232}Th and ^{40}K in soil was estimated using the formula given in UNSCEAR (1988):

$$D = [0.427C_{Ra} + 0.662C_{Th} + 0.0432C_K]nGyh^{-1}$$

Where C_{Ra}, C_{Th} and C_K are the average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples in Bq kg^{-1}. The γ-ray absorbed dose rates due to the corresponding radionuclides in soil samples at 1 m above the ground surface were calculated and the range and mean values are shown in Table 2. The dose rates due to ^{226}Ra, ^{232}Th and ^{40}K in soil samples varied in the range 14.13 ± 2.31 to 383.67 ± 18.75 nGy h^{-1}, with an average value of 180.03 ± 8.17 nGy h^{-1}, which is higher than the world average value of 52 nGy h^{-1} (UNSCEAR, 1988).

***Raduim equivalent activities (Ra_{eq}) and representative level index ($I_{\gamma r}$) in soil*:** The γ-ray radiation hazards due to the radionuclides ^{226}Ra, ^{232}Th and ^{40}K in soil were assessed by two different indices. The most widely used radiation hazard index, Ra_{eq}, can be calculated as (Beretka and Mathew 1985):

$$Ra_{eq} = C_{Ra} + \left(\frac{10}{7}\right)C_{Th} + \left(\frac{10}{30}\right)C_K$$

Where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}, respectively. This equation is based on the estimate that 1 Bq kg^{-1} of ^{226}Ra, 0.7 Bq kg^{-1} ^{232}Th or 13 Bq kg^{-1} of ^{40}K generate the same gamma ray dose rate. The Ra_{eq} values for soil samples are shown in Table 2. The values of Ra_{eq} in soils varied from 121.60 ± 4.177 to 799.02 ± 25.24 Bq kg^{-1} with an average value of 391.16 ± 14.11Bq kg^{-1}. A radiation hazard index, used to estimate the level of γ-radiation hazard associated with the natural radionuclides in soil, representative level index, $I_{\gamma r}$, is defined as (NEA-OECD, 1979):

$$I_{\gamma r} = \left(\frac{C_{Ra}}{150} + \frac{C_{Th}}{100} + \frac{C_K}{1500}\right)$$

Where C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}. The $I_{\gamma r}$ value for soil samples are shown in Table 2. The values of $I_{\gamma r}$ for soil samples varied from 1.11 ± 0.02 to5.91 ± 0.24 Bq kg^{-1} with an average value of 2.82 ± 0.11Bq kg^{-1}.

The radium equivalent, Ra_{eq} and the representative level index $I_{\gamma r}$ values in the soil of Bangladesh are higher than the world average values. The world average values for Ra_{eq} and $I_{\gamma r}$ are 89.25 and 0.66 Bq kg^{-1}respectively (UNSCEAR, 1988).

The activity of ^{137}Cs was not yet found in any sample of water in any location of the country except the present study area, where the activity varied from 1.00 ± 0.34 to 2.21 ± 0.62 Bq L^{-1} with an average value of 1.02 ± 0.53 Bq L^{-1}.

The radioactivity of ^{226}Ra, ^{232}Th and ^{40}K in the present study is comparable with the radioactivity of the respective radionuclides in drinking water from Chittagong region of Bangladesh (Alam *et. al*., 1999), where the ranges were 12.0 to 82.0 Bq L^{-1}, 100.0 to 320.0 Bq L^{-1} and 50.6 to 120.7 Bq L^{-1}respectively.

Conclusion

The activity concentration of ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs in soil and water at *Jaintapur* and *Goainghat* in *Sylhet* were determined. From the data obtained two different indices of γ ray radiation hazards due to radionuclides ^{226}Ra, ^{232}Th and ^{40}K were assed viz. the Radium equivalent activities (Ra_{eq}) and Representative level index ($I_{\gamma r}$) as well as the external dose rate at one meter above the ground surface (D) were calculated. From the result it is being found that the average values of Ra_{eq}, $I_{\gamma r}$ and D in these two areas are higher than their respective world average value. The results would be useful in making base line data of these regions, in formulating sidelines for the radiation protection activities of the health physics, geophysics and environmental science.

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