

Evaluate the Potential Radiological Risks Due to Gamma Emitting Radiation from Soil Samples of Sunamganj area, Bangladesh

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Abstract

The present research work is encouraged by the increasing demand of the data belonging to the earthly γ-ray activities into the soil samples of haor area of Sunamgonj districts of Bangladesh. For measurements of naturally occurring radioactivity and artificial fall out, in the present work a total of 25 soil samples were collected from the eleven upazilas of this district. The samples were then prepared and irradiated for activity measurements using HPGe detecting system. The average concentration of radioactivity in soil samples for ^{238}U is 29.07 ± 2.84 Bq kg^{-1, 232}Th is 26.15±2.34 Bq kg⁻¹ and ⁴⁰K is 346.33±32.66 Bq Kg⁻¹, are below the world-wide average concentration value. No peak of man-made fission fragment ¹³⁷Cs (661.66 keV) was observed in the gamma-ray spectrum. The Radium Equivalent Activity, Dose Rate, External Hazard Index and Annual Effective Dose Equivalent for soil samples in Sunamgonj District has been estimated which are also comparable with worldwide average. The natural radioactivity in the present study poses no health-hazards to the population there. This study will be helpful to provide the possibility to accurately determine the public total effective dose rates due to natural radioactivity.

Key word: Soil sample, HPGe detecting system, Activity concentration, Radiological hazards parameters.

Introduction

Radionuclides are the sources of radioactivity and emit nuclear radiations which have become a part of our daily lives. The majority of naturally occurring radionuclides belong to the radionuclides in the ²³⁸U and ²³²Th series, and the single decay radionuclide, ⁴⁰K [1]. According to the National Council on Radiation Protection and Measurements Report, the most significant source of radiation exposure to humans is due to natural radiation in the environment [2]. This exposure to naturally occurring radiation also accounts for up to 85% of annual exposure dose

received by the world population and any dose of radiation involves a possible risk to human health [3]. Radiation is a natural part of the environment in which we live. All people receive exposure from natural origin (e.g., ²³⁸U, ²³²Th, ²²⁶Ra, ²²²Rn, ²¹⁰Pb, ⁴⁰K etc.), as well as anthropogenic ones (¹³⁷Cs, ¹³⁴Cs, ¹³¹I, ⁹⁰Sr, ⁸⁵Kr, ²³⁹Pu etc.) radionuclides in soil, water, air and food. The natural occurrence of 238U and 232Th in the earth's crust is a source of external exposure due to gammarays. The ²³⁸U decay series consists of 15 members in total including ²²⁶Ra and the series ends with stable ²⁰⁶Pb which is a stable element. After ²²⁶Ra, the series emits eight alpha and six beta particles along with many gamma-rays. There are 12 members in the decay series of ²³²Th and it decays into 208Pb after emitting seven alpha and five beta particles along with many gammarays. In addition, among the naturally occurring potassium isotopes, $40K$ is unstable, and decays by the emission of 1,460 keV gamma-rays and is also a source of gamma-rays. ¹³⁷Cs 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 ¹³⁷Cs world-wide. However, much of the ¹³⁷Cs from the testing has now been decayed. Nuclear reactor accident, such the Chernobyl accident (1986) in the Ukraine, releases ¹³⁷Cs to the environment [4-5]. 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 [6].

In order to protect the general public health against the radiation risk originating from Naturally Occurring Radioactive Material, the measurement of radioactivity in the environment needs to be considered to assess the biological effect on the human. This has also become the focus of greater attention by the IAEA in recent years [7]. The activity concentrations of ²³⁸U, 232 Th and 40 K and their progeny present in soil are measured in many countries of the world [8] including Bangladesh in order to monitor radiation level in the environment. The present study has been carried out to evaluate the level of radioactivity due to ²³⁸U, ²³²Th and ⁴⁰K in soil samples collected from the Sunamganj region of Sylhet Division. The aim of this research not only the determination of natural radioactivity in soil samples but also the determination of the radiological hazard parameters such as Gamma Dose Rate (D), Radium Equivalent Activity (Ra_{eq}), External Hazard Index (H_{ex}) and Annual Effective Dose Equivalent (D_{eff}) for individuals living in this region.

