

# Assessment of primordial and anthropogenic radionuclide contents in the soil samples of lower Himalayas of Jammu & Kashmir, India

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#### Abstract

The present study deals with the assessment of primordial radionuclide contents  $(^{238}$ U,  $^{232}$ Th, and  $^{40}$ K) and anthropogenic radionuclide content  $({}^{137}Cs)$  in the soil samples collected from the villages of Lower Himalayas of Jammu & Kashmir, India by using thallium doped sodium iodide (NaI(Tl)), a scintillator detector. The elemental concentration of U, Th, and K in soil samples has been calculated. The average values of radionuclide contents were found to be well within the average values suggested by UNSCEAR (United Nations Scientific Committee on the effects of atomic radiation, sources and effects of ionizing radiation, [1\)](#page-9-0). All values of various calculated radiological hazards were found to be below the recommended limits suggested by various agencies.

Keywords Radionuclide contents · Hazard indices · Absorbed dose rate · Level indices · Elemental concentration · Soil sample

# Introduction

Radionuclides existing in the environment are either natural or artificial. Uranium and thorium series and  $^{40}$ K nuclides are the most widely dispersed in the lithosphere. The gamma radiations emitted from these primordial radionuclides acts as the main external source of irradiation to the human body. The content of the natural radionuclides in the soil rely predominantly upon the geographical and geological condition of the soil [[1\]](#page-9-0), and to the content in potassium (K), uranium (U), and thorium (Th) of the rock from which the soils emanate in each area. Besides naturally occurring radionuclides, there are also artificial radionuclides have been released into the environment by

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different processes. The  $137Cs$  is one of them and is believed to be deposited by atmospheric weapon tests, medical applications, industrial activities and remains in biota and living organisms because of its relatively long physical half-life [\[2](#page-9-0)]. Its chemical and ecophysiological similarity with potassium, abundance among fission products, long physical half-life (30.14 years) and relatively high volatility makes radiocesium one of the most important man-made radionuclides released into the environment. Human beings have been exposed to a wide range of ionizing radiations from these naturally occurring radioactive materials (NORMs) and from artificial radionuclides [\[3](#page-9-0)]. In order to determine whether the region people live in is safe and healthy in terms of the natural radionuclides concentrations in the environment, the concentration of radionuclides in the soil and the effects of radiation on human health must be known.

The study related the natural radioactivity in the soil is important because of their radiological effect and also they serve as an excellent biochemical and geochemical traces [\[4](#page-9-0)]. The estimation of health effects due to exposure of ionizing radiation from natural sources needs awareness of its distribution in the environment. The level of presence of natural radioactivity in soil and water is significant principally due to two radiological effects. The first is internal

exposure to radiation from radioelements in diet and inhaled radon and its progenies which emanate from soil, sediment and building materials results in irradiation of the lungs by alpha-emitting short-lived progeny of radon and thoron and the second effect is the external irradiation of the body by gamma rays emitted from primordial radionuclides present in the area [\[5](#page-9-0)]. The long-term exposures to internal and external sources have serious health effects such as chronic lung cancer and leukemia [\[6](#page-9-0), [7\]](#page-9-0). Therefore, the assessment of natural radioactivity in the soil is of a great interest for various workers all over the world, which led to the worldwide national surveys in the last two decades [[8–11\]](#page-9-0).

The prime goal of this study is to measure the concentration of primordial radionuclide contents i.e.,  $^{238}$ U,  $^{232}$ Th, and  $^{40}$ K, and anthropogenic radionuclide content i.e., <sup>137</sup>Cs, respectively in the soil samples collected from the Lower Himalayas of Jammu & Kashmir, India. From this, the various radiological hazards i.e., radium equivalent activity, criteria formula, internal and external health effects, gamma index, absorbed dose rate, and annual effective dose equivalent, respectively have been calculated for the health related threat. The elemental concentration of the primordial radionuclide contents has been calculated also. The two physicochemical parameters i.e., pH and electrical conductivity were also determined in the collected soil samples. The result obtained from this study will serve as radioactivity database for the area and will also be relevant in the radiological mapping of the present area.

# Geology

The present study has been placed in the villages of the lower Himalayas of the Jammu & Kashmir, India (Fig. [1](#page-2-0)). The present villages come under the Reasi and Jammu Districts of Jammu & Kashmir. The Jammu District situates in the foothills of the sub-mountainous region of the Himalayas. This District is bounded by Reasi and Udhampur District in the north and northeast, Pakistan and Rajouri district in the west, Punjab state in the south, and Kathua district in the southeast. The selected eight locations of Jammu district come under the northern hilly area underlain by Siwalik rocks of mio-pliocene age. The Siwalik group is delimited by the lesser Himalaya to the north and Indo-Gangetic plain to the south.

