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Natural Radioactivity in Soil, Associated Radiation Exposure and Cancer Risk to Population of Eastern Haryana, India

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ABSTRACT

This study was undertaken to quantify the natural radioactivity in soil and associated radiation exposure to the inhabitants residing in Sonipat district, Haryana, India. A total of 120 soil samples were collected from 30 villages. These soil samples were quantified for naturally occurring radioactive materials activity using HPGe detector. Activity concentration of ^{40}K, ^{232}Th and ^{226}Ra in the soil samples of study area ranged from 463.8 – 696.9 Bq kg^{-1}, 31.4 – 37.9 Bq kg^{-1}and 41.5 – 54.9 Bq kg^{-1}, respectively. Radium equivalent (Ra_{eq}) values of all samples are lesser than maximum permissible limit (370 Bq kg^{-1}) acceptable for safer use of soil as building material. Average value of Air absorbed dose (AAD) in the study area was comparable to the Indian average of 64 nGy h^{-1}. Annual effective dose equivalent (AEDE) in the study area was 0.0831 μSv y^{-1} that is lower than the global average of 70.0 μSv y^{-1}. Annual Gonadal equivalent dose in study area was within the safe limit (1 μSv y^{-1}). Excess life time cancer risk due to natural radioactivity of soil to population of area is negligibly small. Values of Gamma index ($I_{\tilde{a}}$), outside hazard index (H_{ex}) and inside hazard index (H_{in}) are less than unity, indicating that there is no considerable health risk caused by natural radioactivity in soil in the study area.

INTRODUCTION

Nature is major source of radiation exposure through naturally occurring radionuclides and cosmic rays (UNSCEAR, 2000). So, human beings are exposed to these terrestrial radiations and get radiation dose from the ambient environment. Natural radioactivity exists in various matrices including rocks, soil, air, water etc. (Daulta et al. 2017).

Soil is the main constituent of earth crust on which human life dependent to great extent. Soil is a third major source of radiation exposure to human beings after air and water (Mathur et al. 2013). Radionuclides, belonging to radioactive decay series of ^{238}U, ^{235}U, ^{232}Th and ^{40}K, are present in almost all types of soils and are major cause of natural radioactivity (Rajamanna et al. 2013).The descendant products in these radionuclides series, which have comparatively shorter half-lives, add considerable level of radioactivity to average background levels. These natural sources are accountable for about 98% of the entire radiation dose to the people (UNSCEAR, 1993). The outdoor exposure of natural terrestrial radiations to human beings originates mainly from the upper 30 cm layer of the earth crust (Chikaswa et al. 2001). Naturally Occurring Radioactive materials (NORMs) are not uniformly distributed in earth crust. Concentration of radionuclides is highly variable in different type of soils that is affected by local geology, altitude, weather conditions, geochemistry and human economic & technological activities etc. of region (Radhakrishna et al. 1993).Therefore quantitative information about NORMs distribution, in different regions of the world is important to protect life from radiation exposure (Rani et al. 2005; Amanjeet et al. 2017).

Above a level, natural radioactivity may cause detrimental radiological health risks. The dose rate as well as the absorbed dose decides harshness and nature of the biological and clinical symptoms (Singh et al. 2014). Hereditary and somatic effects may occur due to excess of radiation exposure (Ajayi, 2008). Gamma radiation exposure and inhalation of gaseous radionuclides may pose cancer risk to different body parts (Srilatha et al. 2015). If information about the distribution of radioactive elements is available then several diseases and sicknesses can be efficiently managed. Therefore quantification of natural radionuclides in soil is of great importance with respect to radiation exposure.

The study on natural radioactivity in soil and associated risks in different regions has been reported in past. The literature revels that naturally occurring radionuclides have been quantified by the use of HPGe detector in Karnataka (Srilatha and Rangaswamy, 2015); Punjab (Badhan and Mehra, 2012); Haryana (Yadav et al. 2018); Uttar Pardesh (Mathur et al. 2013) and by the use of NaI(TI) detector in Tamil Nadu (Ravisanker et al. 2012); Himachal Pardesh (Bala et al. 2014); Beach sand samples of Kerala (Ramasamy, 2013); Haryana (Panghal et al., 2018) and many other regions of India. Similarly in Rize Province, Turkey (Durusoy and Yildirim, 2017); South-Eastern Nigeria (Osimobi et al. 2018); Xiamen Island, China (Huang et al. 2015); Nile Delta, Egypt (Yousuf et al. 2007); Poland (Saleziak et al. 2010) etc. regions of world have been quantified for natural radioactivity in soil by the use of NaI(TI) detector. A bibliographic survey indicated that no such data is available for Sonipat district of Haryana, India. Keeping this in view, the present study was undertaken to quantify natural radioactivity in the soil of Sonipat district and evaluation of radium equivalent activity, associated radiation hazards to the inhabitants of the study area.

MATERIAL AND METHOD

Study Area

Sonipat district is located at latitude N 28.980 and longitude E 77.020. The area of the district is around 2260 km^2. As per census 2011 the population of the district is 1.48 million. The Yamuna river runs along the eastern boundary of the district. Sonipat district has alluvium soil and loamy soil. Most of the area of district is plain. The climate of Sonipat is dry with a cold winter and a hot summer. The maximum (75% of the annual) rainfall is received in monsoon season. About 80% area of that area is under cultivation. The major crops cultivated in the district are wheat, rice, sugarcane and cotton (Sonipat District, 2018). The main source of irrigation water is from Yamuna river and groundwater. Study area map with sampling locations is given

