Abstract: In the present study, natural environmental radioactivity in fly ash samples collected from Kasimpur Thermal Power Plant has been measured by low level gamma ray spectrometer. The activity concentration of $^{238}$U and $^{232}$Th varies from 61.45±1.37 Bq kg$^{-1}$ to 112.66±2.15 Bq kg$^{-1}$ with an average value of 100.66±1.92 and from 68.04±1.70 Bq kg$^{-1}$ to 117.08±2.49 Bq kg$^{-1}$ with a mean value of 101.84±2.17 Bq kg$^{-1}$ respectively. $^{40}$K in fly ash samples ranges from 236.06±3.39 Bq kg$^{-1}$ to 520.03±6.63 Bq kg$^{-1}$ with an average value of 461.08±5.99 Bq kg$^{-1}$. Calculated values of radium equivalent activity (Ra$_{eq}$) varies from 175.27 Bq kg$^{-1}$ to 308.39 Bq kg$^{-1}$ with an average value of 278.56 Bq kg$^{-1}$. Total absorbed gamma dose rates are found to vary from 79.33 nGy h$^{-1}$ to 140.46 nGy h$^{-1}$ with an average value of 127.24 nGy h$^{-1}$. Indoor and outdoor annual effective dose rate from these fly ash samples are determined from 0.39 mSv y$^{-1}$ to 0.69 mSv y$^{-1}$ and 0.09 mSv y$^{-1}$ to 0.17 mSv y$^{-1}$ respectively. Computed values of $H_a$ vary from 0.64 to 1.14 with an average value of 1.01. External hazard index, $H_e$, for the soil samples studied in this work range from 0.48 to 0.84 with a mean value of 0.76.

1 INTRODUCTION

Environment contains some naturally occurring radioactive materials (NORM) which are found in soils, rocks, vegetation, air, water and also in building materials [1]. Radioactivity is a part of the natural environment [2]. Gamma radiation has always been existed in environment since the big-bang occurred due to the long half-lives of the radionuclides from the $^{238}$U and $^{232}$Th series, and their decay products [3]. Historical antecedents of studies conducted on natural radioactivity have established that the presence of the uranium-thorium series and potassium-40 in various materials constitute potential exposure to the global population [4]. Natural materials contain various trace elements including uranium, thorium and their radioactive decay products. Coal is no exception and the combustion of pulverized coal in power station boilers results in concentrating some 30-40 trace elements, including radioactive nuclides in the mineral residue, in fly ash and bottom ash [5-8]. Fly ash is an aluminosilicate glass consisting of oxides of Si, Al, Fe and Ca with minor amounts of Hg, Na, K, Ti, S and various trace elements including the radioactive elements $^{238}$U, $^{232}$Th and $^{40}$K [9,10]. In a power plant consuming 10 tonne of coal per day, the mobilization of radioactivity to the atmosphere due to $^{238}$U alone is about 1850kBq [11]. Uranium-238, the parent element of the uranium series has a half-life of 4.5x10$^9$ yr. and supports a series of 13 main decay products and several other radionuclides including radon which is considered to be the largest single source of radiation exposure to the human population [12,13]. Analyses of coal and fly ash indicate the enrichment of trace elements in fly ash [14,15]. Indian coal has a high ash content and the total fly ash production from thermal power plants is estimated to be ca 64 million tonne per year, assuming 40% ash content in coal [16]. Fly ash has found wide application world-wide for cement and sand replacement in pre-mixed concrete, for the manufacture of blended fly ash-Portland cement, aerated concrete, fly ash clay bricks and blocks, for road making and as a filler in asphalt. The use of fly ash products, especially as building materials, may result in radiation exposure of humans to unacceptable levels of radiation [17,18]. Thus it is quite important to estimate the radiation risk to the population from the radioactivity of fly ash. In the present study, environmental natural radioactivity in fly ash samples collected from Kasimpur Thermal Power Plant has been measured by low level gamma ray spectrometer. The study is important from environmental radiological point of view.
2 EXPERIMENTAL TECHNIQUE

