



ORIGINAL ARTICLE

Estimation of Natural Radioactivity and Radiation Doses in Soil Samples Collected from Jharkhand State, India

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ABSTRACT

In the present study radon exhalation rate and the activity concentration of radionuclides in soil samples collected from different places in Jharkhand state have been studied and evaluated. Gamma ray spectrometric measurement were carried out by using a NaI(Tl) detector at Atomic Minerals Directorate for Exploration and Research-Department of Atomic Energy (AMD-DAE), New Delhi, India. Radon exhalation rate measurement is done using 'Can technique'. It was found that soil activity ranges from 28.71 to 97.05 Bq/Kg with an average 51.97 Bq/Kg for ^{238}U , 47.53 to 132.81 Bq/Kg with an average 85.34 Bq/Kg for ^{232}Th and 290.30 to 1363.82 Bq/Kg with an average 555.32 Bq/Kg for ^{40}K . Radon surface exhalation rate vary from 2.15 $\text{mBq m}^{-2}\text{h}^{-1}$ to 89.35 $\text{mBq m}^{-2}\text{h}^{-1}$ with an average 37.06 $\text{mBq m}^{-2}\text{h}^{-1}$. Radon mass exhalation rate vary from 0.08 $\text{mBq Kg}^{-1}\text{h}^{-1}$ to 3.44 $\text{mBq Kg}^{-1}\text{h}^{-1}$ with an average 1.42 $\text{mBq Kg}^{-1}\text{h}^{-1}$. The absorbed gamma doses (D) in air due to naturally occurring radionuclide's in the soil sample varied from 56.09 to 175.28 nGy h^{-1} with an average value of 100.16 nGy h^{-1} . The annual effective dose (D_e) varies from 0.069 to 0.214 mSv y^{-1} with average value 0.123 mSv y^{-1} . Measured value of radium equivalent activity (R_{eq}) of sample collected from the area range from 123.14 to 372.71 Bq/Kg with an average 216.77 Bq/Kg. The value of internal hazards index (H_{in}) and external hazards index (H_{ex}) varies from 0.41 to 1.21 and 0.33 to 1.00 respectively. Radiation hazard index (representative level index) I_{yr} varies from 0.73 to 1.97 Bq/Kg with an average value of 1.26 Bq/Kg.

Key words: Natural radioactivity, NaI(Tl) detector, Effective dose rate, Radium equivalent activity, Radiation hazard indices

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INTRODUCTION

Radioactive radionuclide's in soil are either remaining from the Earth's original crust, introduced by cosmic radiation, or absorbed from man-made releases. Humans have always been exposed throughout their period of existence to naturally occurring radionuclides present in variable amounts in our environment. To assess radiological health hazards, naturally occurring radionuclide's are being measured in soil, sand, marble, bricks etc throughout the world (Rahman, *et al.*, 2008). It is a fact that natural radioactivity is present everywhere in earth and the study of naturally occurring radionuclide's uranium, potassium and thorium present in the earth, and radiation level in the environment is important for assessing the effect of radiation exposure to humane being due to both terrestrial and extraterrestrial sources (Rao NS, *et al.*, 2009). According to UNSCEAR (1993), about 87% of the radiation dose received by mankind is due to natural radiation sources and the remaining is due to anthropogenic radiation. The external radiation exposure arises mainly due to cosmic rays and terrestrial

radionuclide's occurring at trace levels in all soil. There are some regions in the world that are known for high background radiation areas (HBRAs), where the local geological controls and geochemical; effects causes enhanced levels of terrestrial radiation (MacKenzie, 2000; UNSCEAR, 2000; Mohanty, *et al.*, 2004). Wide ranging radiation studies have been carried out in the HBRAs in Brazil, in China, in India, in Iran and in USA and Canada and in some other countries to estimate risks and effect of long-term radiation exposure. High radiation above the earth is mainly due to naturally occurring radionuclide's ^{238}U , ^{232}Th and ^{40}K . High altitudes areas are also affected by cosmic radiation. All over the world, many works are being carried out to map radioelements in soil and environmental gamma dose rate. Radon exhalation is one of the important parameter to find out the radiation risk. The objective of this study was radiological characterization of natural radioactivity in the soil, rock, sand and ash sample from various location of Jharkhand state, India. In present study, radon exhalation rates have been measured using 'Sealed Can technique' (Abu-Zarad, *et al.*, 1988) and a low level gamma ray spectrometry was used for the estimation of U, Th and K concentration in the sample collected from different place of Rajmahal of Santhal Pergana Region, Jharkhand.