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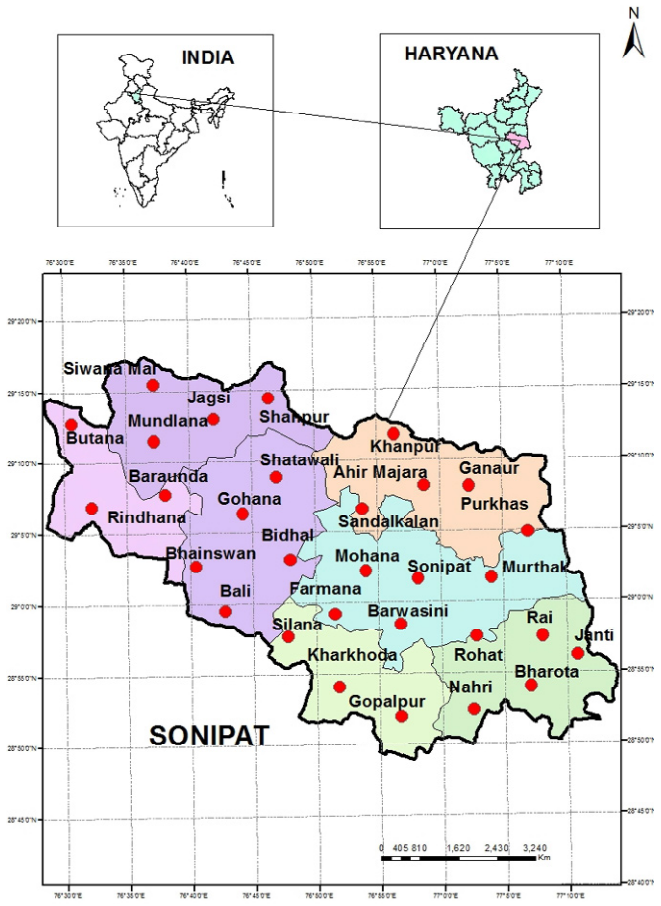

Fig.1. Sampling location map of Sonipat district, Haryana.

in Fig.1.

Sample Collection, Processing and Analysis

A total of 120 soil samples from 30 villages of the district were collected in Rabi and Kharif seasons. To decide sampling location Sonipat district area was divided into grids. Area of each grid was approximately 75 km^2. The soil samples were collected from rice and wheat fields. Rice field soil samples were collect in November, 2014 and November, 2015. Wheat field soil samples were collected in April, 2015 and April, 2016. Collected soil sample was firstly air dried and then oven dried at 110º C to remove moisture until a constant weight was attained. Then after removal of all extraneous matter (pebbles and stones) the soil samples were grounded and sieved through 0.5 mm sieve to homogenize the texture. Samples were filled into the plastic containers of 250 ml capacity having the same geometry and size. All samples were sealed by plastic tape, labeled and kept for one month to achieve secular equilibrium among uranium, thorium and their daughter products.

p-type High-purity Germanium (HPGe) detector (Baltic Scientific Instruments, Latvia) along with acquisition interface module ORION (ITECH Instruments, France) has been used to analyze the soil samples. Qualitative and quantitative analysis of gamma spectra was processed using user interface program INTERWINNER 7.0. The detector is enclosed by a heavy-lead shield (210Pb < 50Bq kg^{-1}) of about 100 mm thickness to minimize the background interference. A 9.0 mm thick layer of copper lined with 1.0 mm tin has been used to reduce characteristic X-rays from lead shield. The instrument has relative efficiency (w.r.t. 3" × 3" NaI detector and Co-60 source mounted 25 cm above the detector) at 1332 keV γ-photon is ≥50% and energy resolution (FWHM) of 0.90 KeV for energy 122 keV(Co- 57) and 2.0 KeV for energy 1332 keV(Co-60).

To ensure quality of measurement, the standard mixed multi-nuclide source, procured from BARC, Mumbai and ESL, Tarapur were used for energy and efficiency calibration of the instrument. Each samples and background were measured for radioactivity for a period of 80,000s. The activity concentration of ^{40}K was measured directly from its gamma emission at 1461 keV (abundance 10.7%). The activity of ^{238}U and ^{232}Th were calculated assuming a secular equilibrium with their daughter products. The activity concentration of ^{238}U was determined through gamma energy photo peak of its daughter ^{214}Bi (609 keV, abundance 45% and 1764 keV, abundance 16%), while the activity of ^{232}Th determined by photo peaks of ^{228}Ac (911.1 keV, abundance 27.8%) and ^{208}Tl (583keV, abundance 86%) (EML, 1990)

Calculations

1) NORMs, ^{40}K, ^{232}Th and ^{226}Ra activity

Different radionuclide's activity was calculated in the samples using the following equation (1) (Knoll, 1998).

$$Activity\ (Bq/kg) = [(g/t_1) - (b/t_2) \pm SD] \times 100/\eta \times 100/a \times 1000/wt \quad (1)$$

where g= Gross count (sample + background) in t_1 seconds, b= Background counts of counter in t_2 seconds, SD= Standard deviation = $[(G/t_1)^2 + (B/t_2)^2]^{1/2}$, η = Percent efficiency of HPGe system for a particular energy peak. a = Abundance factor of radionuclide for a particular energy peak, wt = dry weight of soil sample taken for analysis (in grams).

2) Radium Equivalent (Ra_{eq}) activity

Ra_{eq} is an index that describes gamma amount produced from different concentration of radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in the soil samples.Distribution of radionuclides in soil is not consistent. So, the consistency with respect to radiation exposure to human from ^{226}Ra, ^{232}Th and ^{40}K has been defined in terms of radium equivalent activity. It has been calculated by using the equation (2) (Beretka and Mathew, 1985).

$$Ra_{eq} = R_{Ra} + 1.43 \times R_{Th} + 0.077 \times R_K \quad (2)$$

Where Ra_{eq} (Bq kg^{-1}), R_{Ra}, R_{Th} and R_K are the activity (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, correspondingly.

3) Hazard Indices Calculation

1) Air Absorbed Dose rate (AAD)

Kocher and Sjoreen (1985) have assumed in their study while calculating air absorbed dose that environmental background radiation dose is a result of ^{238}U series, ^{232}Th series,^{40}K only. The contribution of other radionuclides can be neglected as they contribute very little. Same assumption used in the present study. The exterior air absorbed gamma radiation dose rates at about 1.0 m height to the ground were calculated using the conversion factor of 0.462 nGy h^{-1}/ Bq kg^{-1} for ^{226}Ra, and 0.604 nGy h^{-1}/Bq kg^{-1} for ^{232}Th and 0.0417 nGy h^{-1}/Bq kg^{-1} for ^{40}K and is calculated by equation (3) (UNSCEAR 2000).

$$AAD = 0.462 \times R_{Ra} + 0.604 \times R_{Th} \times 0.0417 \times R_K \quad (3)$$

Where AAD is gamma radiation dose in nGy h^{-1}; R_{Ra}, R_{Th} and R_K are the activity (Bq kg^{-1}) of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

2) Annual Effective Dose Equivalent (AEDE)

The outdoor annual effective dose equivalent due to natural radionuclides in the soil samples was quantified using the outdoor

occupancy of 0.2 and dose conversion coefficient for converting the absorbed dose rate in air to the effective dose (0.7 Sv Gy^{-1}) as projected by (UNSCEAR, 2000). The annual effective dose received by the population was calculated using the equation (4) (Jibiri and Adewuyi 2008).

$$AEDE\ (\mu Svyr^{-1}) = D \times T \times OF \times Q \times DCF \quad (4)$$

where, D = absorbed dose rate (nGy h^{-1}), T = time, OF = the occupancy factor which corrects the average time spent outdoor (0.2), Q = the quotient of the effective dose rate and absorbed dose rate in air (0.7 Sv Gy^{-1}). DCF = Dose conversion factor (10^{-6}).

