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Assessment of radiological impacts of natural radionuclides and radon exhalation rate measured in the soil samples of Himalayan foothills of Uttarakhand, India

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Abstract
In order to assess the radiological baseline, the terrestrial radionuclide contents (^{226}Ra, ^{232}Th, and ^{40}K) and radon exhalation rate were measured in the soil samples collected from the vicinity of Himalayan foothills of Uttarakhand, India. Based on the measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, different radiological parameters have been estimated to assess the radiation hazards arising out of the use of these soil samples as a building material in the studied region. The annual effective doses for distinct organs and tissues have also been calculated in order to assess total radiological risk. The overall radiological dose was less than the recommended values of 1 mSv y^{-1}. No significant correlation was observed between ^{226}Ra, ^{232}Th concentration and its exhalation rate in the studied soil samples.

Keywords NaI (TI) gamma detector · Smart RnDuo monitor · Natural radionuclide content · Exhalation rate · Hazard parameter · Air absorbed dose rate

Introduction

Contamination of soil resources with radionuclides is a major health concern in the world. Since soil is composed of various mineral and organic components, it exhibits a wide range of radioactive elements, and their varying concentration level called as NORM's (Naturally Occurring Radionuclide Materials) is mainly dependent on the types of parent rocks of the soil. Most of these radionuclides enter to our soil through dispersion and fall-out process and represent a continuous source entering our body by ingestion/inhalation. These radionuclides irradiate the various organs with alpha, beta particles, and gamma rays. The gamma-emitting radionuclides ^{238}U, ^{232}Th, and ^{40}K are the main contributors to the external exposure to the population in both indoor and outdoor situation [1–5]. The radionuclide ^{226}Ra has greater mobility in the environment. Radon gas is the decay product of ^{226}Ra which generally gets diffused out of the soil, reducing the exposure rate from the ^{238}U series. Some other natural radionuclides (^{235}U series, ^{87}Rb, ^{138}La, ^{147}Sm, and ^{176}Lu) also exist in the environment at low levels so their contributions are very small to the dose in human. Therefore, the assessment of ^{226}Ra, ^{232}Th, and ^{40}K in soil has received special attention and attains a particular interest in the area of radiation protection. The internal and external exposure from these radionuclides is determined in this study.

Gamma radiation notably affects the environment due to the presence of radionuclides in soil and which are used for the assessment of the gamma dose rate. Approximately 87% of the radiation dose received by population is from natural radiation and the remaining is due to anthropogenic radiation [6]. The worldwide value of the average annual effective dose per capita to human is 2.4 mSv [1, 7]. However, much higher levels of radionuclides in the soil become a health hazard for inhabitants. The main sources responsible for high natural background radiation are a high level of ^{238}U and its decay products in soil, rock, and ^{232}Th, which occur in monazites sands and zircons. The local geology and geochemical effects produced enhanced levels of terrestrial radiation in High Background Radiation Areas (HBRA's) [6–8]. In the world, few regions found with very high background radiation due to the deposition of monazite sand and this finding contributes to considerable background exposure in various countries [7–16].

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Many previous studies confirmed that higher level of $^{222}Rn/^{220}Rn$ in the natural environment can significantly increase the risk of lung cancer in the inhabitants [7, 17–20]. Radon/thoron reaches into the indoor environment from soil and building materials through emanation or exhalation process. Radon/thoron exhalation rate from the sub-surface soil depends upon radium (^{226}Ra) content in soil or rock, permeability, porosity, humidity, temperature, and meteorological conditions. Therefore, in the present work exhalation studies for the sediment samples were also taken into consideration for health risk hazard.

In the past decade, some efforts have been made in distinct parts of Uttarakhand along with the main central thrust to quantify the natural radionuclides in soil [21–25]. However, no work has been reported on the viability of these radionuclides in soils of Dehradun and Haridwar district of India. A qualitative study is required for the assessment of radiation hazards in the studied area. In the present study, a systematic analysis has been made to characterize the soil contamination level of natural radionuclides for the assessment of health risk to the resident of the study area. In India, dwellings are mainly constructed using bricks mixed with about 80% of soil [26]. Therefore, the present investigation will also be helpful to check whether the soil of the studied region can be used as building material without posing any health hazard.