Materials And Methods:

Geological Outline

Sunamganj district is in the north-eastern part of Bangladesh (Figure 1). It is a border district between Bangladesh and India. It is located at in between 24°34' and 25°12' north latitudes and

in between 90°56' and 91°49' east longitudes. It was established as a district in 1984 and the area of Sunamganj district is 3669.58 square kilometers. Of the eleven upazilas of this district Dharmpasha is the largest (496.03 sq km) and Bishwambarpur is the smallest (194.25 sq km). The population of Sunamganj district is 2467968. Main sources of income Agriculture 66.99%, non-agricultural labourer 6.52%, industry 0.45%, commerce 9.12%, transport and communication 0.96%, service 3.81%, construction 0.64%, religious service 0.34%, rent and remittance 2.20% and others 8.97%. It is bounded by Meghalaya state of India on the north, Habiganj and Kishoreganj districts on the south, Sylhet district on the east, Netrokona district on the west. The geographical position and natural resources of Sunamganj indicate the likelihood of radioactivity but information on radioactivity is scarce as systematic measurements have not been undertaken in this area so far [9].

Figure 1: Map of collected soil sample from the eleven upazilas of Sunamganj district in Bangladesh.

Sample Collection and Preparation

International Journal of Life Sciences and Technology (2022), Volume 14, Issue 3, Page(s):29-44 Sunamganj district consist of the eleven upazilas. Total 25 Soil samples were collected from the eleven upazilas of this district. All samples were carefully collected at 5cm-10cm depth from the soil-surface in agriculture land by using shovel and each of the samples weighed approximately 1kg. At the laboratory of Health physics Division in Atomic Energy Center Dhaka, the collected samples were transferred from the polyethylene bags to the acetone-cleaned stainless-steel bucket and a crowbar was used to break the big soil stone separately. Then the samples were dried at 105°C until a constant weight was achieved and it was ensured that any significant moisture was removed from the samples. Each of the dried samples was grounded to fine powder in an agate motor separately. The powdered samples were then sieved using a fine aperture mesh screen (mesh size 2µm) in order to remove extraneous items like plant

material, roots, pebbles etc. and to obtain a fine-grained sample that would present a uniform matrix to the detector [10]. Finally, each of the grounded samples was transferred to marinelli beaker which capacity is 500 ml which Height: 4.2 inches (10.7 cm), Diameter: 4.9 inches (12.4 cm), Well Height: 2.7 inches (6.9 cm), Well Diameter: 3.03 inches (7.7 cm) and Endcap Diameter: 3.00 inches (7.6 cm) of Germenium detector. Marinelli Beaker sample containers are used in gamma spectrometry to maximize the efficiency of measurement systems when the radioactivity of the sample material is low and the limits of detection require special geometries. The beakers were then labelled properly and sealed tightly, rapped with thick vinyl tapes around their screw necks. The samples were stored for at least four weeks before counting in order to attain secular equilibrium.

Measurement procedures

A stand-alone high-resolution spectroscopic system is used for the measurement ofthe energy spectrum of the emitted gamma rays in the energy range between 60 and 2000 keV. The system consists of a high-purity germanium (HPGe) detector (coaxial cylinder of 55 mm in diameter and 73 mm in length) with an efficiency of 20% relative to a 3 × 3 NaI (Tl) scintillator. The detector is mounted on a cryostat which is dipped into a 30 l dewar, filled with liquid nitrogen. An advanced multi-channel analyzer (MCA) emulation software (MAESTRO-32) allows data acquisition, storage, display and online analysis of the acquired gamma-spectra. The effective volume of the detector was 83.469 cm³ and energy resolution of the 1.33 MeV energy peak for ⁶⁰Co was found as 1.69 keV at full width half maximum (FWHM). The detector is surrounded by a graded-Z cylindrical shield consisting of lead, iron, and aluminium, which provides an efficient suppression of background gamma radiation present at the laboratory site. The radioactivity concentration of²²⁶U was determined from γ-ray energies of its daughters ²¹⁴Pb (351.92 & 295.21 keV) and ²¹⁴Bi (609.31 & 1120.30 keV), while the ²³²Th was determined from γ-ray energies of its daughters ²¹²Pb (238.63 keV), ²⁰⁸Tl (583.14 keV) and ²²⁸Ac (911.07 & 969.11 keV). The radioactivity concentrations of $40K$ and $137Cs$ were determined from their γ -ray energy of 1460.80 keV and 662 keV respectively [10]. The energy-dependent detection efficiency has been determined using a calibrated ¹⁵²Eu gamma reference source. 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 (Figure 2). The geometry of the counting samples was the same as that of the standard samples and the counting time for all the sample was 10000 sec. 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.