EXPERIMENTAL PROCEDURE

SAMPLING AND SAMPLE PREPARATION:

Samples were collected from different area of Rajmahal of Santhal Pergana Region, Jharkhand. The samples are different in nature like soil, coal, stone, sand etc. After collection, 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 measurement samples were oven dried at 110°C for 24 h and the samples were then packed and sealed in an impermeable airtight PVC container to prevent the escape of radiogenic gases radon and thoron. About 100 gm sample of each material were used for measurement. Before measurement, the containers were kept sealed about four weeks so that equilibrium of the ^{238}U and their daughter products is reached. After attainment of secular equilibrium between ^{238}U , ^{232}Th and their decay products, the samples were subjected to low level gamma spectroscopic analysis.

RADON ACTIVITY AND RADON EXHALATION RATE:

Radon exhalation is one of the important parameter to find out radiation risk from various building materials. The well known 'can-technique' was used using LR-115 type II plastic detector, as a solid state nuclear track detector was used for measurement of radon exhalation rate in which sample of measurement is enclosed in a sealed cylindrical can made of high grade plastic 7.5 cm height, 7.0 cm diameter and 0.5 mm thickness. The sensitive lower surface of Cellulose nitrate LR-115 (12 micrometer thickness) detector is freely exposed to register the alpha particles as shown in fig. 1. Nuclear tracks detected by the detector are not directly visible but can be visible by enlarging the molecular size using chemical etching. Different types of samples (Coal, flyash, soil etc) were collected from different location of power plants in Delhi. Collected samples were crushed in to powder from and sieved through a small mesh size (150 micron) to takeoff large size grain. For measurement, 100 gram of each sample was placed in a plastic can of size 7×7.5 cm. Radon and its progeny reach in equilibrium in nearly 4h and thus equilibrium activity of emergent radon can be obtained from the geometry of Can and time of exposure. After 90 days exposure, detectors were etched in 2.5 NaOH at 60°C for duration of 90 minute in a constant temperature water bath to detect the track. Resulting alpha tracks were counted using spark counter. From the track density the radon activity was obtained using the calibration factor $0.056 \text{ Tr cm}^{-2} \text{ d}^{-1} (\text{Bq m}^{-3})^{-1}$ obtained from an earlier calibration experiment (Singh, *et al.*, 1997). The surface exhalation rate of radon is obtained from the following expression:

$$E_A = \frac{CV\lambda}{A\left[T + \frac{1}{\lambda}(e^{-\lambda T} - 1)\right]} \quad \dots(1)$$

For radon mass exhalation rate-

$$E_M = \frac{CV\lambda}{M\left[T + \frac{1}{\lambda}(e^{-\lambda T} - 1)\right]} \quad \dots(2)$$

Where, E_A is radon surface exhalation rate (Bq/m²/h); E_M is radon mass exhalation rate (Bq/Kg/h); C is a integrated radon exposure as measured by Lr-115 solid state nuclear track detectors (Bq/m³/h); V is the effective volume of the can; λ is the decay constant (hr⁻¹); T is the exposure time (hr); A is the area of can (m²) and M is the mass of the sample (Mahur, *et al.*,2008)