Out of selected eight locations, five locations are situated on middle siwalik and three locations are situated on lower Siwalik. The lower Siwalik group is of middle to upper Miocene age. Red mudstones and sandstones are taken as characteristic for lower Siwalik rocks. The sandstones of the area come under lower Siwalik are less compact and fine to medium grained and have more

fragments. On the other hand, the middle Siwalik group is of upper Miocene to middle Pliocene age and is mainly sandy with subordinate grey and brown mudstones. The sandstones of this area are mainly multistoried, grey, and medium to coarse-grained, soft and pebby salt and pepper sandstones turning into conglomerates.

However, the Reasi District is situated in the Siwalik Hills and it falls in the mid-hill zone of Jammu & Kashmir state. It is underlain by rock formations range in age from pre-cambrian to quaternary period. Reasi District lies between longitude  $33^{\circ}05'56.58''$ N and latitude 74°42′26.88″E, at a distance of 64 km from Jammu and is bounded by Tehsil Gool-Gulabgarh in the north, Tehsil Sunderbani and Kalakote of Rajouri District in the west, Udhampur District in the east, Tehsils Jammu and Akhnoor of district Jammu on the south. There is a sirban limestone also known as reasi limestone, which occurs in a chain of inliers in the tertiary rock formations. It is hard massive compact limestone occurring as an elongated dome of which southern limb has been eroded and new only northern is preserved. The limestone is light grey to dark grey in color. The lower part is dolomite and the upper part is cherty and silicious. The soil cover is thin at higher altitudes and becomes deep to very deep at lower altitudes i.e., at foothills.

# Experimental work

# Sampling strategy

The soil samples were collected at the depth of 15–20 cm from the soil profile to obtain undisturbed and pure soil samples. About 1 kg soil samples were collected in a polyethylene bag from each location. The soil samples were not collected from the surface because at the soil surface the radium nuclides might have been flushed away by wind or water. While collecting the soil samples, this was kept in mind that soil samples must be free from gravels and pebbles. The collected soil samples were packed and named properly. These were dried in open air to remove moisture and humidity totally. The dried samples were crushed by using mortar and pestle and sieved with a standard size of sieve i.e.,  $150 \mu m$  to make homogeneity in the samples. The sieved soil samples were packed in an airtight cylindrical polypropylene container with height 8 cm and diameter 6.5 cm (i.e., vol. is  $265 \text{ cm}^3$ ) and sealed the container with tape properly to avoid the leakage of radon gas from it. The airtight containers were stored for 30 days at room temperature until radium and thorium and their decay products reached their radioactive equilibrium [[11](#page-9-0)].

<span id="page-2-0"></span>

## Physicochemical properties

The pH and electric conductivity in soil samples were determined by using Micro Processor Based Water Analysis Kit NPC 362D. Each soil sample was mixed with distilled water in the proportion of 1:2.5 (w/v) for the measurement of pH and to measure the electrical conductivity (EC), the mixture of soil and distilled water in the proportion of 1:1.5 (w/v) was prepared, respectively [\[12](#page-9-0)]. The soil pH was determined with pH probe in soil water suspension and calibrated with three buffer solution (pH 4.0, 7.0, 11.0).

#### Gamma spectrometry

The radionuclide contents from the prepared soil samples were assessed by using thallium doped sodium iodide detector i.e., NaI(Tl). The scintillation gamma radiation detection unit with the NaI(Tl) crystal of size  $63 \times 63$  mm is used as a gamma radiation detector with multiple channel analyzer. The spectrometer ensures the registration of the gamma radiation within the energy range from 50 to

3000 keV. The NaI(Tl) detector has high detection efficiency, easy to handle and required low maintenance. The efficiency and energy calibration of the spectrometer were determined with standard IAEA-375 calibration material. The detector has a resolution of 10% at the 0.662 MeV line of  $137Cs$ , which is capable of distinguishing the gamma-ray energies of the radionuclides considered in this study. The activity concentration of  $^{238}$ U and  $^{232}$ Th in soil has been determined from the 1764 keV gamma line of  $^{214}$ Bi and 2610 keV gamma line of  $^{208}$ Tl, respectively. In order to calculate the activity concentrations of  $^{137}Cs$  and  $^{40}K$ , the 661.7 and 1460 keV gamma lines were analyzed, respectively. The measurement time for both activity and background measurement was 10,800 s. The Maestro software provides significant photo peaks in the spectrum. The specific radioactivity of the radionuclides present in the samples were evaluated from the neat peak areas of full energy peaks after background subtraction and taking into account the detection efficiency of the system, the branching ratio of gamma transition. Lead absorbers (thickness of 10 cm) were also placed around the apparatus to reduce background radiation. The specific activity of a nuclide is then obtained by using equation [[13\]](#page-9-0):

Activity 
$$
(Bq \, kg^{-1}) = \frac{N_{nE}}{\gamma_D \times \epsilon_E \times t \times M2}
$$
 (1)

The definition of the factors given in the above formula has been provided in Table 1.

