



RESEARCH ARTICLE

STUDY OF RADON EXHALATION RATE AND NATURAL RADIOACTIVITY IN SOIL SAMPLES
COLLECTED FROM 50M AWAY FROM KASIMPUR THERMAL POWER PLANT

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ABSTRACT

In the present study, about 20 samples of soil were collected about 50 m away from Kasimpur Thermal Power Plant to investigate the effect of fly ash spread out from the plant on the nearby soil. Can technique using LR-115 type II solid state nuclear track detector has been employed for the measurement of radon activity and radon exhalation rate. Radon activity for soil samples from inside of the Kasimpur Thermal Power Plant, varies from 4657.14 to 4714.29 Bq m⁻³ with an average value of 4685.72 Bq m⁻³, exhalation rate varies from 1674.35 to 1694.90 mBq m⁻² h⁻¹ with an average value of 1684.63 mBq m⁻² h⁻¹ and effective dose equivalent varies from 197.44 to 199.86 μSv y⁻¹ with an average value of 198.65 μSv y⁻¹, while from a distance of 50m away from power plant, radon activity varies from 2148.57 Bq m⁻³ to 4905.71 Bq m⁻³ with an average value of 3840.75 Bq m⁻³, exhalation rate varies from 772.46 mBq m⁻² h⁻¹ to 1763.72 mBq m⁻² h⁻¹ with an average value of 1380.84 mBq m⁻² h⁻¹ while effective dose equivalent varies from 91.09 μSv y⁻¹ to 207.98 μSv y⁻¹ with an average value of 162.83 μSv y⁻¹. Natural radioactivity in soil samples collected from a distance of 50m from Kasimpur Thermal Power Plant has been measured by low level gamma ray spectrometer. Activity concentrations were found to vary from 24.82 ± 0.68 to 241.55 ± 3.39 Bq kg⁻¹ with an average value of 72.19 ± 1.44 Bq kg⁻¹ for ²³⁸U, from 37.97 ± 1.3 to 97.22 ± 2.16 Bq kg⁻¹ with an average value of 68.81 ± 1.73 Bq kg⁻¹ for ²³²Th and 99.73 ± 1.53 to 710.72 ± 8.56 Bq kg⁻¹ with an average value of 517.89 ± 6.54 Bq kg⁻¹ for ⁴⁰K. Radium equivalent activity (Ra_{eq}) due to the presence of radio nuclides varies from 86.09 to 415.68 Bq kg⁻¹ with an average value of 206.84 Bq kg⁻¹. Total absorbed gamma dose rates in the surrounding air are found to vary from 38.56 to 191.23 nGy h⁻¹ with an average value of 96.51 nGy h⁻¹. Indoor and outdoor annual effective dose rate from these soil samples changes from 0.19 to 0.94 mSv y⁻¹ and 0.05 to 0.23 mSv y⁻¹ respectively. External hazard index, H_{ex} for the soil samples studied in this work range from 0.23 to 1.13 with mean a value of 0.57. Internal hazard index H_{in} varies from 0.3 to 1.79 with an average value of 0.76.

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INTRODUCTION

Noble radon gas (²²²Rn) originates from radioactive transformation of ²²⁶Ra in the ²³⁸U decay chain in the earth's crust (Vaupotic, 2010). The rate at which radon escapes or emanates from solid into the surrounding air is known as radon emanation rate or radon exhalation rate of the solid (Singh, 2010). The assessment of radiological risk related to inhalation of radon and radon progeny is based mainly on the integrated measurement of radon in both indoor and outdoor environments. The exhalation of radon from the earth crust and building materials forms the main source of radon in indoor environment (Gusain, 2009).