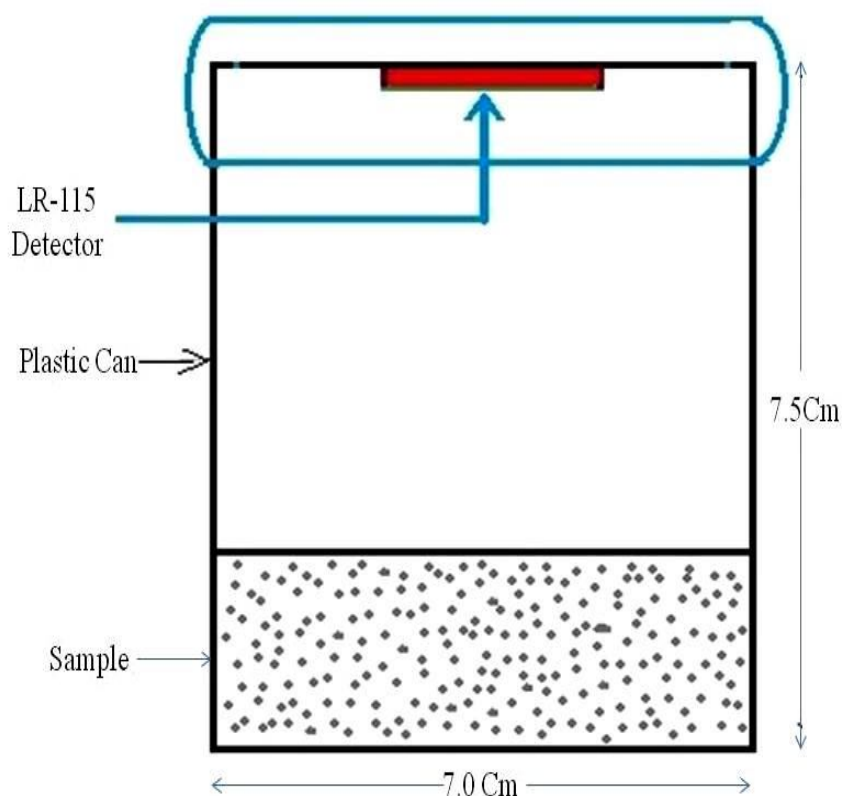


Fig. 1: Can technique setup for measurement of Radon exhalation rate

RADIOMETRIC ANALYSIS:

A low level gamma ray spectrometric measurement was carried out at Atomic Minerals Directorate for Exploration and Research-Department of Atomic Energy (AMD-DAE), New Delhi, India. The individual samples were placed on the detector [5"×4" Na(Tl)] manually and each sample was analyzed for a time of about 2000 sec. The detector has a resolution of 6-7% at 0.662 MeV for ¹³⁷Cs. The activity of ⁴⁰K was measured directly since ⁴⁰K is monoenergetic element emitting gamma photons of energy 1.46 MeV using energy regions 1.36-1.56 MeV. Since both U and Th have a long list of daughter products emitting so many gamma photons of different energies, we select the energy for which the interference from other element is low as well as the relative abundance is good. For ²³⁸U, gamma ray of its daughter ²¹⁴Bi (1.76MeV) and for ²³²Th the gamma ray of its daughter ²⁰⁸Tl (2.62MeV) using energy region 1.66-1.86MeV and 2.42-2.82 MeV respectively are selected for purpose of spectrometry, which are universally accepted. In the present

laboratory setup, the standard sample was prepared using rock powder material having 1100ppm of U_3O_8 and 4400ppm of ThO_2 (contain 176ppm of U_3O_8 also in it).

METHOD OF RADIOMETRIC ANALYSIS:

The relative method of estimation for ^{214}Bi , ^{208}Tl & ^{40}K are used for which the following equations are applicable.

$T_{hn} = T_{hg} - aU_n$, net rate of count in Th channel (T_{hn})

$U_n = U_g - \alpha T_{hn}$, net rate of count in U channel (U_n)

$K_n = K_g - \beta T_{hn}$, net rate of count in K channel (K_n)

T_{hg} , U_g and K_g are gross rate of counts (i.e., total rate of count – background rate of count) in respective channels and a , α , β and γ are stripping constants.

The final concentrations is given by

Conc. Th = T_{hn} / S_{Th} , Conc. U = U_n / S_U , Conc. K = K_n / S_K

Where S_{Th} , S_U and S_K are the respective channel sensitivities expressed as net rate of count per unit concentrations. As the concentrations found are in ppm, we obtain conc. of Th, U and K in Bq/Kg using the following relation

1 ppm of U = 12.36 Bq/Kg

1 ppm of Th = 4.04 Bq/Kg

1% of k = 302.4 Bq/Kg of ^{40}K .