## Elemental concentration of radionuclides

The activity concentrations of  ${}^{40}$ K,  ${}^{238}$ U, and  ${}^{232}$ Th has been converted into the elemental concentrations of K (in %), U (in ppm), and Th (in ppm) and using the following formula [\[5](#page-9-0), [10](#page-9-0), [14,](#page-9-0) [15\]](#page-9-0):

$$
E_C = \frac{A_C \times A_{M,E} \times F}{\lambda_E \times N_{Av} \times h_E}
$$
 (2)

The definition and values of various factors are given in Table 1.

#### Statistical analysis

Descriptive statistical parameters, i.e., minimum, maximum, average and standard deviation of the radionuclide content, radiological hazards and physicochemical parameters have been measured. Correlations were measured of the data by using Pearson's correlation analysis. One sample  $T$  test was applied to compare the means of radionuclide contents in Lower and Middle Siwalik, respectively. All statistical analysis conducted in present paper has been done using SPSS software-version (16.0) at the 95% confidence level.

#### Radiological hazards

The radionuclide contents present in the soil samples contributes to the various radiological health hazards including level indices, hazard indices, and dose rate.

Table 1 Definitions and values of the constant and variables factors

Factors (units)	Definition	Values			
$N_{nE}$	Net count of a nuclide n in a peak at energy E				
$E_{C}$	Elemental concentration of radionuclides				
$A_C$ (Bq kg <sup>-1</sup> )	Measured activity concentration of radionuclides				
M(g)	Mass of sample	150			
t(s)	Counting time for a sample	10,800			
$N_{Av}$ (atoms mol <sup>-1</sup> )	Avogadro's number	$6.023 \times 10^{23}$			
$\gamma_{\rm D}$	Gamma ray abundance per disintegration of the	16.7 for $^{238}$ U,			
	specific nuclide at E	36 for <sup>232</sup> Th and			
		10.7 for ${}^{40}$ K			
$\epsilon_{\rm E}$	Detection efficiency of crystal at energy E	1.47 for $^{238}$ U,			
		0.55 for $^{232}$ Th, and			
		1.732 for ${}^{40}$ K			
		0.097 for ${}^{137}Cs$			
$A_{M, E}$ (kg mol <sup>-1</sup> )	Atomic mass of radionuclides	0.238 for $^{238}$ U,			
		0.232 for $^{232}$ Th,			
		0.039 for ${}^{40}$ K, and			
		0.136 for ${}^{137}Cs$			
$h_E(\%)$	Atomic abundance in nature	99.27 for <sup>238</sup> U			
		100 for $^{232}$ Th and			
		0.012 for ${}^{40}$ K			
		0 (trace) for ${}^{137}Cs$			
$\lambda_{\rm E}$ (s <sup>-1</sup> )	Decay constant	$4.9 \times 10^{-18}$ for <sup>238</sup> U,			
		$1.6 \times 10^{-18}$ for <sup>232</sup> Th and			
		$1.8 \times 10^{-17}$ for <sup>40</sup> K			
		$7.3 \times 10^{-10}$ for $^{137}Cs$			
F (mg kg <sup>-1</sup> ) (%)	A factor	1,000,000 for <sup>238</sup> U and <sup>232</sup> Th			
		100 for ${}^{40}\text{K}$			

These various radiological hazards have been described as below:

## Radium equivalent activity ( $Ra_{eq}$ )

The radium equivalent activity ( $Ra_{eq}$ ) in Bq kg<sup>-1</sup> was introduced to define the uniformity of the dispensation of primordial radionuclides in soils and to evaluate the radiation threat emerging due to the exposure of the presence of primordial radionuclides in the soil [[16\]](#page-9-0). This radium equivalent activity represents a weighted sum of activities of  $40$ K,  $^{238}$ U, and  $^{232}$ Th radionuclides and is based on the estimation that 370, 259, 4810 Bq kg<sup>-1</sup> of <sup>238</sup>U, <sup>232</sup>Th, and 40K produce the same radiation dose rates. It is calculated by the following formula [\[5](#page-9-0)]:

$$
Ra_{eq} = 370 \left( \frac{A_{Th}}{259} + \frac{A_U}{370} + \frac{A_K}{4810} \right)
$$
  
\n
$$
\Rightarrow Ra_{eq} = 1.43 A_{Th} + A_U + 0.077 A_K
$$
\n(3)

where  $A_{Th}$ ,  $A_U$ , and  $A_K$  are the specific activity (Bq kg<sup>-1</sup>) of  $^{232}$ Th,  $^{238}$ U, and  $^{40}$ K, respectively in the soil samples.