3) Annual Gonadal Equivalent Dose (AGED)

Gonads are contemplated as the key organs of interest for dose evaluation from the gamma radiation (UNSCEAR 1982). The annual gonadal equivalent dose in the study area due to ^{226}Ra, ^{232}Th and ^{40}K was calculated using equation (5) (Arafa 2004).

$$AGED\ (\mu Svy^{-1}) = 3.09 \times R_{Ra} + 4.18 \times R_{Th} + 0.314 \times R_K \quad (5)$$

Where R_{Ra}, R_{Th} and R_K are the activities of ^{226}Ra, ^{232}Th and ^{40}K, in that order.

4) Gamma Index (I_{γ})

Gamma Index represents the total radiological hazard due to presence of ^{226}Ra, ^{232}Th and ^{40}K in the soil. This index is given by European Commission 1999. It was calculated using equation (6) (EC 1999).

$$I_{\gamma} = (R_{Ra}/300) + (R_{Th}/200) + (R_K/3000) \quad (6)$$

Where I_{γ} is gamma index and R_{Ra}, R_{Th} and R_K are the activity of ^{226}Ra, ^{232}Th and ^{40}K, in that order.

5) Outside Hazard Index (H_{ex})

Soil is main constituent of building materials that may pose risk of gamma radiation exposure to the inhabitants. Outside hazard index (H_{ex}) was calculated to quantify this radiation exposure using equation (7) (Beretka and Mathew, 1985).Calculation was done based on the estimation assumption that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K all producing the same gamma ray dose rate (Beretka and Mathew, 1985; Yu Guan Stoks and Young, 1992).

$$H_{ex} = (R_{Ra}/370) + (R_{Th}/259) + (R_K/4810) \quad (7)$$

Where H_{ex} is the outside hazard index and R_{Ra}, R_{Th} and R_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, correspondingly.

6) Inside Hazard Index (H_{in})

Radon causes damage to the lungs of human beings which is quantified by inside hazard index and is calculated as per equation (8) (Berekta and Mathew, 1985).The inside hazard index is clear to decrease the adequate upper limit concentration of ^{226}Ra to half the value suitable to outside exposure alone (Ravishanker et al. 2014).

$$H_{in} = (R_{Ra}/185) + (R_{Th}/259) + (R_K/4810) \quad (8)$$

Where H_{in} is inside hazard index and R_{Ra}, R_{Th} and R_K are the activity of ^{226}Ra, ^{232}Th and ^{40}K, correspondingly.The values of H_{ex} and H_{in} indices must be <1.0 $mSvy^{-1}$ in order not to cause any harmful effect to population (Quindos Fernandez and Soto, 1987).

7) Cancer Risk

Outdoor cancer risk was calculated using equation (9) (Ramasamy et al. 2013).

$$Cancer\ risk = AEDE \times DL \times RF \quad (9)$$

Where AEDE is total annual effective dose equivalent (in μSv y^{-1}); DL is duration of life (65 years) (Jain et al. 1995) and RF is risk factor (ICRP 60 uses values of 0.05 Sv^{-1} for the public). (ICRP 1991).

RESULT AND DISCUSSION

Natural Radioactivity in Soil due to ^{40}K, ^{232}Th and ^{226}Ra

The overall activity concentration (calculated by taking average of four seasons) of ^{40}K ^{232}Th and ^{226}Ra in study area ranged 463.8 – 696.9Bq kg^{-1} with mean 581.5 ± 80.9 Bq kg^{-1}, 31.4 – 37.9Bq kg^{-1} with mean 34.9 ± 2.9 Bq $kg^{-1}$and 41.5 – 54.9 Bq kg^{-1} with mean 48.7 ± 5.4 Bq kg^{-1} respectively (Table 1).

Activity concentration in soil during each harvesting season of Rabi and Kharif crop was measured for four seasons. Soil samples collected in Kharif in November - 2014 have activity concentration in the range of 493.6 – 682.2 Bq kg^{-1}, 29.7 – 39.1 Bq kg^{-1} and 36.3 – 56.4 Bq kg^{-1} of ^{40}K, ^{232}Th and ^{226}Ra respectively. The activity of ^{40}K, ^{232}Th and ^{226}Ra in Rabi field soil samples collected in May- 2015 ranged 335.7 - 765.7 Bq kg^{-1}, 31.0 - 40.4 Bq kg^{-1} and 39.4 - 60.4 Bq kg^{-1} correspondingly (Table 2). The activity of ^{40}K, ^{232}Th and ^{226}Ra in Kharif field soil samples collected in November-2015 ranged 443.4 – 737.4 Bq kg^{-1}, 27.9 – 39.6 Bq kg^{-1} and 35.8 – 54.7 Bq kg^{-1} correspondingly. For Rabi soil samples collected in May-2016 the activity ranged from 330.4 - 757.4 Bq kg^{-1}, 28.7 – 40.9 Bq kg^{-1} and 39.6 – 57.9 Bq kg^{-1} for ^{40}K,^{232}Th and ^{226}Ra correspondingly (Table 3). There is no significant seasonal variation in the activity of radionuclides in soil samples in different seasons at same location.

A comparison of radioactivity in the soil of Sonipat district was made from the available literature, it was found that radionuclide concentration is comparable with other parts of Haryana (Yadav et al. 2018): Shivalik range (Singh el al. 2009); have higher values of NORMs. Similar situation is with the other parts of the world, i.e. rock samples of Lombardia, Italy has higher concentration of natural radionuclides due to high density (Guidotti et al., 2015) and sand samples of Xiamen Island of China (Huang et al., 2015) have lower values due to lower density and lesser organic carbon contents while alluvial deposits have comparable values. Comparison of present study with other similar studies conducted worldwide is presented in Table 4.

