Measurement of the Activity Concentrations of the Terrestrial Radionuclides from the Samples Collected from the Northern Part of Chittagong City Corporation, Chittagong, Bangladesh

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ABSTRACT

The concentrations of natural and artificial radionuclides in soil samples collected from the different areas of Chittagong City Corporation were measured by using Gamma-ray Spectrometry of High Purity Germanium (HPGe) detector. The concentration of $^{238}$U was found to be in the range 25.66 ± 5.48 Bq/Kg to 48.87 ± 6.00 Bq/Kg with an average value of 39.11 ± 5.77 Bq/Kg. The activity concentration of $^{232}$Th was found to be in the range of 27.12 ± 7.07 Bq/Kg to 64.71 ± 7.95 Bq/Kg with an average value of 45.65 ± 7.48 Bq/Kg, and the activity concentration of $^{40}$K was found to be in the range of 293.81 ± 75.36 Bq/Kg to 637.49 ± 81.88 Bq/Kg with an average value of 430.56 ± 76.79 Bq/Kg. The level of natural radioactivity in the region has been compared with the other areas of the world and the world average. In the soil samples, the overall activity concentration of Uranium and Potassium has been found to be higher than that of the world average, and the activity concentration of Thorium has been found to be nearly of the world average and the activity concentration. The artificial radionuclide $^{137}$Cs has not been detected in the samples.

Key Words: Activity Concentration, Natural Radionuclides, Gamma Ray Spectroscopy, World Average Values

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INTRODUCTION

The nucleus is the very dense region consisting of protons and neutrons at the center of an atom. Although the volume of a nucleus is negligible with compare to the total volume, but it contains the whole mass of an atom. The atomic nuclei of some isotopes have a surplus of energy, are unstable, and disintegrate to form more stable nuclei of different isotopes. This process is accompanied by the spontaneous emission of particles or energy, termed as nuclear radiation. Radioactivity is a phenomenon of spontaneous transformations resulting in the formation of new elements (Rao). These types of nuclides are called radionuclides, and this process is called as the radioactivity or nuclear decay or nuclear disintegration. Radioactivity discovered by A. H. Becquerel in 1896. Three different types of decays, known as alpha, beta, and gamma rays are associated with radioactivity. The alpha particle is nothing but helium nucleus, where beta is an electron and gamma is one type of electromagnetic radiation.

Therefore, alpha is highly positive, while the beta is negative and gamma is charge less radiation. Each of these reactions may be followed by gamma emission. In earth’s environment everywhere there is a certain amount of ionizing radiation (background radiation) or natural & artificial sources of radioactivity around everything. Consequently everything in the earth is continuously being exposed to ionizing radiation (Eisenbud 1962). The radioactivity process cannot be altered by any external means as atoms lose particles as heavy as nuclei of helium, they become atoms of some other element. That is the radioactive process a process in where a spontaneous transmutation occurs, and it will continue until the new generation is stable one. Irradiated to radiation even in low level, is considered undesirable because it may cause damage to public health and environment (Rutherford and Soddy).

METHODS AND MATERIALS

Study Area

For determining the radioactivity level of naturally occurring and anthropogenic radionuclides and hence for finding the radiological risk (i.e. dose rate) at the different wards of Chittagong city corporation in chittagong district especially South Pahartali (Nandirdigi, Baradigi, Natunpara); Jalalabad (Kulgong, Kuaesh, Bayezeed, Sholoshahar), Panchlaish (Shaheednagar, Kalurghatroad, Bahaddarhat); Chandgaong (Sulokbahar, Chawk-bazar road); West-Sholoshahar (Al-Falhmosque); Sulakbahar (SunniaMadrasha); North pahartali (Foyeslake, UttarKattali); SouthKattali, Saraipara, Pahartali, Chawkbazar, West-Bkalia. The total number of 21 samples was collected from different locations at the 5cm depth of different wards in Chittagong City Corporation.

Systematic grid sampling:

Systematic grid sampling involves subdividing the area of concern by using a square, triangular or herringbone grid and collecting samples from the nodes (intersections of the grid lines). The origin and direction for placement of the grid are done using an initial random point. From that point, a coordinate axis and grid are constructed over the whole site. The distance between sampling locations in the systematic grid is determined by the size of the area to be sampled and the number of samples to be collected. Systematic grid sampling is often used to delineate the extent of contamination and to define contaminant concentration gradients (IAEA-TECDOC-1415, 2004).
The rough distance between two samples would be about 2 kilometers, but due to the different difficulties, the distance could not maintain accurately, I have taken sample from that place which is easy to access. Because different sample locations were highway road, industry, building and face to various obstruct. The geographical locations of all sampling points were recorded by using GPS machine collected from Geography department in the University of Chittagong. With counting radiation of sampling point in dose rate unit of mr/hr by Survey meter.