Plastic track detectors were used to measure the radon concentration and exhalation rate from soil samples (Mujahid *et al.*, 2010). In India there are a quite a few monazite sand-bearing placer deposits causing high background radiation along its long coastline. Ullal in Karnataka (Radhakrishna *et al.*, 1993) Kalpakkam (Kannan *et al.*, 2002) in Tamilnadu and Karala state and the Southwestern coast of India are known high background radiation areas (Mishra *et al.*, 1993; Suntan *et al.*, 1993; Zhu *et al.*, 1990 and Sohrabi, 1993). Some of these areas have been under study for many years in order to determine the risks and effects of long term, low level and natural radiation exposure (Sohrabi *et al.*, 1999). Man is dependent on soils and good soil are dependent upon man and the use he makes of them. Soil is a mixture of natural bodies on the earth surface containing living matter and supporting plants. Soil consists of three- phase system as solids, liquids and gases (Boal, 1976 and Rusal, 1957). Nationwide surveys

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have been carried out to determine the radium equivalent activity of soil samples in many countries (Singh *et al.*, 2003; Al-Jundi *et al.*, 2003; Mireles *et al.*, 2003; Ibrahim *et al.*, 1999; Sroor *et al.*, 2001 and Ibrahim *et al.*, 1993). Radioactivity is a part of the natural environment (Malik, *et al.*, 2011). Environment contains some naturally occurring radioactive materials (NORM) which are found in soils, rocks, vegetation, air, water and also in building materials (Mehra *et al.*, 2009). Naturally occurring radioactive materials (NORM) generally contains radionuclides found in nature i.e. thorium, uranium, and their progeny. Existences of three primordial radio nuclides (^{40}K , ^{238}U and ^{232}Th) in building materials cause internal and external exposures to residents. External exposure is caused by gamma radiation emitted from ^{40}K and daughter products of ^{238}U and ^{232}Th (Nassiri *et al.*, 2011). 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 (Kumar *et al.*, 2000). 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 (Forlkerts, 1984). In the present paper radon exhalation rates have been measured in soil samples, collected about 50 m away from Kasimpur Thermal Power Plant to investigate the effect of fly ash spread out from the plant on the nearby soil. The analysis of radioactivity in soil samples has been measured by low level gamma ray spectrometer. In addition, absorbed radiation doses and radiation risk have also been estimated.

Experiment

Radon exhalation rate

Radon exhalation rate is of prime importance for the estimation of radiation risk from various materials. In such measurements, it is expected that the exhalation rate depends upon the material and its amount as well as on the geometry and dimension of the can. Collected granite samples were dried and sieved through a 100- mesh sieve. They were placed in the cans (7.5cm height and 7.0 cm diameter) similar to those used in previous calibration experiment (Kumar, 2001). In each can a LR-115 type II plastic detector (2cm \times 2cm) was fixed at the top inside of the can, such that the sensitive surface of the detector faces the material and is freely exposed to the emergent radon. Radon decays in the volume of the can record the alpha particles resulting from the Po^{218} and Po^{214} deposited on the inner wall of the can. Radon and its daughters will reach an equilibrium in concentration after one week or more. Hence the equilibrium activity of the emergent radon can be obtained from the geometry of the can and the time of exposure. The detectors were exposed to radon for 100 days. After the exposure the detectors were etched in 2.5 N NaOH at 60°C in a constant temperature water bath for revelation of tracks. The resulting alpha tracks on the exposed face of the track detector were counted using an optical microscope at a magnification of 400X. The radon exposure inside the can was obtained from the track density of the detector by using calibration factor of 0.56 tracks $\text{cm}^{-2} \text{d}^{-1}$ obtained from an earlier calibration experiment (Singh, 1997). The exhalation

rate is found from the expression (Fleischer, 1978; Khan *et al.*, 1992).

$$Ex = \frac{CV\lambda}{A[T + \frac{1}{\lambda}(e^{\lambda T} - 1)]}$$

Where,

Ex = Radon Exhalation rate ($\text{Bq m}^{-2} \text{h}^{-1}$)

C=Integrated radon exposure as measured by LR-115 type II solid state nuclear track detector ($\text{Bq m}^{-3} \text{h}^{-1}$).