Gamma ray spectrometric measurements were carried out at Inter-University Accelerator Centre, New Delhi, India using a coaxial n-type HPGe detector (EG&G, ORTEC, Oak Ridge, USA) for estimation of the natural radionuclides, Uranium \(^{238}\)U), thorium \(^{232}\)Th) and potassium \(^{40}\)K). The samples were crushed into fine powder by using Mortar and Pestle. Fine quality of the sample is obtained by using scientific sieve of 150 micron-mesh size. Before measurements samples were oven dried at 110°C for 24h and the samples were then packed and sealed in an impermeable airtight PVC container to prevent the escape of radiogenic gases radon \(^{222}\)Rn) and thoron \(^{220}\)Rn). About 300g sample of each material was used for measurements. Before measurements, the containers were kept sealed about 4 weeks in order to reach equilibrium of the \(^{238}\)U and \(^{232}\)Th and their respective progenies. After attainment of secular equilibrium between \(^{238}\)U and \(^{232}\)Th and their decay products, the samples were subjected to high resolution gamma spectroscopic analysis. HPGe detector (EG&G, ORTEC, Oak Ridge, USA) having a resolution of 2.0 keV at 1332 keV and a relative efficiency of 20% was placed in 4” shield of lead bricks on all sides to reduce the background radiation from building materials and cosmic rays [19]. The detector was coupled to a PC based 4K multi channel analyzer and an ADC for data acquisition. The calibration of the low background counting system was done with a secondary standard which was calibrated with the primary standard (RGU-1) obtained from the International Atomic Energy Agency (IAEA). The efficiency for the system was determined using secondary standard source of uranium ore in the same geometry as available for the sample counting. For activity measurements the samples were counted for a period of 72000 seconds. The activity concentration of \(^{40}\)K (C\(_K\)) was measured directly by its own gamma ray of 1461 keV. As \(^{238}\)U and \(^{232}\)Th are not directly gamma emitters, their activity concentrations (C\(_U\) and C\(_Th\)) were measured through gamma rays of their decay products. Decay products taken for \(^{238}\)U were \(^{214}\)Pb: 295 and 352 keV and \(^{214}\)Bi: 609, 1120 and 1764 keV whereas for \(^{232}\)Th were \(^{228}\)Ac : 338, 463, 911 and 968 keV, \(^{212}\)Bi : 727 keV, \(^{212}\)Pb : 238 keV and \(^{234}\)Pa : 1001 keV gamma ray by assuming the decay series to be in equilibrium [20]. Weighted averages of several decay products were used to estimate the activity concentrations of \(^{238}\)U and \(^{232}\)Th. The gamma ray spectrum of typical flyash sample is shown in Fig. 1. was analyzed using the locally developed software “CANDLE” (Collection and Analysis of Nuclear Data using Linux Net work). The net count rate under the most prominent photo peaks of radium and thorium daughter peaks are calculated from respective count rate after subtracting the background counts of the spectrum obtained for the same counting time. Then the activity of the radionuclide is calculated from the background subtracted area of prominent gamma ray energies. The concentration of uranium, thorium and potassium is calculated using the following equation:

\[
\text{Activity (Bq.kg}^{-1}\) = \frac{(S \pm \sigma) \times 100 \times 1000 \times 100}{E \times W \times A}
\]

Where S is the net counts/sec (cps) under the photo peak of interest, \(\sigma\) the standard deviation of S, E the counting efficiency (%), A the gamma abundance or branching intensity (%) of the radionuclide and W is the mass of the sample (Kg).