In γ-ray spectrometry, the full energy peak efficiency of a high purity Germanium (HpGe) detector is the number of γ- rays detected by the detector to the number of photons emitted by the source for specific energy, which is defined as (6)

\[ \varepsilon (E) = \frac{n(E)}{A \times I_\gamma} \]

(1)

Where, \(n(E)\) is the net count rate of the photo peak for the corresponding energy \(E\), \(A\) is the present activities of the standard reference source which were calculated by using the well-known decay law: \(A = A_0 e^{-\lambda t}\). And \(I_\gamma\) is the Intensity of the gamma energy.

In the present study, the International Atomic Energy Agency (IAEA) reference samples were used for the calibration of detector efficiencies. The IAEA reference samples are: RGU-
1. Uranium is in a silica matrix, RGTh-1: Thorium is in silica matrix and RGK-1: Potassium Sulphate. The standard reference source has the same diameter as the soil samples of known concentrations of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ radionuclides supplied by the Canada Centre for Mineral and Energy Technology (CAMET) under a contract with the IAEA.

**Sample analysis**

The detection and measurement of radionuclides in the samples were carried out by gamma spectrometry system using a vertical coaxial cylindrical HPGe detector of 172 cm$^3$ active volume with 38% relative efficiency. The p-type HPGe detector supplied by CANBERRA (Model IGG 2020) had a resolution of 2 keV at 1332 keV of $^{60}\text{Co}$. The detector was coupled to a 16 k-channel computer analyzer. The analysis was carried out using a Genie 2000 software, which matched various gamma energy peaks to a library of possible radionuclides. The detector was enclosed in a cylindrical shielding container made of lead and iron with 11.3 cm thickness, 51 cm height and 28 cm internal diameter and with a fixed bottom and moving cover to reduce the external $\gamma$-ray background. All the samples were counted for 50 ks. The energy regions selected for the corresponding radionuclides were 295 keV and 352 keV of $^{214}\text{Pb}$ and 609 keV, 1120 keV and 1764 keV of $^{214}\text{Bi}$ for $^{226}\text{Ra}$, 583 keV and 2614 keV of $^{208}\text{Tl}$, 911 keV and 969 keV of $^{228}\text{Ac}$ for $^{228}\text{Th}$ and 1460 keV for $^{40}\text{K}$ (Roessler et al. 1970).

**Calibration of HPGe detector**

In the present study, the calibration for the efficiency of the detector was performed by the International Atomic Energy Agency (IAEA) reference samples of solid. The IAEA reference samples are: RGU-1, Uranium is in a silica matrix, RGTh-1: Thorium is in silica matrix and RGK-1: Potassium Sulphate. The standard reference source has the same diameter as the soil samples of known concentrations of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ radionuclides supplied by the Canada Centre for Mineral and Energy Technology (CAMET) under a contract with the IAEA. The detector efficiency calibration curves as a function of energy for both solid matrices have been shown in Figure 3.

![Figure 3: Counting efficiency curve of the HPGe Detector (For solid standard sample)](image-url)
Calculation of Activity Concentration

The radioactivity of each sample was measured using the calibrated high purity Germanium (HpGe) Detector of energy resolution of 2.0 KeV at 1.33 MeV of \(^{60}\)Co. For a period of 20,000 s. keeping the samples one by one on the top of the detector and counted for a period of 20,000 s. The activity concentration (A) of each radionuclide in the sample was determined by using the net count rates (Nc) after subtracting the background counts from the gross counts for the same counting time under the selected photo peaks, weight of the sample, the photo peak efficiency and the gamma intensity at a specific energy as:

\[ A = \frac{(Nc \times 1000)}{\varepsilon \times I_{\gamma} \times W} \] ……………..(2)

Where A = Activity concentration of the sample in Bq/.Kg Net count rate, Nc = Gross counts per second from the samples - background counts per second \( \varepsilon = \) Efficiency of the detector for the specific energy. \( I_{\gamma} = \) Intensity of the gamma ray. \( W = \) Sample weight in gm. For the analysis of peak areas of gamma spectra, a Computer software programming (GENIE 2000) was used.