RESULTS AND DISCUSSIONS

Table 1: Radon activity and radon exhalation rate in different soil samples

Sample code	Nature	Location	Track density (tr.cm ² d ⁻¹)	Radon activity (Bq m ⁻³)	Surface exhalation rates (mBqm ⁻² h ⁻¹)	Mass exhalation rates (mBq Kg ⁻¹ h ⁻¹)
R1	Soil	Rajmahal	39.08	662.49	14.12	0.54
R2	Soil	Mangalahate	85.91	1456.14	31.05	1.19
R3	Soil	Karanpurato	230.95	3914.44	83.47	3.21
R4	Soil	Sahibgang	116.46	1974.03	42.09	1.62
R5	Soil	Borio	247.22	4190.20	89.35	3.44
R6	Ash	Rajmahal Coal	15.67	265.67	5.66	0.21
R7	Soil	Barharwe	219.44	3719.39	79.31	3.05
R8	Sand	Mangla Hate	198.41	3362.92	71.71	2.76
R9	Rock	Dhamdhamia	38.88	659.13	14.05	0.54
R10	Sand	Jumani	27.18	460.72	9.82	0.37
R11	Soil	Barharwa	129.16	2189.26	46.68	1.79
R12	Soil	Barharwa	22.42	380.01	8.10	0.31
R13	Soil	Hiranpur	5.95	100.88	2.15	0.08
R14	Sand	Litipara	58.92	998.78	21.29	0.81
Minimum			5.95	100.88	2.15	0.08
Average			102.54	1738.14	37.06	1.42
Maximum			247.22	4190.20	89.35	3.44

Result of radon activity and radon exhalation from soil, ash, sand and rock samples are presented in different location of Jharkhand state is presented in table 1. Value of radon activity is vary from 100.88 Bq m⁻³ to 4190.20 Bq m⁻³ with an average 1738.14 Bq m⁻³. Radon mass exhalation and radon surface exhalation results are presented in Table 1. The average activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K were 51.97, 85.34 and 555.32 Bq/Kg respectively. The absorbed gamma doses (D) in air due to naturally occurring radionuclide's in the soil sample varied from 56.09 to 175.28 nGyh⁻¹ with an average value of (100.16) nGyh⁻¹. Radium equivalent activity, Effective dose rate, absorbed gamma dose, Level Index, Internal hazard index and external hazard index were also calculated and presented in table 3.

SPECIFIC ACTIVITY:

The radioactivity concentration of naturally occurring radionuclide's ^{226}Ra , ^{232}Th and ^{40}K and ratio of $^{232}\text{Th}/^{238}\text{U}$ in Bq/Kg of samples with their type and location are presented in Table 2.

Table 2: Natural Radioactivity concentration of ^{226}Ra , ^{232}Th and ^{40}K (Bq/Kg) and ratio of $^{232}\text{Th}/^{238}\text{U}$ for soil, ash, sand and rock samples

Sample code	Location	^{226}Ra (Bq kg ⁻¹)	^{232}Th (Bq kg ⁻¹)	^{40}K (Bq kg ⁻¹)	$^{232}\text{Th}/^{238}\text{U}$
R1	Rajmahal	33.64	75.30	577.58	2.23
R2	Mangalahate	29.66	49.73	290.30	1.67
R3	Karanpurato	32.80	47.53	483.84	1.44
R4	Sahibgang	54.08	78.28	408.24	1.44
R5	Borio	57.75	52.97	347.76	0.91
R6	Rajmahal Coal	97.05	122.91	350.78	1.26
R7	Barharwe	81.33	116.87	317.52	1.43
R8	Mangla Hate	77.76	132.81	1363.82	1.70
R9	Dhamdhamia	BDL	BDL	BDL	--
R10	Jumani	40.98	121.38	798.33	2.96
R11	Barharwa	BDL	BDL	BDL	--
R12	Barharwa	BDL	BDL	BDL	--
R13	Hiranpur	37.83	79.02	517.10	2.08
R14	Litipara	28.71	61.91	653.18	2.15
Maximum		97.05	132.81	1363.82	2.96
Average		51.97	85.34	555.32	1.63
Minimum		28.71	47.53	290.30	0.91

Table 3: Radium equivalent, dose rate, representative level index, internal hazards and external hazards index