#### Criteria formula (CF)

The results of Keller and Muth [[17\]](#page-9-0), used as a standard to bound the yearly radiation dose from soil samples, such as based on the following formula:

$$
CF = \frac{A_U}{740 B q kg^{-1}} + \frac{A_{Th}}{520 B q kg^{-1}} + \frac{A_K}{9620 B q kg^{-1}} \le 1
$$
\n(4)

#### External ( $H_{ext}$ ) and internal ( $H_{int}$ ) health effects

The external and internal health effects due to the presence of a radionuclide in the soil are given by the formula UNSCEAR [\[5](#page-9-0)]. The external hazard index is a radiation threat index which evaluates the indoor radiation dose due to the external exposure to gamma radiation from the natural radionuclides in the construction building materials of dwellings.

$$
H_{ext} = \frac{A_U}{370 Bq kg^{-1}} + \frac{A_{Th}}{259 Bq kg^{-1}} + \frac{A_K}{4810 Bq kg^{-1}} \le 1
$$
\n(5)

$$
H_{int} = \frac{A_U}{185 \, Bq \, kg^{-1}} + \frac{A_{Th}}{259 \, Bq \, kg^{-1}} + \frac{A_K}{4810 \, Bq \, kg^{-1}} \le 1 \tag{6}
$$

# Gamma index  $(I_{\gamma})$

The radiation risk from excessive gamma exposure can be calculated by gamma activity index  $I_{\gamma}$  as suggested by the European Commission [[18\]](#page-9-0):

$$
I_{\gamma} = \frac{A_U}{300 Bq kg^{-1}} + \frac{A_{Th}}{200 Bq kg^{-1}} + \frac{A_K}{3000 Bq kg^{-1}} \le 1
$$
\n(7)

## Absorbed dose rate (ADR)

The absorbed dose rates (D) due to primordial gamma rays and anthropogenic gamma rays are obtained from  $^{40}$ K,  $238$ U,  $232$ Th, and  $137$ Cs concentration, which arise from radioactive sources in the soil and is significant in the radiation risk study since it measures the quantity of radiation lodged per unit time. The gamma absorbed dose rate calculated using the Eq.  $8$  given below  $[5, 10, 14, 15]$  $[5, 10, 14, 15]$  $[5, 10, 14, 15]$  $[5, 10, 14, 15]$  $[5, 10, 14, 15]$  $[5, 10, 14, 15]$  $[5, 10, 14, 15]$  $[5, 10, 14, 15]$  $[5, 10, 14, 15]$ :

$$
D (nGy h^{-1}) = 0.043AK + 0.662ATh + 0.427AU
$$
  
+ 0.030A<sub>Cs</sub> (8)

where the numerical factors are the conversion factors from Bq kg<sup>-1</sup> to nGy h<sup>-1</sup> [[5,](#page-9-0) [10,](#page-9-0) [14](#page-9-0), [15](#page-9-0)].

## Annual effective dose equivalent (AEDE)

To make a basic estimate of the annual ambient dose, the conversion coefficient from absorbed dose in the air to effective dose and the outdoor occupancy factor has been used. The annual effective dose resulting from the absorbed dose is also obtained using following formula [[5,](#page-9-0) [14](#page-9-0), [15](#page-9-0)]:

AEDE 
$$
(\mu Sv y^{-1})
$$
 = Absorbed dose  $(nGy h^{-1}) \times T_{exp}$   
  $\times \text{ OF } \times \text{ DCF } \times 10^{-3}$  (9)

where  $T_{exp}$  is the outdoor exposure duration per year, i.e., 8760 h  $y^{-1}$ , OF is an occupancy factor for outdoor i.e., 0.2 [\[5](#page-9-0), [19](#page-9-0)] effective dose and DCF is an effective dose of the absorbed dose conversion factor  $(0.7 \text{ Sv } \text{Gy}^{-1})$ .

## Results and discussion

#### Physicochemical parameters

The values of measured two physicochemical parameters i.e., pH and electrical conductivity, EC in assembled soil samples are presented in Table [2](#page-5-0). The value of pH has been ranged from 6.2 to 8.7 with an average value of  $7.2 \pm 0.7$ which indicated the slightly alkaline nature. The average value of EC was  $246 \pm 131 \,\mu\text{S cm}^{-1}$  with the minimum