RESULTS AND DISCUSSION

Activity Concentrations of all Samples

Activity concentrations for nuclides \(^{238}\)U, \(^{232}\)Th, \(^{40}\)K and \(^{137}\)Cs in Chittagong Urea Fertilizer limited (CUFL) samples were determined by equation (2) and the results for the same have been shown in Table 1 with the uncertainty of 1σ level. Here, the table shows that the highest value is found for sample S-3. The highest value of the nuclide may vary from place to place and this variation may it is a chemical composite [First Reformer Catalyst, Ni]. Moreover, it is also possible due to the excess use of agricultural fertilizers and pesticides. The lowest value is found for sample L-5 because it is a liquid sample. All the associated errors were added in quadrature to obtain the total uncertainty. The results for the nuclides \((^{238}\)U, \(^{232}\)Th and \(^{40}\)K) are also shown independently through Figure 3.

Table 1: The comparative data of the activity concentrations of parent radionuclides, \(^{238}\)U, \(^{232}\)Th, \(^{40}\)K and \(^{137}\)Cs of all soil samples

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Activity of (^{238})U, (Bq.Kg(^{-1})) with (±1σ)</th>
<th>Activity of (^{232})Th, (Bq.Kg(^{-1})) with (±1σ)</th>
<th>Activity of (^{40})K, (Bq.Kg(^{-1})) with (±1σ)</th>
<th>Activity of (^{137})Cs, (Bq.Kg(^{-1})) with (±1σ)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AH-01</td>
<td>48.87 ± 6.00</td>
<td>64.71 ± 7.95</td>
<td>494.09 ± 78.79</td>
<td>ND</td>
</tr>
<tr>
<td>AH-02</td>
<td>38.64 ± 5.68</td>
<td>53.79 ± 7.64</td>
<td>536.96 ± 61.03</td>
<td>ND</td>
</tr>
<tr>
<td>AH-03</td>
<td>41.12 ± 5.75</td>
<td>45.00 ± 7.47</td>
<td>449.67 ± 77.94</td>
<td>ND</td>
</tr>
<tr>
<td>AH-04</td>
<td>40.52 ± 5.77</td>
<td>51.49 ± 7.62</td>
<td>463.70 ± 78.20</td>
<td>ND</td>
</tr>
<tr>
<td>AH-05</td>
<td>41.57 ± 5.83</td>
<td>43.60 ± 7.41</td>
<td>477.73 ± 78.47</td>
<td>ND</td>
</tr>
<tr>
<td>AH-06</td>
<td>40.69 ± 5.81</td>
<td>40.99 ± 7.46</td>
<td>331.99 ± 75.93</td>
<td>ND</td>
</tr>
<tr>
<td>AH-07</td>
<td>45.65 ± 5.89</td>
<td>45.92 ± 7.45</td>
<td>293.81 ± 75.36</td>
<td>ND</td>
</tr>
<tr>
<td>AH-08</td>
<td>31.21 ± 5.52</td>
<td>52.74 ± 7.65</td>
<td>406.03 ± 77.15</td>
<td>ND</td>
</tr>
<tr>
<td>AH-09</td>
<td>38.80 ± 5.73</td>
<td>44.49 ± 7.48</td>
<td>335.89 ± 75.99</td>
<td>ND</td>
</tr>
<tr>
<td>AH-10</td>
<td>42.37 ± 5.92</td>
<td>43.51 ± 7.41</td>
<td>416.94 ± 77.34</td>
<td>ND</td>
</tr>
<tr>
<td>AH-11</td>
<td>39.55 ± 5.81</td>
<td>48.64 ± 7.51</td>
<td>353.03 ± 76.26</td>
<td>ND</td>
</tr>
<tr>
<td>AH-12</td>
<td>25.66 ± 5.48</td>
<td>30.01 ± 7.23</td>
<td>360.05 ± 76.37</td>
<td>ND</td>
</tr>
</tbody>
</table>
The Activity of $^{238}$U: The specific activity of $^{238}$U in the soil samples has been found in the range of $25.66 \pm 5.48$ to $48.87 \pm 6.00$ Bq/Kg with an average value of $39.11 \pm 5.77$ Bq/Kg. The comparison of activity concentrations of $^{238}$U of all samples is shown in table 1. The variation of the mean activity concentration of $^{238}$U of present result with worldwide value and other study areas of the world has been shown in table 2 and corresponding graphical representation in figure 4 and the comparison of the mean result of $^{238}$U of the present study with worldwide value and other study has been shown in figure 7.

![Figure 4: Activity concentration of all samples with average world value](image)

The Activity of $^{232}$Th: The activity concentration of $^{232}$Th has been obtained in a range from $27.12 \pm 7.07$ Bq/Kg to $64.71 \pm 7.95$ Bq/Kg with an average value of $45.65 \pm 7.48$ Bq/Kg.