Sample code	Radium equivalent activity Ra_{eq}	Effective dose rate D_e	Absorbed gamma dose rate D	Effective dose equivalent E_p	Level Index I_{yr}	Internal Hazard Index H_i	External Hazard Index (H_e)
R1	185.79	0.105	86.39	0.25	1.03	0.59	0.50
R2	123.14	0.069	56.69	0.55	0.73	0.41	0.33
R3	138.04	0.079	64.85	1.48	0.73	0.46	0.37
R4	197.46	0.111	90.62	0.75	1.20	0.67	0.53
R5	160.27	0.090	74.07	1.59	0.96	0.58	0.43
R6	299.82	0.166	135.79	0.10	1.96	1.07	0.80
R7	272.91	0.151	123.39	1.41	1.79	0.95	0.73
R8	372.71	0.214	175.28	1.27	1.96	1.21	1.00
R9	--	--	--	0.25	--	--	--
R10	276.03	0.156	127.60	0.17	1.57	0.85	0.74
R11	--	--	--	0.83	--	--	--
R12	--	--	--	0.14	--	--	--
R13	190.66	0.108	88.12	0.03	1.10	0.61	0.51
R14	167.55	0.096	78.95	0.37	0.86	0.53	0.45
Maximum	372.21	0.214	175.28	0.03	1.96	1.21	1.00
Average	216.77	0.123	100.16	0.66	1.26	0.73	0.59
Minimum	123.14	0.069	56.09	0.73	0.73	0.41	0.33

RADIUM EQUIVALENT ACTIVITY:

To compare the specific activity of the samples containing ^{226}Ra , ^{232}Th and ^{40}K , the radium equivalent index ($^{226}\text{Ra}_{\text{eq}}$) has been calculated to obtain the total amount of activities using the following equation-

$$\text{Ra}_{\text{eq}} = 1.43\text{C}_{\text{Th}} + \text{C}_{\text{Ra}} + 0.077\text{C}_{\text{K}} \quad \dots(3)$$

Where C_{Th} , C_{Ra} and C_{K} are the activities concentration in (Bq/Kg) of ^{226}Ra (U series), ^{232}Th and ^{40}K respectively (Kant, *et al.*, 2010)

ABSORBED GAMMA DOSE RATE AND THE EFFECTIVE DOSE RATE:

The measured absorbed gamma dose rate in air at 1 m above ground level due to activity concentration of ^{232}Th and ^{238}U series and ^{40}K are calculated using the formula ((UNSCEAR 1988)-

$$D_R = K_K C_K + K_{Th} C_{Th} + K_{Ra} C_{Ra} \quad \dots(4)$$

Where D_R is the absorbed dose rate (nGy hr^{-1}), and K_K , K_{Th} and K_{Ra} are the conversion factors (or dose rate coefficient) for potassium (0.043), Thorium (0.662) and Radium (0.427) respectively measured in nGy hr^{-1} per Bq/Kg^{-1} . To estimate the annual effective dose rates, the conversion coefficient from absorbed dose in air to effective dose rates (0.7 Sv Gy^{-1}) and outdoor occupancy factor (0.2) proposed by UNSCEAR (2000) were used. Effective dose rate (mSv y^{-1}) = Absorbed dose rate in air-

$$(\text{nGy h}^{-1}) \times 8760 \text{ h} \times 0.2 \times 0.7 \text{ Sv Gy}^{-1} \times 10^{-6} \quad \dots(5)$$

The results of absorbed gamma dose rate and effective dose rate are given in Table (2).

EFFECTIVE DOSE EQUIVALENT (E_p):

A parameter for measure the risk of lung cancer due to radon and its progeny can be found out is effective dose equivalent (E_p). Contribution of indoor radon concentration can be measured from the expression ((Nazaroff and Nero, 1988)-

$$C_{Rn} = (E_x \times S) / (V \times \lambda_v) \quad \dots(6)$$

Where C_{Rn} -radon concentration (Bq m^{-3}), E_x -radon exhalation rate ($\text{mBq m}^{-2} \text{h}^{-1}$), V -room volume; λ_v -air exchange rate (h^{-1}). In calculation, the maximum radon concentration from the building material was assessed by assuming the room as a cavity with $S/V = 2.0 \text{ m}^{-1}$ and air exchange rate of 0.5 h^{-1} . Annual exposure to potential alpha energy (E_p) is related to average radon concentration-

$$C_{Rn} E_p (\text{WLM y}^{-1}) = (8760 \times n \times F \times C_{Rn}) / (170 \times 3700) \quad \dots(7)$$

Where n is the fraction of time spent indoors taken as 0.8; 8760, the number of hours per year; 170, the hours per working month and F is the equilibrium factor taken as 0.4 as suggested by UNSCEAR (2000).