Sr. no.	<b>Villages</b>	Rock types	Physico-chemical parameters				Radionuclide contents (Bq $kg^{-1}$ )		Elemental concentration			
			pH	EC $(\mu \text{Scm}^{-1})$	$\rm A_{U}$	$A_{Th}$	$A_K$	$A_{Cs}$	$E_{\rm H}$ $(mg kg^{-1})$	$E_{Th}$ $(mg kg^{-1})$	$E_K$ $(\%)$	
$\mathbf{1}$	Aghar Jitto	M.S.	6.45	104	$30 \pm 6$	$38 \pm 8$	$367 \pm 133$	$3 \pm 1$	2.4	$\mathbf{Q}$	1.1	0.8
2	Laiter	M.S.	7.4	222	$30 \pm 5$	$54 \pm 7$	$373 \pm 110$	$5 \pm 1$	2.4	13	1.1	0.6
3	Pouni-mari	M.S.	7.6	126	$31 \pm 5$	$49 \pm 6$	$399 \pm 109$	$11 \pm 3$	2.5	12	1.2	0.6
$\overline{4}$	Pouni-purria	M.S.	7.73	154	$38 \pm 6$	$42 \pm 7$	$281 \pm 110$	$14 \pm 4$	3.1	10	0.8	0.9
5	Bhagha	M.S.	7.8	273	$36 \pm 6$	$39 \pm 8$	$368 \pm 115$	$12 \pm 3$	2.9	9	1.1	0.9
6	Katra	M.S.	7.05	536	$20 \pm 4$	$28 \pm 4$	$305 \pm 99$	$3 \pm 1$	1.6	$\overline{7}$	0.9	0.7
$\tau$	Dabh jagir	M.S.	8.67	114	$33 \pm 6$	$45 \pm 7$	$325 \pm 115$	$2 \pm 0.6$	2.7	11	1.0	0.7
8	Dhanoa	M.S.	7.72	261	$28 \pm 5$	$38 \pm 5$	$304 \pm 108$	$13 \pm 3$	2.3	9	0.9	0.7
9	Serli- chamba	M.S.	7.7	378	$27 \pm 6$	$45 \pm 7$	$334 \pm 103$	$8 \pm 2$	2.2	11	1.0	0.6
10	Maghal	M.S.	8.05	273	$29 \pm 5$	$53 \pm 7$	$348 \pm 113$	$7 \pm 2$	2.4	13	1.0	0.5
11	Akhli	M.S.	6.2	112	$22 \pm 6$	$30 \pm 5$	$378 \pm 110$	$12 \pm 3$	1.8	$\tau$	1.1	0.7
12	Aghar Ballian	M.S.	6.7	245	$24 \pm 7$	$28 \pm 7$	$357 \pm 111$	$11 \pm 3$	1.9	$\tau$	1.0	0.9
13	Chaurakot	M.S.	6.9	289	$30 \pm 7$	$29 \pm 6$	$289 \pm 106$	$8 \pm 2$	2.4	7	0.9	1.0
14	Charalahkot	M.S.	7.5	432	$29 \pm 6$	$30 \pm 5$	$312 \pm 110$	$10 \pm 2$	2.4	$\overline{7}$	0.9	1.0
15	Garan khalsa	M.S.	7.8	345	$31 \pm 6$	$29 \pm 5$	$368 \pm 104$	$8 \pm 1$	2.5	$\overline{7}$	1.1	1.1
16	Jhajhar kotli	M.S.	6.15	563	$35 \pm 6$	$47 \pm 7$	$367 \pm 118$	$6 \pm 1$	2.8	11	1.1	0.7
17	Chappar	M.S.	6.25	332	$22 \pm 5$	$43 \pm 6$	$370 \pm 105$	$10 \pm 2$	1.8	10	1.1	0.5
18	Churta	L.S.	6.8	234	$32 \pm 6$	$53 \pm 7$	$298 \pm 107$	$4 \pm 1$	2.6	13	0.9	0.6
19	Saranjali	L.S.	7.22	120	$30 \pm 5$	$54 \pm 7$	$326 \pm 136$	$13 \pm 2$	2.4	13	1.0	0.6
20	Badsoo	M.S.	6.66	176	$28 \pm 5$	$48 \pm 6$	$373 \pm 108$	$5 \pm 1$	2.3	12	1.1	0.6
21	Otta	L.S.	6.39	163	$24 \pm 5$	$53 \pm 7$	$409 \pm 110$	$3 \pm 1$	1.9	13	1.2	0.5
22	Garni	M.S.	6.84	164	$34 \pm 6$	$64 \pm 7$	$378 \pm 114$	$15 \pm 4$	2.8	15	1.1	0.5
23	Nagrota rocks	M.S.	7.32	92	$28 \pm 7$	$45 \pm 8$	$322 \pm 98$	$14 \pm 3$	2.3	11	1.0	0.6
Minimum 6.2 92		20	28	281	$\overline{c}$	1.6	$\tau$	0.8	0.5			
Maximum 8.7 563		38	64	409	15	3.1	15	1.2	1.1			
7.2 Average		246	30	43	347	9	2.4	11	1.0	0.7		
S.D.			0.7	131	5	8	36	$\overline{4}$	0.4	$\overline{c}$	0.1	0.1

<span id="page-5-0"></span>Table 2 The values of radionuclide content and their elemental concentration along their physico-chemical parameters in the soil samples of Lower Himalayas of India

S.D. standard deviation, M.S. middle siwalik, L.S. lower siwalik

and maximum value was 92 and 563  $\mu$ S cm<sup>-1</sup>, respectively. The average value of pH was  $7.4 \pm 0.7$  and  $6.8 \pm 0.4$  for Lower and Middle Siwalik, respectively. The average value of EC was  $241 \pm 113 \mu S \text{ cm}^{-1}$  for Lower Siwalik and  $223 \pm 173 \mu S \text{ cm}^{-1}$  for Middle Siwalik, respectively. Both the values of pH and EC are comparable equal for both areas. Moreover, the one sample  $T$  test also indicated that the average value of EC is not statistically difference among different areas with  $T$  Test statistic was 0.5 and a  $p$  value was found to be greater than 0.05 (Fig. [2](#page-6-0)). On the other hand Fig.  $2$  visualized that the  $T$  Test statistic and p value were 2.8 and  $0.02 \lt 0.05$ , indicating that the means of pH was significant different for lower and middle siwalik.