V=Volume of can (m^3)

λ =Decay constant for radon (h^{-1})

T=Exposure time (h)

A=Area covered by the can (m^2)

Estimation of ^{238}U , ^{232}Th and ^{40}K

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 (Kumar *et al.*, 2000). 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 (Forlkerts, 1984). Weighted averages of several decay products were used to estimate the activity concentrations of ^{238}U and ^{232}Th . The gamma ray spectrum 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:

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

Where S is the net counts/sec (cps) under the photo peak of interest, σ 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:

$$\text{Activity(Bq)} = \text{CPS} \times 100 \times 100 / \text{B.I} \times \text{Eff} \pm \text{CPS}_{\text{error}} \times 100 \times 100 / \text{B.I} \times \text{Eff} \quad (2)$$

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

RESULTS AND DISCUSSION

Table 1 Presents the measured data for the soil samples collected from the places 50m away from the power plant...

It is apparent from Table-1 that, radon activity for soil samples from inside of the Kasimpur Thermal Power Plant, varies from 4657.14 to 4714.29 Bq m⁻³ with an average value of 4685.72 Bq m⁻³, exhalation rate varies from 1674.35 to 1694.90 mBq m⁻² h⁻¹ with an average value of 1684.63 mBq m⁻² h⁻¹ and effective dose equivalent varies from 197.44 to 199.86 $\mu\text{Sv y}^{-1}$ with an average value of 198.65 $\mu\text{Sv y}^{-1}$, while from a distance of 50m away from power plant, radon activity varies from 2148.57 Bq m⁻³ to 4905.71 Bq m⁻³ with an average value of 3840.75 Bq m⁻³, exhalation rate varies from 772.46 mBq m⁻²h⁻¹ to 1763.72 mBq m⁻² h⁻¹ with an average value of 1380.84 mBq m⁻² h⁻¹ while effective dose equivalent varies from 91.09 $\mu\text{Sv y}^{-1}$ to 207.98 $\mu\text{Sv y}^{-1}$ with an average value of 162.83 $\mu\text{Sv y}^{-1}$. The fly ash spread over the soil in nearby places of the plant lowers the values of radon activity and radon exhalation. The gamma ray spectrum of a typical soil sample is shown in Fig 2. The measured activity concentration of ²³⁸U, ²³²Th and ⁴⁰K together with their average values in soil samples collected from the distance of 50m from Kasimpur Thermal Power Plant (U.P) are given in Table 2. 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 3.

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 (Yu *et al.*, 1992 ; Hayambu *et al.*, 1995) :

$$Ra_{eq} = C_U + 1.43 C_{Th} + 0.07C_K \quad (3)$$

Table 1. Radon activity concentration, radon exhalation rate and indoor inhalation exposure (radon)-effective dose from soil samples 50 m away from Kasimpur thermal power plant, India

	No. of samples	Track Density (track/cm ² d)	Radon Activity (Bq m ⁻³)	Exhalation rate (mBq m ⁻² h ⁻¹)	Effective dose equivalent ($\mu\text{Sv y}^{-1}$)
Inside Power Plant					
Minimum	20	260.8	4657.14	1674.35	197.44
Maximum	20	264.0	4714.29	1694.90	199.86
Average value	20	262.4	4685.72	1684.63	198.65
S.D	20	2.26	40.41	14.53	1.71
R.S.D%	20	0.86	0.86	0.86	0.86
50m away from power plant					
Minimum	20	120.32	2148.57	772.46	91.09
Maximum	20	274.72	4905.71	1763.72	207.98
Average value	20	215.07	3840.75	1380.84	162.83
S.D	20	44.19	789.41	283.81	33.47
R.S.D%	20	20.55	20.55	20.55	20.56