REPRESENTATIVE LEVEL INDEX (I_{yr}):

In order to examine whether the samples meet these limits of dose criteria, Radiation hazard index (representative level index) I_{yr} associated with the natural radionuclides in specific investigated samples is given by NEA-OECD (1979) as-

$$I_{yr} (\text{Bq/Kg}) = (1/150) C_{Ra} + (1/100) C_{Th} + (1/1500) C_K \quad \dots(8)$$

EXTERNAL AND INTERNAL HAZARD INDEX:

To limit the annual external gamma-ray dose to 1.5 mSv y^{-1} (UNSCEAR 2000), the external hazard index is used. The external hazard index is obtained from $R_{a_{eq}}$ expression through the supposition that its allowed maximum value (equal to unity) corresponds to the upper limit of $R_{a_{eq}}$ (370 Bq/Kg) given by the following equation (Beretka & Mathew, 1985)

$$H_{ex} = C_{Ra} / 370 + C_{Th} / 259 + C_K / 4810 \leq \quad \dots(9)$$

The internal exposure to ^{222}Rn and its radioactive progeny is controlled by internal hazard index, (H_{in}) which is given by (Beretka & Mathew, 1985):

$$H_{in} = C_{Ra} / 185 + C_{Th} / 259 + C_K / 4810 \leq 1 \quad \dots(10)$$

For the safe use of a material in the construction of dwellings and to keep the radiation hazard to be insignificant, this index value must be less than unity.

The value of radium equivalent in Bq/Kg , representative level index, internal hazards and external hazards of soil, ash, sand and rock sample are listed in table 2. From the table it is

clear that $^{232}\text{Th}/^{238}\text{U}$ for all samples are less than Clark's value (3.5), (U- enrichment) (Buket Canbaz, 2010).