## Primordial and anthropogenic radionuclides

The values of calculated radionuclide contents in the soil samples collected from lower Himalayas of Jammu & Kashmir, India are given in Table 2. The values of primordial radionuclides i.e.,  $^{238}$ U,  $^{40}$ K and  $^{232}$ Th content have been varied from 20 to 38 Bq  $kg^{-1}$ , 281 to 409 Bq  $kg^{-1}$ , and 28 to 64 Bq  $kg^{-1}$ , respectively and the value of anthropogenic radionuclide i.e.,  $137Cs$  varied from

<span id="page-6-0"></span>

Fig. 2 Distribution of pH and EC in two different areas

2 to 15 Bq  $kg^{-1}$ , respectively. The average values of  $A_U$ , A<sub>Th</sub>, A<sub>K</sub>, and A<sub>Cs</sub> were found to be  $30 \pm 5$ ,  $43 \pm 8$ ,  $347 \pm 36$ , and  $9 \pm 4$  Bq kg<sup>-1</sup>, respectively. UNSCEAR [\[1](#page-9-0)] has given the worldwide average value of  $A_{\text{U}}$ ,  $A_{\text{Th}}$ ,  $A_{\text{K}}$ , and  $A_{Cs}$  as 33, 45, 412, and 14.8 Bq  $kg^{-1}$ , respectively. The studied average values of  $A_U$ ,  $A_{Th}$ ,  $A_K$ , and  $A_{Cs}$  were found to be well within the average value suggested by UNSCEAR [\[1](#page-9-0)]. The activity concentration of thorium is higher than the concentration of uranium at all places because this area is situated at limestone [\[13](#page-9-0)] with quartzite at the top. The lower part is dolomite and the upper part is cherty and silicious.

Table  $3$  shows  $137Cs$  activity concentration ranges reported from some of the other locations in the world. Measured values of  $137$ Cs activity concentration in the present work are compared with these values, and it is observed that the measured  $137Cs$  activity concentration range is below the reported mean value from Turkey [[20\]](#page-9-0) and Yugoslavia [\[21](#page-9-0)]. However, values of  $137Cs$  activity concentration measured in this study are found higher than the reported ranges from Pakistan [[22\]](#page-9-0), Greece [\[23](#page-9-0)], Algeria [\[24](#page-9-0)], and Saudi-Jordanian Border [\[25](#page-9-0)]. The measured mean value of  $137Cs$  was found to be comparable with the mean values of Italy [[26\]](#page-9-0), Bangladesh [\[27](#page-9-0)], and Taiwan [[28\]](#page-9-0), respectively.

Figure [3](#page-7-0) shows the distribution of radionuclide contents in lower and middle Siwalik of Jammu & Kashmir, India. The values of radionuclide contents were  $29 \pm 5$  Bq kg<sup>-1</sup> for <sup>238</sup>U content,  $51 \pm 10$  Bq kg<sup>-1</sup> for <sup>232</sup>Th content,  $391 \pm 72$  Bq kg<sup>-1</sup> for <sup>40</sup>K content, and  $7 \pm 4$  Bq kg<sup>-1</sup> for <sup>137</sup>Cs content respectively for lower Siwalik of lower Himalayas. However, the values of radionuclide contents were  $31 \pm 3$  Bq kg<sup>-1</sup> for <sup>238</sup>U content,  $54 \pm 7$  Bq kg<sup>-1</sup> for <sup>232</sup>Th content,  $353 \pm 24$  Bq kg<sup>-1</sup> for <sup>40</sup>K content, and 11  $\pm$  4 Bq kg<sup>-1</sup> for <sup>137</sup>Cs content, respectively for middle Siwalik of lower Himalayas. The values of radionuclide contents in lower and middle Siwalik were found to be comparable with each other due to same lithology and geology, respectively. Moreover, the one sample  $T$  test was used to compare the means of radionuclide content in lower and middle Siwalik (Fig. [3](#page-7-0)). As all the  $p$  values of radionuclide contents except cesium-137 were greater than 0.05 (Fig. [3\)](#page-7-0) then this indicated that the average value of radionuclide contents among lower and middle Siwalik was not statistically significantly difference.