The concentrations of Uranium, Thorium and Potassium are calculated using the following equation:

\[
\text{Activity} = \text{CPS} \times 100 \times 100 / \text{B.I.} \times \text{Eff} \pm \text{CPS_error} \times 100 \times 100 / \text{B.I.} \times \text{Eff}
\]

Where, CPS - Net count rate per second
B.I. - Branching intensity, and
E - Efficiency of the detector

3 COMPUTATION OF RADIOLOGICAL EFFECTS

From the activity concentrations of the natural radionuclides, uranium \(^{238}\)U), thorium \(^{232}\)Th) and potassium \(^{40}\)K), radiological parameters can be calculated. The results are of great interest in the environmental radiological protection study, since the samples may widely be used as building construction materials.
3.1 Radium equivalent activity (Ra eq)
Exposure to radiation is defined in terms of radium equivalent activity (Ra eq) in Bq kg⁻¹ to compare the specific activity of materials containing different amounts of ²³⁸U (²²⁶Ra), ²³²Th and ⁴⁰K. It is calculated by the following expression [21,22]:

\[ \text{Ra eq} = C_U + 1.43C_{Th} + 0.07C_K \]  
(3)

Where \( C_U \), \( C_{Th} \) and \( C_K \) are the activity concentrations of ²³⁸U, ²³²Th and ⁴⁰K in Bq kg⁻¹ respectively. In the above equation for defining Ra eq activity it has been assumed that the same gamma dose rate is produced by 370 Bq kg⁻¹ of ²³⁸U or 259 Bq kg⁻¹ of ²³²Th or 4810 Bq kg⁻¹ of ⁴⁰K. There will be variations in the radium equivalent activities of different materials and also within the same type of materials. The results may be important from the point of view of selecting suitable materials for use in building construction materials.

3.2 Absorbed gamma dose rate measurement (D)
Outdoor air absorbed dose rate D in nGy h⁻¹ due to terrestrial gamma rays at 1m above the ground can be computed from the specific activities, \( C_U \), \( C_{Th} \) and \( C_K \) of ²³⁸U/²²⁶Ra, ²³²Th and ⁴⁰K in Bq kg⁻¹ respectively by Monte Carlo method [23]:

\[ D \text{ (nGy h}^{-1}) = 0.462C_U + 0.604C_{Th} + 0.0417C_K \]  
(4)

To estimate the annual effective dose rate, E, the conversion coefficient from absorbed dose in air to effective dose (0.7 Sv Gy⁻¹) and outdoor occupancy factor (0.2) proposed by UNCSEAR (2000) were used. The indoor effective dose rate in units of mSv y⁻¹ was calculated by the following relation:

\[ E \text{ (mSv y}^{-1}) = \text{Dose rate (nGy h}^{-1}) \times 8760 \text{ h} \times 0.8 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6} \]  
(5)

The outdoor effective dose rate in units of mSv y⁻¹ was calculated by the following relation:

\[ E \text{ (mSv y}^{-1}) = \text{Dose rate (nGy h}^{-1}) \times 8760 \text{ h} \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6} \]  
(6)

3.3 External (H ex) and Internal (H in) hazard index
The external hazard index is obtained from Ra eq expression through the supposition that its allowed maximum value (equal to unity) corresponds to the upper limit of Ra eq (370 Bq kg⁻¹). For limiting the radiation dose from building materials in Germany to 1.5 mGy y⁻¹. Krieger (1981) proposed the following relation for H ex:

\[ H_{ex} = \frac{C_U}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \]  
(7)

This criterion considers only the external exposure risk due to γ-rays and corresponds to maximum Ra eq of 370 Bq kg⁻¹ for the material. These very conservative assumptions were later corrected and the maximum permission concentrations were increased by a factor of 2 [24] which gives

\[ H_{ex} = \frac{C_U}{740Bqkg^{-1}} + \frac{C_{Th}}{520Bqkg^{-1}} + \frac{C_K}{9620Bqkg^{-1}} \leq 1 \]  
(8)

Internal exposure to ²²²Rn and its radioactive progeny is controlled by the internal hazard index (H in) as given below [25].

\[ H_{in} = \frac{C_U}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \]  
(9)