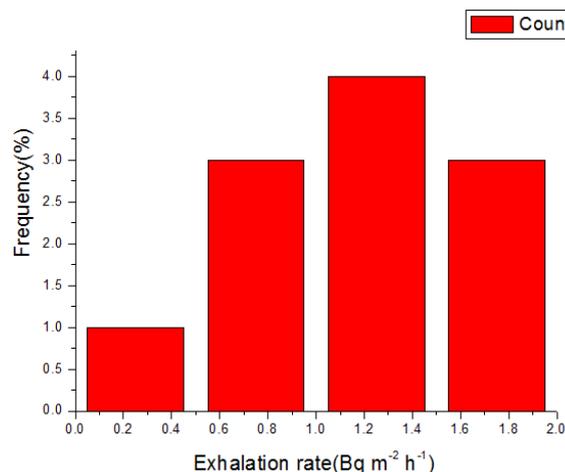


Fig 1. Presents the frequency distribution chart of exhalation rate of different samples studied

Where C_U , C_{Th} and C_K are the activity concentrations of ^{238}U , ^{232}Th and ^{40}K 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 ^{238}U or 259 $Bq\ kg^{-1}$ of ^{232}Th or 4810 $Bq\ kg^{-1}$ of ^{40}K . 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.

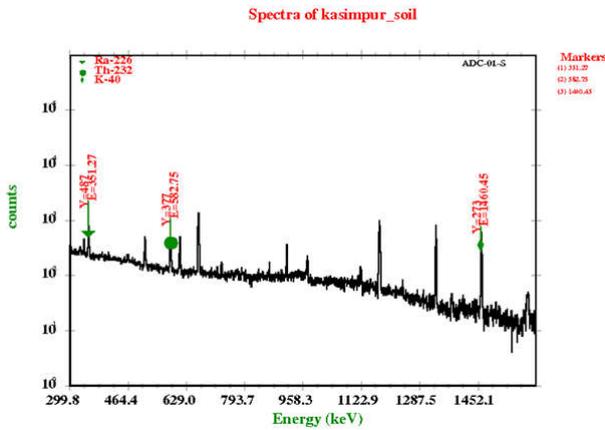


Fig 2. Gamma ray spectrum of a soil sample

Table 2. Activity concentration of ^{238}U , ^{232}Th , and ^{40}K in soil samples from a distance of 50 m from Kasimpur thermal power plant

Sample Code	^{238}U ($Bq\ kg^{-1}$)	^{232}Th ($Bq\ kg^{-1}$)	^{40}K ($Bq\ kg^{-1}$)
S1	58.19±1.37	62.83±1.60	468.48±6.09
S2	53.82±83	75.89±1.82	604.02±7.51
S3	71.24±1.61	79.56±1.92	501.49±6.46
S4	241.55±3.39	97.22±2.16	501.49±6.45
S5	63.44±1.44	73.28 ± 1.80	707.31 ±8.51
S6	79.85±1.64	92.66±2.13	545.90±6.92
S7	24.82±68	37.97±1.30	99.73±1.53
S8	67.27 ±1.51	69.08 ±1.73	536.49 ± 6.82
S9	43.32±1.08	63.09 ± 1.65	501.45 ±6.44
S10	43.31± 1.13	50.61± 1.39	519.71 ± 6.64
S11	47.32 ± 1.21	54.67 ±1.49	710.72 ± 8.56
Min	24.82±68	37.97±1.3	99.73±1.53
Max	241.55±3.39	97.22 ± 2.16	710.72 ±8.56
Average Value	72.19±1.44	68.81±1.73	517.89±6.54
S.D	58.23±.71	17.64±.28	161.16±1.86

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 ^{238}U , ^{232}Th and ^{40}K in $Bq\ kg^{-1}$ respectively by Monte Carlo method (UNSCEAR, 2000)

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

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 UNSCEAR (2000) were used. The indoor effective dose rate in units of $mSv\ y^{-1}$ was calculated by the following relation:

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

The outdoor effective dose rate in units of $mSv\ y^{-1}$ was calculated by the following relation:

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

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} :

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

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 (34) which gives

$$H_{ex} = \frac{C_U}{740\ Bq\ kg^{-1}} + \frac{C_{Th}}{520\ Bq\ kg^{-1}} + \frac{C_K}{9620\ Bq\ kg^{-1}} \leq 1 \quad (8)$$

Internal exposure to ^{222}Rn and its radioactive progeny is controlled by the internal hazard index (H_{in}) as given below (Cottens, 1990).

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

Effective Dose Equivalent (E_p)

The risk of lung cancer from domestic exposure of ^{222}Rn and its daughters can be estimated directly from the indoor inhalation exposure (radon) effective dose. The contribution of indoor radon concentration from the samples can be calculated from the expression (Nazaroff *et al.*, 1988):

$$C_{Rn} = \frac{E_x \times S}{V \times \lambda_v}$$

Where C_{Rn} , E_x , S, V, and λ_v are radon concentration ($Bq\ m^{-3}$), radon exhalation rate ($Bq\ m^{-2}\ h^{-1}$), radon exhalation area (m^2), room volume (m^3) and air exchange rate (h^{-1}) respectively. In these calculation, the maximum radon concentration from the building material was assessed by assuming the room as a cavity with $S/V = 2.0\ m^{-1}$ and air exchange rate of $0.5\ h^{-1}$. The annual exposure to potential alpha energy E_p (effective dose equivalent) is then related to the average radon concentration C_{Rn} by the following expression:

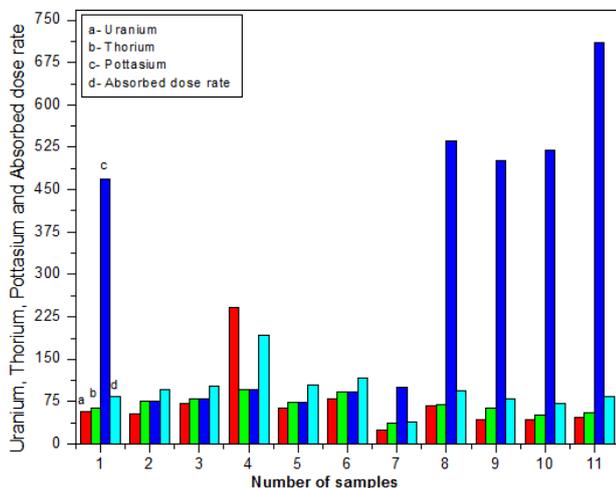
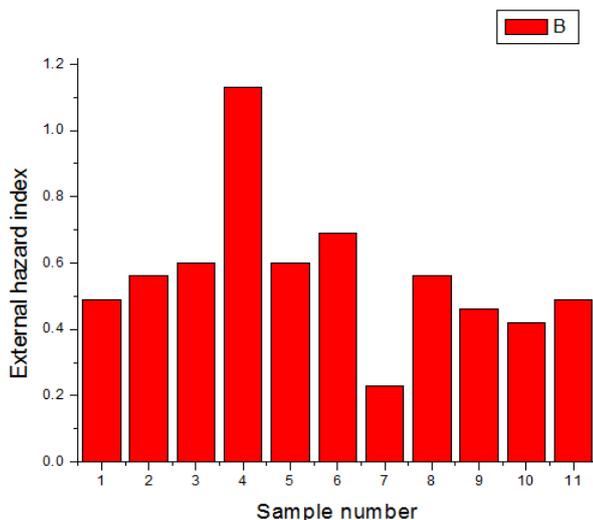
$$E_p (WLM\ yr^{-1}) = 8760 \times n \times f \times C_{Rn} / 170 \times 3700$$

Where C_{Rn} is in $Bq\ m^{-3}$; n, the fraction of time spent indoors; 8760, the number of hours per year; 170, the number of hours per working month and F is the equilibrium factor for radon. Radon progeny equilibrium factor is the most important quantity when dose calculations are to be made on the basis of the measurement of radon concentration.