The elemental concentration of primordial radionuclides in collected soil samples has been also calculated and is presented in Table [2](#page-5-0). The elemental concentration of  $^{238}$ U, <sup>40</sup>K, and <sup>232</sup>Th has been ranged from 1.6 to 3.1 mg kg<sup>-1</sup>, 7 to 15 mg  $kg^{-1}$ , and 0.8 to 1.2%, respectively. The average values of elemental concentration were  $2.4 \pm 0.4$ ,  $11 \pm 2$  mg kg<sup>-1</sup>, and  $1.0 \pm 0.1\%$ , respectively. UNSCEAR [[5\]](#page-9-0) has suggested the worldwide average values of elemental concentration as 2.8, 7.4 mg  $kg^{-1}$ , and 1.3% for  $^{238}$ U,  $^{232}$ Th, and  $^{40}$ K, respectively. The average elemental concentration of  $^{238}$ U and  $^{40}$ K was found to be



Table 3 Activity concentrations  $(Bq kg^{-1})$  of  $137Cs$  in soil samples reported from some of the other countries

<span id="page-7-0"></span>

Fig. 3 Distribution of radionuclide contents in lower and middle siwalik areas

well within the average values suggested by UNSCEAR [[5\]](#page-9-0) except the average value of  $^{232}$ Th.

## Correlations

Pearson's coefficient, used as a measure of correlation between radionuclide contents and pH, was found to be 0.3 for <sup>238</sup>U content; 0.02 for <sup>232</sup>Th content; and  $-$  0.15 for  $^{40}$ K content, respectively (Fig. 4). The results of measured Pearson's coefficients implied that the  $^{238}$ U and  $^{232}$ Th content were weak positive correlated with pH and there was no correlation observed between  ${}^{40}$ K and pH [\[2](#page-9-0), [20](#page-9-0)]. Similarly, a  $p$  value of 0.2 was obtained for <sup>238</sup>U content; a p value of 0.9 was observed for  $^{232}$ Th content; and that of 0.6 for the  $^{40}$ K content, respectively. The results of p value indicated that there were no statistically significant correlation between radionuclide contents and pH. Figure 5



Fig. 4 Correlation of radionuclide contents with pH under 2-tailed test



Fig. 5 Correlation of radionuclide contents with EC ( $\mu$ Scm<sup>-1</sup>) under 2-tailed test

visualized the correlation coefficient of radionuclide contents with electrical conductivity. The Pearson correlation was observed  $-0.3$  for <sup>238</sup>U content;  $-0.4$  for <sup>232</sup>Th content; and  $-0.2$  for <sup>40</sup>K content, respectively. The results indicated that all the radionuclide contents were not correlated with the electrical conductivity. Moreover, the p value for all radionuclide contents was greater than 0.05, further illustrated that the correlations were not statistically significant.

# Radiological parameters

The statistics values of measured various radiological parameters such as radium equivalent activity  $Ra_{eq}$ , criteria formula CF, internal  $H_{int}$  and external  $H_{ext}$  hazards effects, gamma index  $I_{\gamma}$ , absorbed dose rate (ADR), annual effective dose equivalent (AEDE), and annual gonald equivalent dose (AGED), respectively are given in Table [4.](#page-8-0) The calculated value of radium equivalent activity ranged from 84 to 154 Bq kg<sup>-1</sup> with an average  $\pm$  S.D. of  $117 \pm 15$  Bq kg<sup>-1</sup>. All observed values have shown Ra<sub>eq</sub> activities lower than the limit set by the Organization for Economic Cooperation and Development [\[17](#page-9-0)] report  $(370 Bq kg<sup>-1</sup>)$  and do not cause any significant health hazards to the inhabitants of the studied area. The values ranged from 0.11 to 0.21 with an average of 0.16  $\pm$  0.02 for criteria formula. The average value (0.16) of the studied samples was found below the recommended maximum value  $(< 1)$ . This indicates that gamma activity in the soil samples does not exceed the proposed criterion level. Table [4](#page-8-0) shows that the mean value of  $0.32 \pm 0.04$  with range 0.23–0.42 of H<sub>ex</sub> and found below the criterion value  $(< 1)$  [\[5](#page-9-0)]. An average value of  $H_{in}$  is determined to be 0.39, which is  $\lt 1$ , indicating that

<span id="page-8-0"></span>Table 4 Measured values of  $Ra_{eq}$ , CF, H<sub>ex</sub>, H<sub>in</sub>, I<sub>y</sub>, absorbed dose rate, annual effective dose, and annual gonald equivalent dose values in studied soil samples collected from Lower Himalayas of Jammu & Kashmir, India