The comparative data of the activity concentrations of $^{232}$Th of all samples has been shown in table 1 and their corresponding graphical representation has been shown in figure 5. The comparison of the activity concentrations of the present study with the worldwide value and other study areas shown in table 2 and their graphical representation in figure 7.
The Activity of $^{40}$K: The mean activity concentration of $^{40}$K in the soil has been found in the range started from 293.81 ± 75.36 Bq. Kg$^{-1}$ to 637.49 ± 81.88 Bq. Kg$^{-1}$ with an average value of 430.56 ± 76.79 Bq/ Kg. The distribution of the activity concentrations of $^{40}$K of all samples has been shown in table 1. The comparison of the activity concentrations of the present study with the worldwide value and average value are shown in table 2 and corresponding graphical representation in figure 7.

The Activity of $^{137}$Cs: The artificial radionuclide $^{137}$Cs has been detected in some samples but it was too small to take into account. The activity concentration of $^{137}$Cs was from 2 to 5 Bq.Kg$^{-1}$, which is negligible in any comparison with the world’s standard (1000 Bq/kg for $^{137}$Cs set by IAEA) and Bangladesh standard (50 Bq/kg for $^{137}$Cs set by Bangladesh Atomic Energy Commission).
Mean activity concentration with average world value:

Now the comparison of the activity concentration of present study result with worldwide activity concentration is shown in below:

Table 2: The activity concentration in some region also worldwide value

<table>
<thead>
<tr>
<th>Sl. No</th>
<th>Reference</th>
<th>Area</th>
<th>$^{238}$U (Bq kg$^{-1}$)</th>
<th>$^{232}$Th (Bq kg$^{-1}$)</th>
<th>$^{40}$K (Bq kg$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>01</td>
<td>Present study</td>
<td>Along the Chittagong City Corporation</td>
<td>$39.11 \pm 5.77$</td>
<td>$45.65 \pm 65$</td>
<td>$430.56 \pm 76.79$</td>
</tr>
<tr>
<td>02</td>
<td>Worldwide</td>
<td>UNSCEAR, (2000)</td>
<td>$33(17-60)$</td>
<td>$45(11-64)$</td>
<td>$420(140-850)$</td>
</tr>
<tr>
<td>03</td>
<td>China</td>
<td>P. Ziqiang</td>
<td>$61.5 \pm 37$</td>
<td>$89.8 \pm 74$</td>
<td>$524 \pm 162$</td>
</tr>
<tr>
<td>04</td>
<td>USA</td>
<td>R.D. Delune</td>
<td>$34 \pm 29$</td>
<td>$36 \pm 12$</td>
<td>$472 \pm 223$</td>
</tr>
</tbody>
</table>

Figure 7: Comparison of mean activity concentration of $^{238}$U, $^{232}$Th and $^{40}$K respectively.

**CONCLUSION**

There have no chance to deny that radioactivity and radiation have become a great threat to humans as well as aquatic animals due to both are exposed to ionizing radiation and the effects of radiation after certain levels are very detrimental. So the purpose of my research work is to study the distribution of radionuclides, their concentrations due to different radionuclides present in the soil samples. In the present work, the concentration of natural radionuclides $^{238}$U, $^{232}$Th, and $^{40}$K and anthropogenic radionuclides $^{137}$Cs in different soil samples were analyzed by HPGe gamma-ray spectrometry. In the present study, the average activity concentration of $^{238}$U, $^{232}$Th, and $^{40}$K were measured $39.11 \pm 5.77$, $45.65 \pm 7.48$ and $430.56 \pm 76.79$ Bq kg$^{-1}$ respectively. In this study, the presence of anthropogenic radionuclide $^{137}$Cs was not observed. This is because the concentration of this radionuclide is below detectable range and we can ignore its presence in the soil samples of Chittagong city corporation area. This data may be used in future as base line data, so that we can able to
decide that if nuclear waste or fallout affected our City Corporation area or not. The anthropogenic radionuclides concentration, particularly the level of $^{137}$Cs, present in the soil sample at the negligible amount, whereas no such radioactivity exists in other samples of investigated areas. The level of fall-out radionuclide is much below the world’s standard (1000 Bq/kg for $^{137}$Cs set by IAEA) and Bangladesh standard (50 Bq/kg for $^{137}$Cs set by Bangladesh Atomic Energy Commission). The research breakthrough indicates that the investigated parts of the Chittagong City Corporation pose no threat to flora and fauna and thereby for human health at large. Further research needs to be finding out in other parts of the Chittagong City Corporation to prepare a research-data-bank for the radioactivity level for the researchers. The output may work as a useful guideline for the government agencies in taking the proper initiatives regarding radiation hazard in the Chittagong City Corporation.

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