Table 3. Radium equivalent activity, absorbed gamma dose rate, annual effective doses, External hazard index and internal hazard index in soil samples

Samples Code	Radium equivalent activity Ra_{eq} (Bq kg^{-1})	Absorbed gamma dose rate D(nGyh $^{-1}$)	Annual effective dose (mSv y $^{-1}$)		External Hazard Index (H_{ex})	Internal Hazard Index (H_{in})
			Indoor	Outdoor		
S1	180.83	84.37	0.41	0.10	0.49	0.65
S2	204.62	95.89	0.47	0.12	0.56	0.71
S3	220.12	101.88	0.49	0.12	0.60	0.79
S4	415.68	191.23	0.94	0.23	1.13	1.79
S5	217.74	103.07	0.51	0.13	0.60	0.78
S6	250.57	115.62	0.57	0.14	0.69	0.99
S7	86.09	38.56	0.19	0.50	0.23	0.30
S8	203.61	95.17	0.47	0.12	0.56	0.74
S9	168.64	79.03	0.39	0.09	0.46	0.58
S10	152.06	72.25	0.35	0.09	0.42	0.54
S11	175.25	84.52	0.41	0.10	0.49	0.61
Average value	206.84	96.51	0.47	0.12	0.57	0.76
S.D	81.58	37.36	0.18	0.04	0.22	0.38
R.S.D%	39.44	38.71	38.29	33.33	38.59	50

Equilibrium factor F quantifies the state of equilibrium between radon and its daughters and may have values $0 < F < 1$.

**Fig 3. Bar diagram showing activity concentration of ^{238}U , ^{232}Th , and ^{40}K and absorbed gamma dose rate in different samples****Fig 4. Bar diagram showing the values of external hazard index at locations**

The value of F is taken as 0.4 as suggested by UNSCEAR (1988). Thus the values of $n = 0.8$ and $F = 0.4$ were used to calculate E_p . From radon exposure, effective dose equivalents were estimated by using a conversion factor of 6.3 mSv WLM $^{-1}$ (ICRP, 1987). Fig. 3 shows the frequency plot of the variation of ^{238}U , ^{232}Th , ^{40}K and absorbed dose rate in these soil samples. Table 2 it can be observed that radium equivalent activity (Ra_{eq}) due to the presence of radio nuclides varies from 86.09 to 415.68 Bq kg^{-1} with an average value of 206.84 Bq kg^{-1} . Total absorbed gamma dose rates in the surrounding air are found to vary from 38.56 to 191.23 nGy h^{-1} with an average value of 96.51 nGy h^{-1} . Indoor and outdoor annual effective dose rate from these soil samples changes from 0.19 to 0.94 mSv y^{-1} and 0.05 to 0.23 mSv y^{-1} respectively.

External hazard index, H_{ex} for the soil samples studied in this work range from 0.23 to 1.13 with mean a value of 0.57. Internal hazard index H_{in} varies from 0.3 to 1.79 with an average value of 0.76. Since most of these values of H_{ex} are less than unity except one soil samples S-4 showing higher value than unity (shown in Fig 4), the use of soil from this region can be used as construction material without posing significant radiological threat to population.

Conclusion

The fly ash spread over the soil in nearby places of the plant lowers the values of radon activity and radon exhalation. The radium equivalent activity in mosaic samples is less than 370 Bq kg^{-1} , which are acceptable for safe use (Organization for Economic Cooperation and Development (1979), Mahur *et al.*, 2010). Since most of these values of H_{ex} are less than unity except one soil samples S-4 showing higher value than unity (shown in Fig 4), therefore the use of soil from this region can be used as construction material without posing significant radiological threat to population.

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