Figure 2: Efficiency calibration Curve using Eu-152 gamma source

Theoretical calculations

The activity concentrations:

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

$$
A(Bq/kg) = \frac{C_a}{\varepsilon P_Y M_s}
$$

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).

The errors in the measurements were expressed in terms of standard deviation ($±σ$), where $σ$ is expressed as [11]:

$$
\sigma = \sqrt{\left[\frac{N_s}{T_s^2} + \frac{N_b}{T_b^2}\right]}
$$

Where, N_s is the sample counts measured in time T_s , and N_b is the background counts measured in time. The standard deviation $\pm \sigma$ in cps was converted into activity in Bqkg⁻¹. .

Radium equivalent activity

For the purpose of comparing the radiological effect or activity of materials that contain ²²⁶Ra, 232 Th and 40 K by a single quantity, 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 [8]:

 $Ra_{eq} = A_{Ra} + 1.43A_{Th} + 0.077A_{K}$

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

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 ²²⁶Ra, ²³²Th and ⁴⁰K was estimated using formula given in UNSCEAR, 1988[12]:

 $D=[0.427A_{Ra} + 0.662 A_{Th} + 0.0432 A_k]$ nGyh⁻¹

where $A_{R\omega}$, A_{Th} and A_k are the same meaning as in equation 2.

External Hazard index

The soils are used for manufacturing earthen huts, bricks and pottery materials and, hence, the external radiation hazard index, Hex and internal radiation hazard index were using the following relations [10]:

$$
H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_k}{4810} \le 1
$$

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

The values of those parameters used in the UNSCEAR report (2000) are 0.7 Sv. Gy⁻¹ for the conversion coefficient from absorbed dose in air to effective dose received by adults and 0.2 Sv. $\mathrm{Gy^1}$ for the outdoor occupancy factor. The annual effective dose equivalent can be estimated using the following formula [8]:

$$
D_{eff}(\mu Sv/y) = D(nGy/h) \times 8760h \times 0.2 \times 0.7Sv/Gy \times 10^{-3}
$$

Results and Discussion

Activity Concentrations in Soil Samples

The measurements made showed that the levels of radioactivity from the decay chain of ²³⁸U and ²³²Th, as well as the primordial radionuclide ⁴⁰K are observed in all soil samples. The activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K for different soil samples of Sunamgonj district are presented in Table 1. The highest activity concentrations for ²³⁸U and ²³²Th have been observed in Dakshin Sunamgonj and Jagannathpur soils which are 35.01 ± 3.04 Bq kg⁻¹ and 29.96 ± 1.09 Bq $kg⁻¹$ respectively, whilst the lowest activity concentrations for 238 U and 232 Th have been observed in Dharamapasha and Tahirpur which are 23.05 ± 1.70 Bq kg⁻¹ and 21.56 ± 3.37 Bq kg⁻¹, respectively (Table 1). The average concentrations of 238 U and 232 Th are measured as 29.07 ± 2.84 Bq kg⁻¹ and 26.15±2.34 Bq kg⁻¹, respectively. The highest activity concentration for ⁴⁰K have been observed in Derai which is 392 ± 0.74 Bq kg⁻¹ whilst the lowest activity concentration for ⁴⁰K have been observed in Jamalganj soil which is $285.74\pm$ 15.68 Bq kg⁻¹. The average concentration of $40K$ has been measured as 346.33 ± 32.66 Bq kg⁻¹.