Kurtosis analysis of the ^{40}K, ^{226}Ra and ^{232}Th activity distribution in soil samples showed values -0.47, -0.263 and -0.249 respectively. Negative values indicates that the distribution of all three radionuclides is relatively flat or platykurtic. Such a distribution may be result of local geological and mineralogical difference of the Sonipat district as Kadar, alluvial and sandy soil have different concentrations of essential and non-essential elements. Frequency distribution is graphically presented in Figs. 2,3 and 4 for ^{40}K, ^{226}Ra and ^{232}Th. Overall village wise distribution of activity of ^{40}K, ^{232}Th and ^{226}Ra is depicted in Fig 5. The worldwide activity concentrations of, ^{40}K, ^{232}Th and ^{226}Ra are in the range of 140 - 850 Bq kg^{-1}, 11 –64 Bq $kg^{-1}$and 17 – 60 Bq kg^{-1} with mean concentrations of 400, 30 and 35 Bq kg^{-1} respectively (UNSCEAR, 2000). In present study range of activity concentration is equivalent to the global range but average concentration is higher than global average. In the study area, fertilizers are used in excess quantity to increase crop production. Excess use of fertilizers during the agronomic practices may contribute to terrestrial background activity (Almayahi et al. 2012). ^{40}K contribution of activity is higher, it is probably due to the use of inorganic fertilizer as soil

Table 1. Average activity concentration, Radium Equivalent activity, Air absorb dose(AAD), Annual Effective dose equivalent(AEDE), Annual Gonadal Equivalent Dose(AGED), Gamma Index, Hazard Index and Cancer risk in soil of Sonipat district, Haryana

Sample	Location	Activity (Bq kg^{-1})				AAD (nGy h^{-1})	AEDE (μSv y^{-1})	AGED (μSv y^{-1})	Gamma Index	Hazard Index		CR× 10^{-3}
		^{40}K	^{232}Th	^{226}Ra	Ra_{eq}					Outside	Inside	
SRW-1	Shaupur	636.9	37.45	52.6	155.2	73.4	0.090	519.0	0.575	0.419	0.561	0.293
SRW-2	Mundlana	639.3	37.45	51.5	154.3	73.0	0.089	516.5	0.572	0.417	0.556	0.291
SRW-3	Gohana	619.1	37.87	52.7	154.6	73.0	0.089	515.6	0.571	0.417	0.560	0.291
SRW-4	Gannaur	538.3	31.43	43.9	130.3	61.7	0.075	436.1	0.483	0.352	0.471	0.246
SRW-5	Murthal	583.0	34.51	49.0	143.2	67.7	0.083	478.6	0.530	0.387	0.519	0.270
SRW-6	Sewali	571.3	34.29	46.6	139.6	66.0	0.080	466.6	0.517	0.377	0.503	0.263
SRW-7	Janti	623.4	36.13	50.9	150.5	71.3	0.087	503.9	0.558	0.407	0.544	0.284
SRW-8	Nahri	628.7	36.38	49.7	150.1	71.1	0.087	502.9	0.557	0.405	0.540	0.283
SRW-9	Gopalpur	624.7	34.86	48.2	146.2	69.3	0.085	490.8	0.543	0.395	0.525	0.276
SRW-10	Kharkhoda	615.6	36.56	53.1	152.7	72.2	0.088	510.0	0.565	0.413	0.556	0.288
SRW-11	Rohat	534.0	32.27	41.5	128.8	60.9	0.074	430.9	0.478	0.348	0.460	0.243
SRW-12	Barwasini	549.3	32.79	44.2	133.3	63.1	0.077	446.0	0.494	0.360	0.480	0.251
SRW-13	Sandalkalan	572.7	34.81	48.9	142.8	67.4	0.082	476.4	0.528	0.386	0.518	0.269
SRW-14	Purkhas	605.6	35.46	51.4	148.7	70.4	0.086	497.2	0.551	0.402	0.541	0.280
SRW-15	Ahir Majara	606.7	36.16	50.5	148.9	70.4	0.086	497.7	0.551	0.402	0.539	0.281
SRW-16	Jagsi	576.2	35.04	47.2	141.6	66.9	0.082	473.1	0.524	0.383	0.510	0.267
SRW-17	Siwana Mal	527.1	32.50	43.8	130.9	61.8	0.075	436.8	0.484	0.354	0.472	0.246
SRW-18	Butana	610.7	34.94	47.4	144.4	68.4	0.083	484.3	0.536	0.390	0.518	0.273
SRW-19	Barauda	533.6	33.42	43.9	132.7	62.7	0.076	442.7	0.491	0.358	0.477	0.250
SRW-20	Rindhana	616.8	34.04	47.7	143.9	68.3	0.083	483.3	0.535	0.389	0.517	0.272
SRW-21	Bhainswan	619.5	33.93	45.5	141.7	67.3	0.082	477.0	0.528	0.383	0.506	0.268
SRW-22	Bali	696.9	36.42	54.0	159.8	76.0	0.093	538.0	0.595	0.432	0.578	0.303
SRW-23	Farmanah	613.3	35.08	51.8	149.3	70.7	0.086	499.4	0.553	0.403	0.543	0.282
SRW-24	Silana	590.0	36.68	54.9	152.7	72.0	0.088	508.1	0.563	0.413	0.561	0.287
SRW-25	Mohana	601.1	36.71	51.2	150.0	70.8	0.086	500.4	0.555	0.405	0.543	0.282
SRW-26	Bidhal	559.5	35.69	51.3	145.4	68.6	0.084	483.5	0.536	0.393	0.532	0.273
SRW-27	Khanpur	522.8	35.73	50.0	141.3	66.4	0.081	467.9	0.519	0.382	0.517	0.265
SRW-28	Bharoth	497.8	33.46	48.4	134.6	63.3	0.077	445.8	0.495	0.364	0.494	0.252
SRW-29	Sonipat	468.5	32.02	42.8	124.7	58.6	0.071	413.2	0.459	0.337	0.452	0.234
SRW-30	Shatawali	463.8	32.80	45.2	127.8	60.0	0.073	422.3	0.469	0.345	0.467	0.239

Table 2. Naturally occurring radionuclides activity in soil samples collected in harvesting season of Kharif – 2014 and Rabi – 2015 crop field's