4 RESULTS AND DISCUSSION
The gamma ray spectrum of a fly ash sample is shown in Fig 1.

The measured activity concentration of ²³⁸U, ²³²Th and ⁴⁰K together with their average values in fly ash samples collected from Kasimpur Thermal Power Plant (U.P) are given in Table 1. Computed values of radium equivalent activity, absorbed gamma dose rate, annual effective doses, external hazard index and internal hazard index in fly ash samples are given in Table 2.
Table 1
Activity concentration of $^{238}\text{U}$, $^{232}\text{Th}$, and $^{40}\text{K}$ in fly ash samples

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>$^{238}\text{U}$ (Bq kg$^{-1}$)</th>
<th>$^{232}\text{Th}$ (Bq kg$^{-1}$)</th>
<th>$^{40}\text{K}$ (Bq kg$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>KF1</td>
<td>112.66 ± 2.35</td>
<td>108.01 ± 2.35</td>
<td>520.05 ± 6.85</td>
</tr>
<tr>
<td>KF2</td>
<td>93.15 ± 2.14</td>
<td>98.83 ± 2.01</td>
<td>551.96 ± 3.75</td>
</tr>
<tr>
<td>KF3</td>
<td>112.76 ± 2.18</td>
<td>108.32 ± 2.29</td>
<td>497.32 ± 2.08</td>
</tr>
<tr>
<td>KF4</td>
<td>81.45 ± 2.39</td>
<td>81.04 ± 2.70</td>
<td>220.06 ± 2.59</td>
</tr>
<tr>
<td>KF5</td>
<td>101.72 ± 2.35</td>
<td>103.12 ± 2.35</td>
<td>402.19 ± 2.37</td>
</tr>
<tr>
<td>KF6</td>
<td>104.13 ± 2.12</td>
<td>101.34 ± 2.35</td>
<td>479.98 ± 2.65</td>
</tr>
<tr>
<td>KF7</td>
<td>90.36 ± 2.12</td>
<td>97.36 ± 2.81</td>
<td>444.29 ± 2.64</td>
</tr>
<tr>
<td>KF8</td>
<td>110.62 ± 2.10</td>
<td>107.05 ± 2.27</td>
<td>510.96 ± 2.64</td>
</tr>
<tr>
<td>KF9</td>
<td>102.01 ± 2.32</td>
<td>102.21 ± 2.33</td>
<td>489.09 ± 2.47</td>
</tr>
<tr>
<td>KF10</td>
<td>103.45 ± 2.13</td>
<td>101.04 ± 2.81</td>
<td>258.00 ± 2.65</td>
</tr>
</tbody>
</table>

It is apparent from Table 1 that the activity concentration of $^{238}\text{U}$ and $^{232}\text{Th}$ varies from 61.45 ± 1.37 Bq kg$^{-1}$ to 112.66 ± 2.15 Bq kg$^{-1}$ with an average value of 100.66 ± 1.92 Bq kg$^{-1}$ and from 68.04 ± 1.70 Bq kg$^{-1}$ to 117.08 ± 2.49 Bq kg$^{-1}$ with a mean value of 101.84 ± 2.17 Bq kg$^{-1}$, respectively. $^{40}\text{K}$ in fly ash samples ranges from 236.06 ± 3.39 Bq kg$^{-1}$ to 520.03 ± 6.63 Bq kg$^{-1}$ with an average value of 461.08 ± 5.99 Bq kg$^{-1}$. To establish the correlation between $^{238}\text{U}$ and $^{232}\text{Th}$ activity concentration and $^{232}\text{Th}$ and $^{40}\text{K}$ activity concentrations in the fly ash samples of the present study, the values are plotted as shown in Figs. 2 & 3. A positive correlation exists between $^{238}\text{U}$ and $^{232}\text{Th}$ activity concentration and a Positive correlation between $^{232}\text{Th}$ and $^{40}\text{K}$ activity concentration in the fly ash samples studied here.

Correlation Coefficient (R) = .92349

Table 2
Radium equivalent activity, Absorbed gamma dose rate, Annual effective doses, External hazard index and Internal hazard index in fly ash samples.