Table 1. Activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in soil samples at different locations of Sunamgonj District.

The calculated activity concentrations of thorium, uranium and potassium are graphically presented in Figure-3 for 25 soil samples collected from the different upazilas of Sunamgonj district. It has been also observed that the measured activity concentration of ⁴⁰K exceeded markedly the values of both uranium and thorium as it is the most abundant radioactive element under consideration. The errors quoted were the standard deviations from the means and represent the spread in the concentrations of the natural radionuclides in the soil.

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Figure 3: Graphically shows that Activity concentrations in soil samples from different locations with sample code

The activity concentration of 238 U, 232 Th and 40 K in this study are comparable with other published results in Bangladesh in figure -4. The results of the present study are compared with those of the northern region at first. The ²³⁸U and ²³²Th concentrations of Sunamgonj District are about 3.19 and 5.85 times less than the high radioactivity zones of Rangpur, Lalmonirhat and Kurigram [10]. The ⁴⁰K activity concentration of the former is about 5.74 times less than the latter. ²³⁸U content in the Sunamgonj District is comparable with the values of other districts in Bangladesh except that in Sylhet where the value is about 53% higher. ²³²Th content in most of the districts are almost similar to this region except that in Sylhet and Kuakata Sea Beach where the values are 83% and 56% higher respectively. The concentration of $40K$ in this region is similar with the values in all other districts. The ²³⁸U and ²³²Th concentrations of Sunamgonj District are higher than Habiganj and Gopalgonj districts. No peak of man-made fission fragment ¹³⁷Cs (661.66 keV) was observed in the gamma-ray spectrum. It may be concluded that there is no ¹³⁷Cs radionuclide in the soil samples under study. However, it may be stated that the $137Cs$ activity was beyond the detection limit of 1.54 Bq in our measurement system. It seems that there is fresh nuclear fallout in the region studied.

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Figure 4: Comparison of Average activity concentration of ²³⁸U, ²³²Th and ⁴⁰K in soil samples of different countries and Worldwide average with the present study

Moreover, the average values obtained fell within the range of corresponding world values and other published results mentioned in Table 2. The world average activity concentration and ranges of 238 U, 232 Th , and $^{40}\rm{K}$ are 35 Bq kg 1 with a range of 17 - 60 Bq kg 1 , 30 Bq kg 1 with a range of 11 - 64 Bq kg $^{\text{-1}}$ and 400 Bq kg $^{\text{-1}}$ with a range of 140 - 850 Bq kg $^{\text{-1}}$, respectively [8]. The average activity concentrations for $238U$, $232Th$ and $40K$ in this study were lower than world averages for these radionuclides in the soils. In fact, the averages of this region are roughly equal to the world average. The average activity concentration for ²³⁸U and ²³²Th in present

study is higher than Denmark, Poland, Greece, Saudi Arabia, Nigeria, Turkey and lower than West Bank-Palestine, Pakistan, Luxembourg, Spain, Romania. The average activity concentration for ⁴⁰K in present study is higher than Saudi Arabia, Turkey and lower than Denmark, Poland, Nigeria, West Bank-Palestine, Pakistan, Luxembourg, Spain, Romania. It is similar to Greece. Investigations on terrestrial natural radiation have received particular attention worldwide and led to extensive surveys in many countries. They mainly serve as baseline data of natural radioactivity such that man made possible contaminations can be detected and quantitatively determined. They can further be used to assess public dose rates and to perform epidemiological studies. The results obtained in each country can be exploited to enrich the world's data bank, which is highly needed for evaluating worldwide average values of radiometric and diametric quantities.