Sample	Activity Concentration (Bq Kg^{-1})					
	Kharif Field's Soil (November – 2014)			Rabi Field's Soil (May – 2015)		
	^{40}K	^{232}Th	^{226}Ra	^{40}K	^{232}Th	^{226}Ra
SRW-1	580.1 ± 15.2	33.1 ± 1.2	49.9 ± 1.7	679.3 ± 17.6	39.3 ± 1.4	53.0 ± 1.9
SRW-2	591.8 ± 15.7	35.2 ± 1.3	49.7 ± 1.8	678.3 ± 16.7	38.8 ± 1.3	54.2 ± 1.9
SRW-3	566.7 ± 15.0	36.3 ± 1.3	52.4 ± 1.8	649.2 ± 16.6	38.7 ± 1.3	53.4 ± 1.9
SRW-4	560.3 ± 15.4	32.1 ± 1.2	43.1 ± 1.7	544.3 ± 15.4	32.4 ± 1.2	47.1 ± 1.8
SRW-5	632.2 ± 16.5	34.2 ± 1.3	52.5 ± 1.9	526.3 ± 15.1	33.0 ± 1.2	45.6 ± 1.8
SRW-6	573.9 ± 15.5	33.7 ± 1.2	41.8 ± 1.6	643.9 ± 17.7	37.7 ± 1.4	52.6 ± 2.0
SRW-7	603.9 ± 16.4	38.2 ± 1.4	52.0 ± 1.9	702.9 ± 18.4	38.7 ± 1.4	56.4 ± 2.1
SRW-8	567.4 ± 14.9	33.5 ± 1.2	45.1 ± 1.6	598.2 ± 16.1	35.4 ± 1.3	48.1 ± 1.8
SRW-9	632.0 ± 16.7	35.1 ± 1.3	45.8 ± 1.8	632.5 ± 16.2	36.3 ± 1.3	51.1 ± 1.8
SRW-10	542.1 ± 14.8	38.7 ± 1.3	54.5 ± 1.8	706.5 ± 18.5	39.6 ± 1.4	60.4 ± 2.1
SRW-11	525.4 ± 14.9	30.4 ± 1.2	38.9 ± 1.6	490.7 ± 13.5	32.2 ± 1.1	39.9 ± 1.5
SRW-12	551.3 ± 14.8	31.9 ± 1.2	43.6 ± 1.6	497.1 ± 13.6	31.0 ± 1.1	41.3 ± 1.5
SRW-13	500.1 ± 14.3	35.1 ± 1.2	47.8 ± 1.7	614.0 ± 16.7	34.1 ± 1.3	47.8 ± 1.8
SRW-14	523.2 ± 14.9	34.0 ± 1.3	49.3 ± 0.7	735.1 ± 18.2	37.4 ± 1.4	56.5 ± 2.0
SRW-15	598.1 ± 16.1	37.6 ± 1.3	52.8 ± 1.9	652.2 ± 17.1	38.4 ± 1.4	54.7 ± 2.0
SRW-16	579.2 ± 15.6	39.1 ± 1.3	56.4 ± 1.9	584.7 ± 15.5	35.6 ± 1.3	45.7 ± 1.7
SRW-17	585.0 ± 15.2	33.1 ± 1.2	49.4 ± 1.7	506.3 ± 13.6	31.2 ± 3.4	39.4 ± 1.5
SRW-18	493.6 ± 13.9	30.8 ± 1.2	36.3 ± 1.5	646.2 ± 16.9	37.5 ± 1.3	51.9 ± 1.9
SRW-19	549.5 ± 14.7	34.7 ± 1.2	47.0 ± 1.7	538.3 ± 14.2	34.0 ± 1.2	44.2 ± 1.6
SRW-20	610.0 ± 16.2	31.5 ± 1.2	47.4 ± 1.8	709.0 ± 18.7	38.3 ± 1.4	51.0 ± 2.0
SRW-21	652.9 ± 16.4	31.6 ± 1.2	43.2 ± 1.7	623.9 ± 16.5	35.4 ± 1.3	48.8 ± 1.8
SRW-22	682.2 ± 17.1	36.5 ± 1.3	54.1 ± 1.9	765.7 ± 19.8	40.4 ± 1.5	59.9 ± 2.2
SRW-23	567.0 ± 15.1	30.8 ± 1.2	47.9 ± 1.7	636.7 ± 16.2	37.5 ± 1.3	55.3 ± 1.9
SRW-24	512.2 ± 14.5	31.9 ± 1.2	55.6 ± 1.9	558.4 ± 14.4	36.2 ± 1.2	55.1 ± 1.9
SRW-25	650.5 ± 16.7	34.3 ± 1.3	51.7 ± 1.9	495.9 ± 13.1	35.0 ± 1.2	46.7 ± 1.7
SRW-26	558.9 ± 15.1	34.5 ± 1.2	51.87 ± 1.8	522.0 ± 13.6	36.1 ± 1.3	55.0 ± 2.0
SRW-27	521.7 ± 14.7	34.0 ± 1.2	52.1 ± 1.8	464.4 ± 11.9	36.5 ± 1.3	52.7 ± 1.9
SRW-28	538.2 ± 14.8	31.3 ± 1.1	45.7 ± 1.7	511.6 ± 13.4	39.2 ± 1.5	59.9 ± 2.2
SRW-29	570.1 ± 15.5	31.7 ± 1.2	44.5 ± 1.7	393.1 ± 10.9	33.5 ± 1.3	49.2 ± 1.9
SRW-30	534.3 ± 15.1	29.7 ± 1.1	48.5 ± 1.8	335.7 ± 09.1	33.0 ± 1.2	50.2 ± 1.8

Table 3. Naturally occurring radionuclides activity in soil samples collected in harvesting season of Kharif -2015 and Rabi- 2016 crop field's