Geology of the area

The present study has been performed in the vicinity of the Himalayas foothills of the Uttarakhand, India. The chosen sites come under the Dehradun and Haridwar districts of Uttarakhand (Fig. 1). The Dehradun district is situated in the northwest region of Uttarakhand (29° 58′ N to 31° 2′ N and 77° 34′ E to 78° 18′ E). Dehradun district covers the

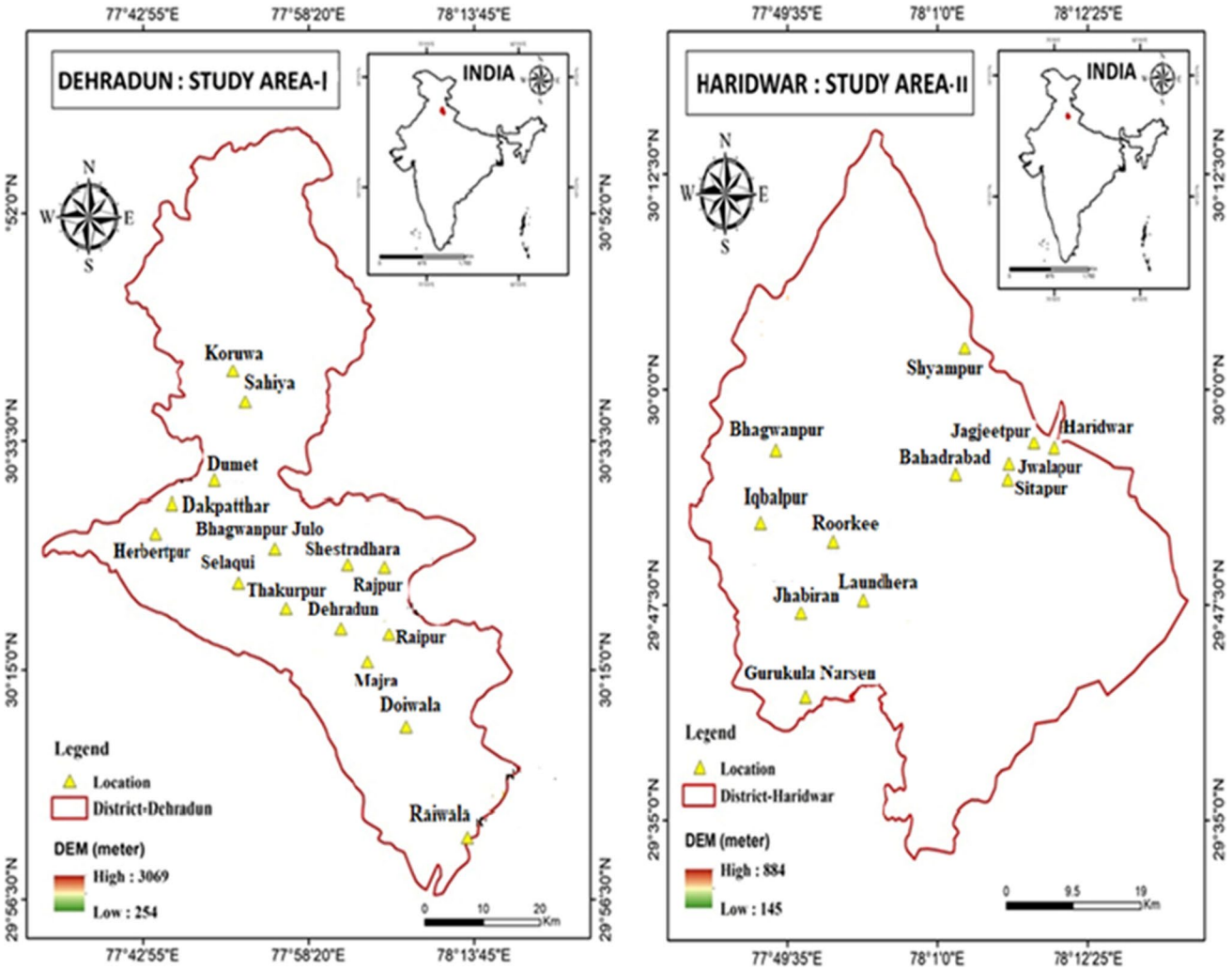

Fig. 1 Sampling sites on the map of the studied region

total area of 3088 km^2 with an average altitude of 640 m above mean sea level (MSL). Dehradun is an integral part of the Doon valley which is about 80 km long and 20 km wide. Physiographically, Doon valley extends from the crest of the Siwalik Hills of Garhwal Himalaya in the south to the Mussoorie range of lesser Himalayas in the north and the Ganga River in the east to Yamuna River in the west. The Jaunsar-Bawar area of Dehradun lies in the North West. The Doon valley is a crescent-shaped, longitudinal and synclinal asymmetric valley, nestled between the main boundary thrust (MBT) in the north, where pre Tertiary rocks from Lesser Himalaya zone nullify the Tertiary rocks of the Siwalik group and Himalayan Frontal Thrust (HFT) or locally known as the Mohand Thrust in south where the rocks of the Siwalik group nullify the alluvial sediments. Whereas, it is bounded by Yamuna tear fault (YTF) in the west and Ganga tear fault (GTF) in the east. The MBT is the major structure that divided the Siwalik and the Lesser Himalaya. The Doon valley and Siwalik range are primarily composed of the rocks of the Siwalik groups. The rocks of the Siwalik groups are divided into the lower (mudstone), the middle (sandstone) and the upper (conglomerate) Siwalik. On the basement of Siwalik rocks, the piedmont fan deposits, locally known as Doon gravels. However, Doon gravels are absent at those places where Siwalik rocks are exposed and increase in thickness towards the south. Based on rock and soil types, the valley has been categorized as the lesser Himalayan belt, Doon valley, and Siwalik belt. The soil of Dehradun is arenaceous in Lesser Himalayan Belt, acidic to near neutral and fine loamy in texture in the boulder belt of the valley and sandy loam on Siwalik Belt.

However, Haridwar district is located at 29° 35′ to 30° 40′ North and 77° 43 ‘ to 78° 22′ East in the southwestern part of Uttarakhand. The geographical area of district Haridwar is 2360 km^2 with a height of 249.7 m above the sea level. Geographically; Haridwar is located between Siwalik Hills in the North and the Ganga basin in the South. Geologically, the area categorized into three zones viz. the Siwaliks Hills, Bhabar and Alluvial Plains (Tarai) from North to South. The Siwalik region is constituted of boulders, pebbles of quartzites, grey micaceous sandstone, and siltstone. Bhabar is