Table 2. Comparison of Average activity concentration of ²³⁸U, ²³²Th and ⁴⁰K in soil samples of different countries and worldwide average with the present study

International Journal of Life Sciences and Technology (2022), Volume 14, Issue 3, Page(s):29-44 The values of the radiological hazard parameters are shown in Table -3. The Radium Equivalent Activity Ra_{eq} ranges from 82.40 to 105.99 Bq.kg⁻¹ with mean 93.15±6.48 Bq.kg⁻¹ for the

Sunamgonj District soil samples It is found that the Ra_{eq} values for all soil samples in the present work are lower than the accepted safety limit value of 370 Bq.kg⁻¹ as recommended by the Organization for Economic Cooperation and Development (OECD) [8]. The values are graphically shown in Figure 5(a). The estimated Absorbed Dose Rate D based on soil radioactivity ranges from 39.34 to 50.60 nGy.h^{.1} with a mean value 44.62±3.12 nGy.h^{.1} for the Sunamgonj District soil samples. These values are lower than the worldwide mean value 60 n G y.h⁻¹ [8]. The comparison is shown in Figure 5(b). As listed in Table 4, the calculated values of the External Hazard Index H_{ex} vary from 0.22 to 0.28 and the average value is found to be 0.25±0.017 for the Sunamgonj District. The results show that the Hex values from all soil samples are below the limit of unity, meaning that the radiation dose is below the permissible limit of 1 mSv.y 1 recommended by ICRP 60 [10]. This is displayed in Figure 5 (c). The Annual Effective Dose Equivalent D_{eff} from outdoor terrestrial gamma radiation was estimated by taking into account the conversion coefficients from absorbed dose in air to effective dose and the outdoor occupancy factor. The Annual Effective Dose Equivalent D_{eff} for the Sunamgonj District in this study vary from 48.0 to 62.0 µSv.y ¹ with mean value of 48.0 µSv.y^{.1}, which is similar with the worldwide effective dose of 80 μ Sv.y⁻¹ [10]. The values are graphically depicted in Figure 5(d).

Table 3. Radium Equivalent Activity, Dose Rate, External Hazard Index and Annual Effective Dose Equivalent for soil samples in Sunamgonj region.

5(a)

5(d)

Figure 5a, b, c & d: Comparison Radium Equivalent Activity, Dose Rate, External Hazard Index and Annual Effective Dose Equivalent for soil samples in Sunamgonj District with worldwide standard values.

Conclusions

High-resolution gamma-ray spectroscopy is a powerful experimental tool in studying natural radioactivity and determining activity concentrations and radiological hazard in soil samples. The concentrations and distributions of natural and anthropogenic radioactive materials in soils of the Sunamgonj District has been investigated with the aim of evaluating the environmental radioactivity and radiation hazard. The average activity concentrations of ²³⁸U

and ²³²Th in the samples of the Sunamgonj District are found to be similar with those in most districts in Bangladesh except in Sylhet and in the high radiation background of Rangpur, Lalmonirhat and Kurigram districts. The $40K$ concentrations in the samples of the present study are lower than those in the samples of all other districts in Bangladesh. The average activity concentrations of ²³⁸U and ²³²Th in the samples of the present study are lower than the world average. The average activity concentration of ⁴⁰K in the samples of the present study is lower than the world average. The artificial radionuclide ¹³⁷Cs was not observed in statistically significant amounts above background level in this study. It seems that there is no fresh nuclear fallout in places under study. The Radium Equivalent Activity Ra $_{eq}$ due to the radionuclides in the samples has been estimated. This value is found to be about 4 times less than the permissible maximum value of 370 Bq.kg-1 . The Absorbed dose rate D has been estimated from the soil activity of the present study which is lower than the worldwide mean value 60 nGy.h-1 . The External Hazard Index H_{ex} of the Sunamgonj District is found to be 0.25 which is much below the maximum limit of unity. The Effective Dose Equivalents D_{eff} of the present study is found to be 54µSv y ¹ which is similar with the worldwide effective dose of 80µSv y ¹. The natural radioactivity in the present study poses no health-hazards to the population there. This study will be helpful to provide the possibility to accurately determine the public total effective dose rates due to natural radioactivity.

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