Sample	Activity Concentration (Bq Kg^{-1})					
	Kharif Field's Soil (November – 2015)			Rabi Field's Soil (May – 2016)		
	^{40}K	^{232}Th	^{226}Ra	^{40}K	^{232}Th	^{226}Ra
SRW-1	667.0 ± 17.2	39.6 ± 1.4	54.7 ± 1.9	621.8 ± 17.0	37.9 ± 1.4	52.7 ± 20
SRW-2	643.3 ± 16.2	37.3 ± 1.3	50.4 ± 1.8	643.5 ± 17.3	38.4 ± 1.4	51.8 ± 1.9
SRW-3	650.1 ± 17.3	36.9 ± 1.4	49.2 ± 1.9	610.2 ± 16.5	39.6 ± 1.4	55.8 ± 2.0
SRW-4	575.4 ± 15.5	32.4 ± 1.2	45.8 ± 1.7	473.1 ± 14.3	28.7 ± 1.2	39.6 ± 1.6
SRW-5	590.9 ± 16.5	35.5 ± 1.3	50.1 ± 1.9	582.5 ± 16.3	35.4 ± 1.3	47.6 ± 1.8
SRW-6	484.2 ± 13.8	31.5 ± 1.2	41.3 ± 1.6	583.2 ± 16.1	34.3 ± 1.3	50.4 ±1.9
SRW-7	652.8 ± 17.7	36.0 ± 1.4	51.0 ± 1.9	533.9 ± 14.3	31.6 ± 1.2	44.0 ± 1.6
SRW-8	681.4 ± 16.8	35.6 ± 1.3	47.5 ± 1.7	668.0 ± 17.3	40.9 ± 1.4	57.9 ± 2.0
SRW-9	611.9 ± 15.8	32.7 ± 1.2	45.1 ± 1.7	622.4 ± 16.0	35.3 ± 1.3	50.9 ± 1.8
SRW-10	737.4 ± 18.3	33.8 ± 1.3	50.9 ± 1.9	476.4 ± 13.9	34.1 ± 1.2	46.4 ± 1.7
SRW-11	554.6 ± 14.8	31.6 ± 1.2	41.5 ± 1.6	565.1 ± 15.6	34.8 ± 1.3	45.7 ± 1.8
SRW-12	584.7 ± 15.9	34.3 ± 1.3	47.0 ± 1.8	563.9 ± 15.1	33.8 ± 1.2	44.7 ± 1.7
SRW-13	586.2 ± 15.6	32.3 ± 1.2	47.8 ± 1.7	590.3 ± 16.4	37.7 ± 1.4	52.2 ± 1.9
SRW-14	568.3 ± 15.3	32. 2 ± 1.2	46.7 ± 1.7	595.9 ± 16.4	38.2 ± 1.4	53.0 ± 1.9
SRW-15	556.7 ± 14.9	35.0 ± 1.2	49.3 ± 1.7	619.8 ± 16.4	33.6 ± 1.3	45.3 ± 1.8
SRW-16	537.7 ± 14.3	32.4 ± 1.2	40.3 ± 1.5	603.0 ± 16.4	33.1 ± 1.3	46.2 ± 1.8
SRW-17	530.1 ± 14.8	32.6 ± 1.2	45.0 ± 1.7	486.8 ± 13.9	32.8 ± 1.2	41.6 ± 1.6
SRW-18	635.0 ± 16.9	34.3 ± 1.3	47.8 ± 1.8	668.0 ± 17.1	37.1 ± 1.3	53. ± 1.9
SRW-19	494.7 ± 14.2	31.8 ± 1.2	39.7 ± 1.6	552.1 ± 14.9	33.1 ± 1.2	44.5 ± 1.7
SRW-20	578.1 ± 16.3	34.0 ± 1.3	46.1 ± 1.8	570.2 ± 16.2	32.3 ± 1.3	45.3 ± 1.8
SRW-21	443.4 ± 12.7	27.9 ± 1.1	35.8 ± 1.4	757.9 ± 19.8	40.9 ± 1.5	54.3 ± 2.1
SRW-22	609.8 ± 15.5	31.3 ± 1.2	46.9 ± 1.7	729.8 ± 18.0	37.4 ± 1.4	55.3 ± 2.0
SRW-23	610.0 ± 16.4	37.6 ± 1.3	54.1 ± 1.9	639.7 ± 16.4	34.4 ± 1.3	50.1 ± 1.8
SRW-24	599.5 ± 16.1	38.1 ± 1.3	52.0 ± 1.8	689.9 ± 17.6	40.6 ± 1.5	56.7 ± 2.1
SRW-25	600.2 ± 15.7	37.8 ± 1.3	49.8 ± 1.7	657.9 ± 16.4	39.8 ± 1.5	56.6 ± 2.0
SRW-26	549.0 ± 15.1	33.9 ± 1.2	44.9 ± 1.6	607.9 ± 14.6	38.3 ± 1.4	53.5 ± 1.9
SRW-27	664.9 ± 17.7	37.4 ± 1.4	49.0 ± 1.8	440.2 ± 11.3	35.0 ± 1.2	46.1 ± 1.6
SRW-28	493.1 ± 13.8	29.2 ± 1.1	38.6 ± 1.4	448.1 ± 11.9	34.1 ± 1.3	49.5 ± 1.8
SRW-29	550.4 ± 14.9	31.8 ± 1.2	37.4 ± 1.4	360.3 ± 09.9	31.0 ± 1.2	40.1 ± 1.5
SRW-30	654.6 ± 16.7	33.9 ± 1.3	38.2 ± 1.5	330.4 ± 09.2	34.7 ± 1.3	43.6 ± 1.6

Table 4. Comparison of present study soil natural radioactivity results with other worldwide study

Region	Methodology	^{40}K	^{232}Th	$^{226}Ra/^{238}U$	References
Eastern Haryana	HPGe	46.3 - 696	31.4 - 37.9	41.5 – 54.4	Present Study
South-Eastern Nigeria	NaI (Tl)	100.7	77.7	33.2	Osimobi, et al. (2018)
Shabwah and Hadramout, Yemen	HPGe	566.0	25.7	14.3	Nafee et al. (2017)
Wadi Arar, Saudi Arabia	HPGe	132-305.9	8.5-20.1	19.8-26.2	Alghamdi and Diab (2017)
Itagunmodi, Nigeria	NaI(Tl)	102.8	17.5	8.8	Ademola et al. (2014)
Hisar, Haryana	HPGe	360.0	45.5	17.8	Kansal *et al*. (2010)
Haryana, India	HPGe	445–708.7	24.2–39.0	31.9– 50.9	Yadav et al. (2018)
Karnataka	HPGe	791.58	77.44	33.78	Srilatha et al. (2015)
Shivalik Hills, Himachal Pradesh	HPGe	472 - 630	53 -78	31 - 63	Chauhan et al. (2014)
Abha, Saudi Arabia	NaI (Tl)	182- 251	23.4- 41.9	38.2 –44.1	Ibraheem et al. (2018).
Sarikamis Kars	NaI (Tl)	148-909	7.6-53	BDL-38.1	Cengiz & Çaglar (2017)
Kogi, Nigeria	NaI (Tl)	508.86	18.90	41.27	Ilemona et al. (2016)
Rudoltowice, Poland	HPGe	428 – 580	53 - 83	33 -155	Sleziak (2010)
Jessore, Bangladesh	HPGe	345-674	33-70	28-67	Kabir et al. (2009)
Ramanagara and Tumkur, Karnataka	HPGe	607 - 1108	52.5 - 117.8	22.1 - 59.8	Rangaswamy et al. (2016)
Tiruchirappalli, Tamil Nadu	HPGe	55.9 - 717.5	BDL - 464.8	BDL- 60.0	Pillai et al. (2016)
Jalandhar and Hoshiarpur Punjab	HPGe	738- 1064	24 - 334	6.37- 56.7	Badhan et al. (2017).
Chittagong, Bangladesh	HPGe	787- 1125	67-102	41 – 86	Yesmin et al. (2018)
Raichur district, Karnataka,	NaI(Tl)	46–1646	8–285	10–119	Rajesh and Kerur (2018)
Tumkur, Karnataka	HPGe	194 - 1528	12.3 - 333	9.6 -71.6	Jayasheelan et al. (2014)
Jadugura, Jharkhand	HPGe	291 - 1391	8.7 - 237	6.1 - 826	Mahur et al. (2013)
Kerala	NaI(Tl)	<21.5 - 693	<5.5 - 5328	<5.5 - 1187	Ramasamy et al. (2013)
Upper Shivaliks, Northern India	HPGe	363 - 1002	61.2 - 140	28.3 - 81	Singh et al. (2009)
West Khasi Hills, Meghalaya		173 - 359	175 - 284	51-335	War et al. (2008)
Rize Province city, Turkey	NaI (TI)	35.7-913	9.5-170.8	7.4-79.8	Durusoy &Yildirim (2017)
Lombardia, Italy	HPGe	242-1434	24-231	20-70	Guidotti et al. (20015)