<table>
<thead>
<tr>
<th>Sample Code</th>
<th>Radium Equivalent Activity (Bq kg$^{-1}$)</th>
<th>Absorbed Gamma Dose Rate (mGy h$^{-1}$)</th>
<th>Annual Effective Dose (mSv)</th>
<th>External Hazard Index (HL)</th>
<th>Internal Hazard Index (IL)</th>
</tr>
</thead>
<tbody>
<tr>
<td>KF1</td>
<td>254.75</td>
<td>0.04</td>
<td>0.13</td>
<td>0.32</td>
<td>1.14</td>
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<tr>
<td>KF2</td>
<td>257.54</td>
<td>0.19</td>
<td>0.12</td>
<td>0.37</td>
<td>0.98</td>
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<tr>
<td>KF3</td>
<td>217.78</td>
<td>0.67</td>
<td>0.17</td>
<td>0.31</td>
<td>1.12</td>
</tr>
<tr>
<td>KF4</td>
<td>151.25</td>
<td>0.29</td>
<td>0.08</td>
<td>0.16</td>
<td>0.64</td>
</tr>
<tr>
<td>KF5</td>
<td>215.41</td>
<td>0.85</td>
<td>0.16</td>
<td>0.76</td>
<td>1.68</td>
</tr>
<tr>
<td>KF6</td>
<td>257.05</td>
<td>0.28</td>
<td>0.12</td>
<td>0.73</td>
<td>1.73</td>
</tr>
<tr>
<td>KF7</td>
<td>258.71</td>
<td>0.29</td>
<td>0.12</td>
<td>0.71</td>
<td>1.72</td>
</tr>
<tr>
<td>KF8</td>
<td>258.41</td>
<td>0.87</td>
<td>0.18</td>
<td>0.32</td>
<td>1.32</td>
</tr>
<tr>
<td>KF9</td>
<td>216.89</td>
<td>0.24</td>
<td>0.16</td>
<td>0.18</td>
<td>1.01</td>
</tr>
<tr>
<td>KF10</td>
<td>210.25</td>
<td>0.20</td>
<td>0.12</td>
<td>0.34</td>
<td>1.11</td>
</tr>
</tbody>
</table>

Average Values

<table>
<thead>
<tr>
<th>B</th>
<th>Linear Fit of Data1_B</th>
</tr>
</thead>
<tbody>
<tr>
<td>60</td>
<td>70</td>
</tr>
<tr>
<td>60</td>
<td>70</td>
</tr>
</tbody>
</table>

Fig 2 Variation of $^{238}\text{U}$ activity concentration versus $^{232}\text{Th}$ activity in fly ash samples.
Indoor and outdoor annual effective dose rate from these fly ash samples are determined from 0.39 mSv y⁻¹ to 0.69 mSv y⁻¹ and 0.09 mSv y⁻¹ to 0.17 mSv y⁻¹ respectively. Computed values of $H_{in}$ vary from 0.64 to 1.14 with an average value of 1.01. External hazard index, $H_{ex}$ for the soil samples studied in this work range from 0.48 to 0.84 with a mean value of 0.76. since most of the values are less than unity (shown in Fig 5), the fly ash may be safe to be used as a construction material without posing any radiological implications. But care must be taken in using the fly ash as building material.

5. CONCLUSIONS

The radium equivalent activity in mosaic samples is less than 370 Bq Kg⁻¹, which are acceptable for safe use [26]. since most of the values of $H_{ex}$ are less than unity (shown in Fig 5), the fly ash may be safe to be used as a construction material without posing any radiological implications. But care must be taken in using the fly ash as building material. Health risks are especially high in the area downward of the power plant. Being very minute, fly ash particles may tend to remain airborne for long periods leading to serious health problems as the airborne ash can enter the lungs through inhalation and may stick to lung tissues. Due to higher radon emanating power the lung tissues may be irradiated with $\alpha$- particles from radon progeny to a high degree, increasing the possibility of
lung cancer. These radionuclides in the fly ash may migrate from the waste disposal site to the underlying ground water body and may also accumulate in the top soil giving sufficient chances for the radionuclides to become enriched in soil.

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REFERENCES