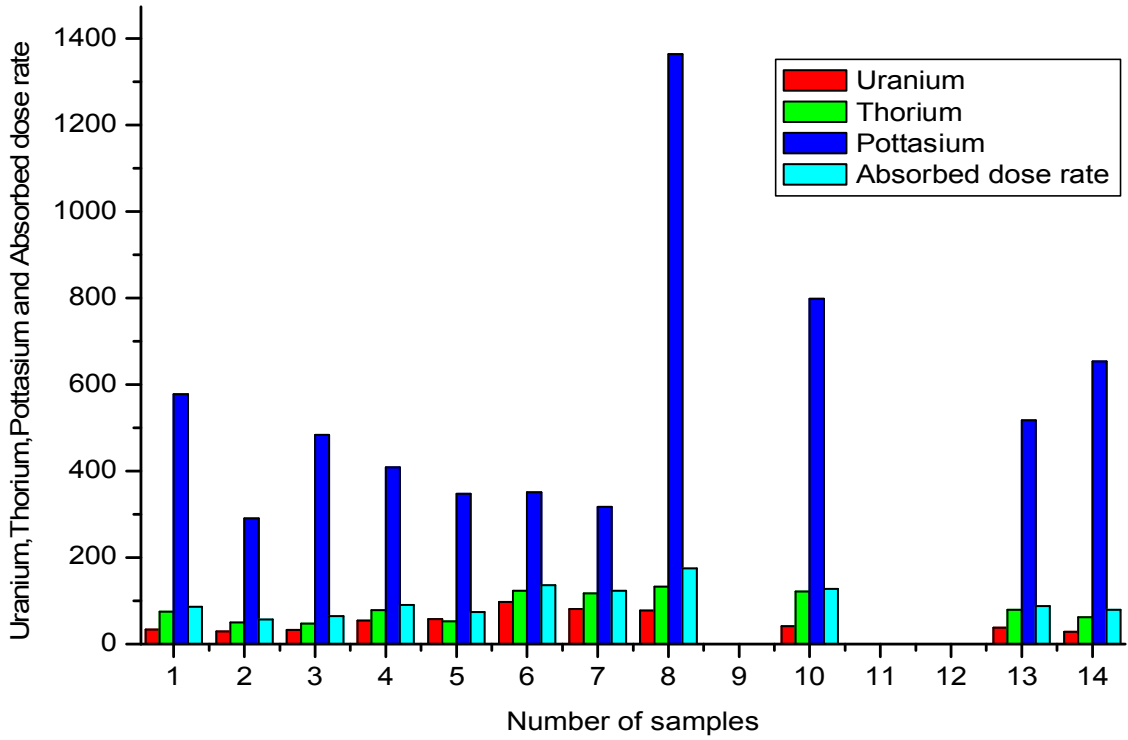


Fig. 2: Bar diagram showing the activity concentration of radionuclide's

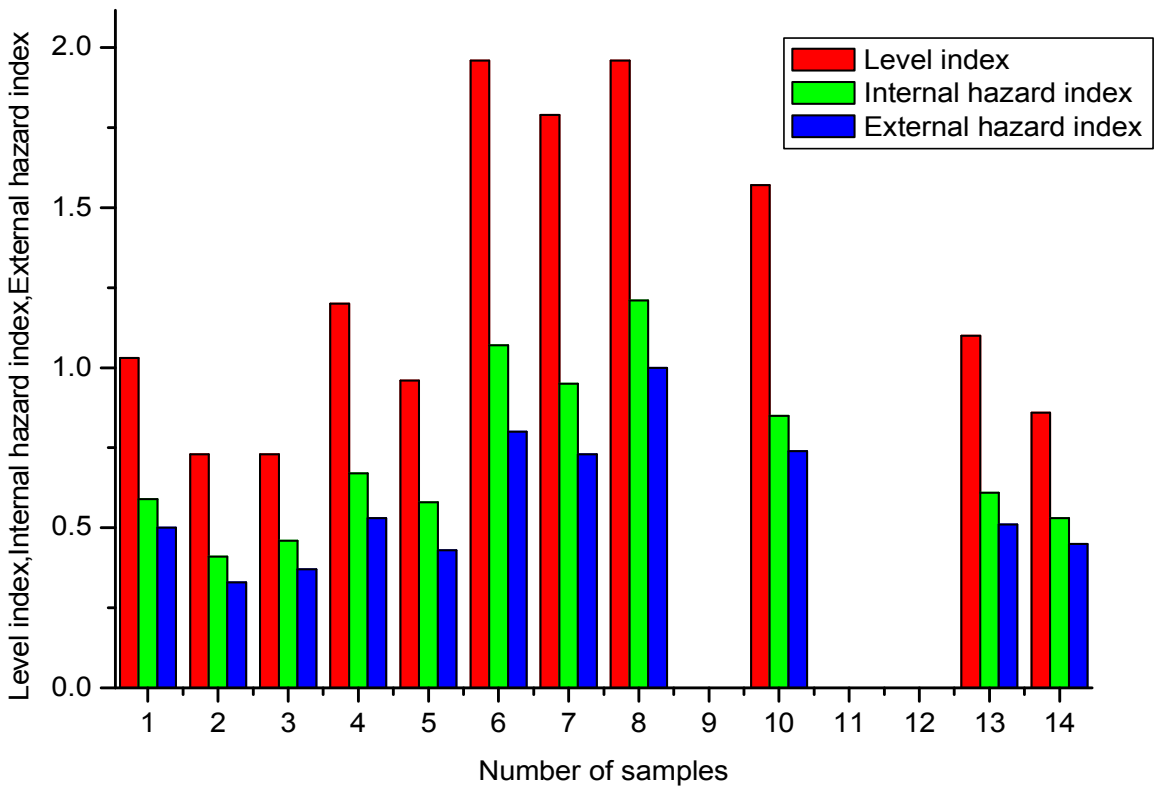


Fig. 3: Bar diagram showing the absorbed dose, Level index, internal hazard index and external hazard index

CONCLUSION

Gamma ray spectrometry has been used to determine the radioactivity concentration ^{226}Ra , ^{232}Th and ^{40}K in the studied sample collected from various location of Jharkhand state. The radiological parameters such as radium equivalent activity (Ra_{eq}) and activity index shows that internal dose due to natural radioactivity in the samples used not exceed the dose criteria. The ratio of $^{232}\text{Th}/^{238}\text{U}$ (Clark's value = 3.5) for all samples are less than Clark's value, (U enrichment). Also internal hazard index and external hazard index are less than the international average value. Fig. 2 and Fig. 3 shows the bar diagram showing the activity concentration of radionuclide's and Absorbed dose, Level index, internal hazard index and external hazard index. According to recommendation of International Commission on Radiation Protection (ICRP -65, 1993) remedial action is justified above a continuous effective dose of 10mSv and proposed the action level for radon concentration should be in range between 200 and 600 Bqm⁻³.

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