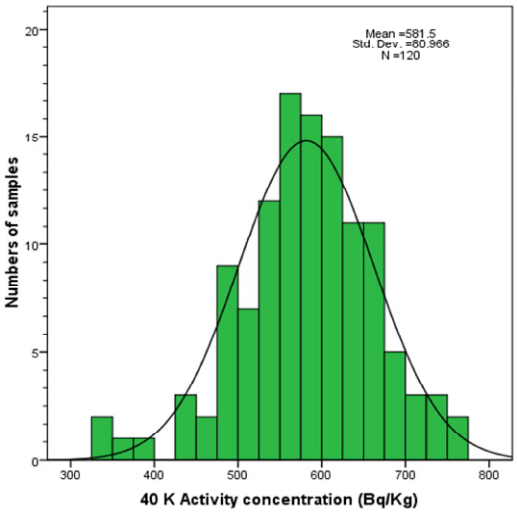

Fig.2. Frequency distribution for activity concentration of ^{40}K

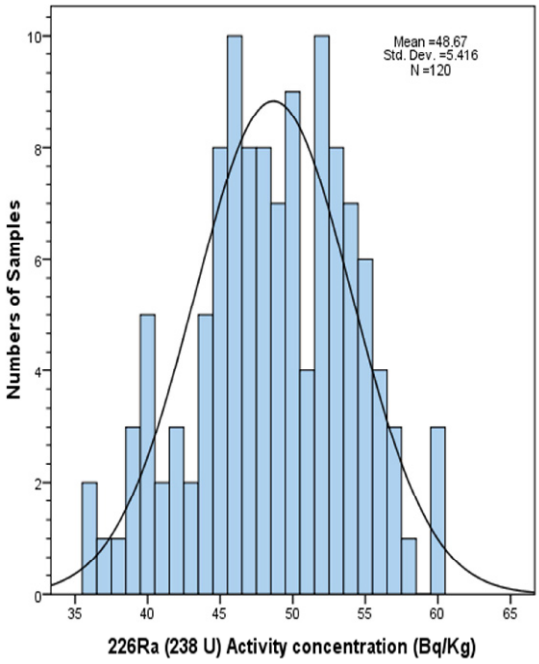

Fig.3. Frequency distribution of activity concentration of ^{226}Ra (^{238}U)

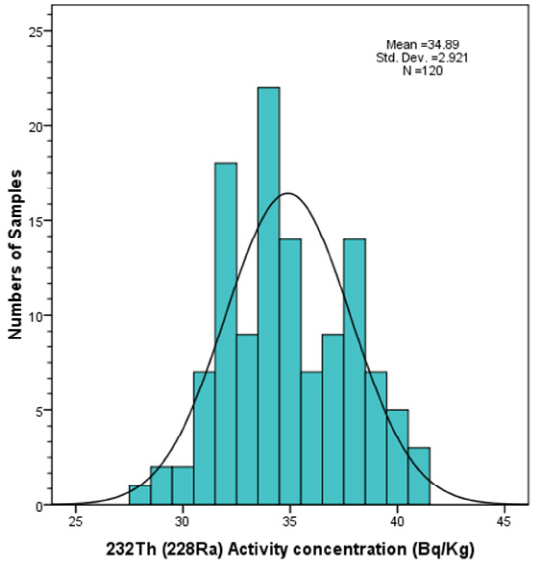

Fig.4. Frequency distribution of activity concentration of ^{232}Th

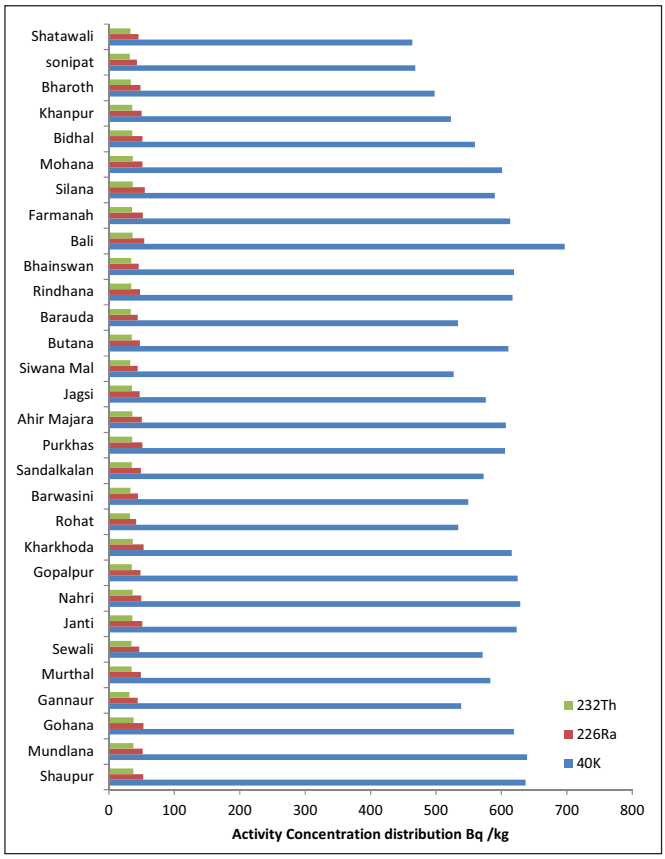

Fig.5. Distribution of activity concentration of ^{40}K, ^{232}Th and ^{226}Ra in different villages of Sonipat district, Haryana

samples were collected from agriculture fields and may also be due to the weathering of potassium from potassium rich rocks (Sherifet al.2017). Moreover, the soil of study area has been deposited from the Shivalik hills by Yamuna river. The deposit consists of gravels, kankars and unconsolidated sand which are rich in long lived uranium and thorium (Singh et al. 2007; Kumar et al. 2014).