Sr. no.	Name of villages	${\rm Ra}_{\rm eq}$ $(Bq\bar{kg}^{-1})$	$CF \leq 1$	$H_{ex} \leq 1$	$H_{in} \leq 1$	$I_{\gamma} \leq 1$	ADR $(nGyh^{-1})$		AEDE $(\mu Svy^{-1})$		<b>AGED</b>
							$^{238}U,$ $^{232}$ Th, $^{40}$ K	$137C_s$	$238$ U <sub>,</sub> $232$ Th, $40$ K	$\overline{^{137}}\mathrm{Cs}$	$(\mu Svy^{-1})$
$\mathbf{1}$	Aghar Jitto	113	0.15	0.30	0.39	0.41	54	0.09	66	0.11	367
$\overline{\mathbf{c}}$	Laiter	136	0.18	0.37	0.45	0.49	65	0.15	79	0.18	436
3	Pouni-mari	132	0.18	0.36	0.44	0.48	63	0.33	77	0.40	426
4	Pouni-purria	120	0.16	0.32	0.43	0.43	56	0.42	69	0.52	381
5	Bhagha	120	0.16	0.32	0.42	0.44	57	0.36	70	0.44	389
6	Katra	84	0.11	0.22	0.28	0.30	40	0.09	49	0.11	274
7	Dabh jagir	122	0.16	0.33	0.42	0.44	58	0.06	71	0.07	392
8	Dhanoa	106	0.14	0.29	0.36	0.38	50	0.39	62	0.48	341
9	Serli-chamba	117	0.16	0.32	0.39	0.43	56	0.24	68	0.29	376
10	Maghal	132	0.18	0.36	0.43	0.48	62	0.21	77	0.26	420
11	Akhli	94	0.13	0.25	0.31	0.3	56	0.42	68	0.52	312
12	Aghar Ballian	92	0.12	0.25	0.31	0.3	46	0.36	56	0.44	303
13	Chaurakot	94	0.13	0.25	0.33	0.3	44	0.33	54	0.40	305
14	Charalahkot	96	0.13	0.26	0.34	0.4	44	0.24	54	0.29	313
15	Garan khalsa	101	0.14	0.27	0.36	0.4	46	0.30	56	0.37	333
16	Jhajhar kotli	130	0.18	0.35	0.45	0.47	48	0.24	59	0.29	420
17	Chappar	112	0.15	0.30	0.36	0.41	54	0.30	66	0.37	364
18	Churta	131	0.18	0.35	0.44	0.47	62	0.12	76	0.15	414
19	Saranjali	132	0.18	0.36	0.44	0.48	63	0.39	77	0.48	421
20	Badsoo	125	0.17	0.34	0.41	0.46	60	0.15	73	0.18	404
21	Otta	131	0.18	0.35	0.42	0.48	63	0.09	77	0.11	424
22	Garni	155	0.21	0.42	0.51	0.56	73	0.45	90	0.55	491
23	Nagrota rocks	117	0.16	0.32	0.39	0.43	56	0.42	68	0.52	376
Minimum		84	0.11	0.23	0.28	0.31	40	0.06	49	0.07	275
Maximum		154	0.21	0.42	0.51	0.56	73	0.45	90	0.55	491
Average		117	0.16	0.32	0.39	0.42	56	0.26	68	0.32	378
S.D.		15	0.02	0.04	0.05	0.05	8	0.12	9	0.15	45

the internal hazard is below the critical value and it indicates that the soil samples are free from radiation hazards. The calculated range of gamma index from 0.31 to 0.56 with an average  $\pm$  S.D. was 0.42  $\pm$  0.05. Since I<sub>v</sub> values of the studied samples follow the criterion ( $I_{\gamma} \le 1$ ) therefore it may be concluded that the soil samples used in the studied area is safe and does not pose any significant health hazards. The average gamma absorbed doses in soil samples for primordial and anthropogenic radionuclides was  $56 \pm 8$  and  $0.26 \pm 0.12$  nGy h<sup>-1</sup>. 65% gamma dose rate due to primordial radionuclides and all gamma dose rate due to anthropogenic radionuclides were found below the limit recommended by UNSCEAR  $[5]$  $[5]$  i.e., 60 nGy h<sup>-1</sup>. The present value of the annual effective dose due to the primordial radionuclides ranged between 49 and 90 with the mean value of  $68 \pm 9 \mu Sv$  y<sup>-1</sup>; the annual effective dose due to the  $^{137}$ Cs ranged between 0.07 and 0.55 with the mean value of  $0.32 \pm 0.15 \mu Sv$  y<sup>-1</sup> (Table 4). As can be seen from the table, the annual effective dose received due to the exposure to primordial radionuclides is relatively higher than the annual effective dose due to  $137$ Cs. The mean value of 68  $\mu$ Sv y<sup>-1</sup> (due to primordial radionuclides) is found to be less than the worldwide annual effective dose value of 70  $\mu$ Sv y<sup>-1</sup> reported by UNSCEAR [\[5](#page-9-0), [14](#page-9-0), [15](#page-9-0)].

# Conclusions

The estimated average activity concentration of radionuclide contents were found to be lower than the worldwide average values recommended by UNSCEAR [\[5](#page-9-0)]. The values of radionuclide contents and physicochemical parameters in lower and middle Siwalik were found to be comparable with each other due to same lithology and

<span id="page-9-0"></span>geology, respectively. Moreover, the one sample T test also implied that the average values of radionuclide contents among Lower and Middle Siwalik were not statistically significant different except the average values of cesium-137 content and pH. The results of  $p$  value indicated that there were no statistically significant correlation between radionuclide contents among pH and EC. All values of radiological parameters such as radium equivalent content, criteria formula, internal and external health hazards, gamma index, absorbed dose rate and annual effective dose equivalent were found to be well within the limits suggested by various agencies.

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#### Compliance with ethical standards

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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