Radium Equivalent (Ra_{eq}) Activity

The Ra_{eq} activity is used to compare the activity of ^{226}Ra, ^{232}Th and ^{40}K in the building materials such as bricks soil, tiles etc. This is a common radiological index helps to estimate total radiation exposure to human in terms of Radium equivalent (Ra_{eq}) activity. The Ra_{eq} in soil samples were in range of 124.7 – 159.8Bq kg^{-1} with the mean value of 143.3 ± 9.3 Bqkg^{-1}. Ra_{eq} values at all locations are less than maximum permissible limit (370 Bq kg^{-1}) that has been accepted for safer use. Average Ra_{eq} value is 2.4 times lesser than the maximum permissible value. Radium equivalent activity was maximum in Bali village and minimum in Sonipat. This may be related to geology, groundwater use for irrigation and other agricultural practices like kind of fertilizer addition. Water table is low in Bali village region compared to Sonipat village region and this may affect the leaching of metals in this region.

Dose Rate, Dose Equivalents and Other Hazard Indices

The external air absorbed dose of gamma radiation due to ^{226}Ra, ^{232}Th and ^{40}K at about 1.0 m on top of the ground was calculated. Air absorbed dose due to natural radioactivity in the soil of Sonipat district ranged from 58.6 to 76.0 nGy h^{-1} with mean value of 67.8 ± 4.5 nGy h^{-1}. Air absorbed dose in study area is nearly comparable to the Indian average of 64 nGy h^{-1}. The outdoor annual effective dose equivalent ranged from 0.071 to 0.093 with the mean value 0.083± 0.005 μSv y^{-1}. The annual effective dose equivalent, at all the location

in study area is very much lower than the worldwide average of 70.0 μSv y^{-1} (UNSCEAR 2000). Annual Gonadal Equivalent Dose values ranged from 413.2 to 538.0 μSv y^{-1} with mean value 478.8 ± 32.1 μSv y^{-1}. Calculated values are greater than the worldwide average 300 μSv y^{-1} (UNSCEAR 2000). Average value in study area was 1.6 times higher than the world average value. Active bone marrow, bone surface cell and gonads are considered to be mainly affected by gamma radiations of natural radionuclides (UNSCEAR 1982). The higher values of calculated gonadal dose may be due high natural radionuclides activity of ^{226}Ra and ^{232}Th this may be attributed to Shivalik hill's plain (Chauhan, 2010) and Yamuna river as it brings silt with it which has comparatively higher activity.

Gamma index represents the total radiological hazard associated with soil due to presence of ^{226}Ra, ^{232}Th and ^{40}K. In the soil, gamma index in study area ranged from 0.459-0.595 with average value 0.531 ±.035. According to European Commission recommended value of gamma Index must be < 1.0 to keep the radiation hazards insignificant to population hence it is criteria of unity corresponds to effective dose of 300 μSv y^{-1}. Gamma Index is <1.0, in study area indicating that soil of study area do not pose any radiation hazard and is safe to the population.

The Outside hazard index due to soil in study area ranged from 0.337 to 0.432 with mean value 0.387 ±0.02. All calculated values are less than unity so if soil of Sonipat district is used for making buildings materials will not have external hazard. Calculated inside hazard index in study area due to ^{226}Ra, ^{232}Th and ^{40}K due to soil ranged from 0.452 to 0.578 μSv y^{-1} with the mean value 0.519 ± 0.03 μSv y^{-1}. Inside hazard index is for all studied locations is <1.0, indicating that all values are below the recommended value. Based on results it can be inferred that there is no potential internal radiation hazard due to these naturally occurring radionuclides present in the soil of the study area. Outdoor excess lifetime cancer risk in soil of study area due to ^{226}Ra, ^{232}Th and ^{40}K was calculated and overall cancer risk ranged from 0.236×10^{-3} to 0.303×10^{-3} with the mean value $(0.270 \pm 0.01) \times 10^{-3}$. However the world average value is 0.29×10^{-3}, indicating that in study area cancer risk was lower than world average.

CONCLUSION

The average activity concentration of ^{40}K, ^{232}Th and ^{226}Ra in study area is higher than the global average value 400 Bq kg^{-1}, 30Bq kg^{-1} and 35 Bq kg^{-1} correspondingly (UNSCEAR, 2000). Excess use of fertilizers during the agricultural practices, may contaminate the top soil and enhanced the terrestrial background activity in the study area. Organic and Inorganic matters from Shivalik hills and sediments of Yamuna river may be other causes of enhancement of natural radionuclides content. Radium equivalent activity is 2.4 times lower than the maximum permissible value. Average value for Air Absorbed Dose is comparable to Indian average. Annual effective dose equivalent is very much lower than world average. AGED in study area is much lower than (1.0 mSy^{-1}) recommended by International Commission on Radiological Protection for general public. The average outdoor cancer risk due to natural radioactivity in soil samples of study area was less than the world average. Average Gamma index, outside hazard index and inside hazard index is less than unity indicating that there is no significant health risk due to radioactivity in soil and this soil can also be safely used as construction material. Present study provides useful reference information about baseline data on soil natural radioactivity level in study area, which is useful for further studies. Moreover, understanding of radionuclides environmental actions by knowing distribution prototype in different segment of the soil is of major significance for the selection of best possible remedial technologies. It is recommended to study cyclic variations in determining radioactivity. This is because during rainy season rivers and groundwater tables which irrigate the study area might have higher concentration of radionuclides due to the soil corrosion. The investigation of Th/U ratios possibly will help us to know the reduction/enrichment due to weathering and complex metamorphic history that have affected the soil of study area.

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