formed just below the Siwaliks and characterized by the alluvial fans. A gangetic alluvial plain is further divided into lower piedmont plain, the older and the younger alluvial plains. Lithologically, Tarai is characterized by coarse to fine-grained sand, slit, kankar, gravels, and clays. According to the geological distribution of the area, the soil can also be divided into three types. The important soils are Ultisols (brown hill soil). These soils are highly fertile and occurring in the northern part of the district along Siwaliks. The Entisols soil (Bhabar soil) occurs along the Siwaliks foothills and extends up to Alluvial Plains. Mollisols soil or Tarai soil occurs in the southern part of the district. These soils are mineral soils with less than 25% organic matter.

Experimental work

Soil sampling and pretreatment of samples

Basic criteria concerning the surroundings, meteorological factors, and accessibility, following a grid mapping that covers all the geological structures of the study area, were used for the site selection. To obtain a representative distribution of radionuclides in natural soil, soil samples were collected from aforementioned sites of Dehradun and Haridwar districts of Uttarakhand, India. Natural soil samples were taken from an auger hole at a depth of 0.75 m level from the ground. About 1 × 2 m area was marked at each sampling site and clearing the roots and stones from the ground surface to make the sampling area homogenized. About 1 kg of soil sample was taken from each site in a sealed plastic container. At the laboratory, all the collected samples were pulverized using a pestle and then sieved through a 150 μm mesh size to obtain more homogenized samples. The samples were then dried in an oven of restrained temperature at 110 °C for 2 h to assure that the moisture is thoroughly removed and then were cooled to room temperature. Each sample was weighed, carefully sealed and stored into cylindrical airtight polyethylene containers (6 × 4 cm) for at least 30 days before the measurement to attain a radioactive secular equilibrium between ^{226}Ra and ^{232}Th and their progenies.

Measurement setup for radionuclide content and exhalation rate in soil

The in situ measurement of activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in stored soil samples at the end of 30 days was carried out using γ-ray counts a lead-shielded NaI (Tl) detector. The instrumentations consisted of a large volume NaI (Tl) detectors (63 mm × 63 mm) coupled with a multichannel analyzer (MCA) through pre-amplifier. The relative efficiency calibration of the system was done using International Atomic Energy Agency reference standard materials for ^{226}Ra, ^{232}Th, and ^{40}K, activity measurements: RGU-1, RGTh-1, and RGK-1 respectively. Stabilization of gamma peak was performed with reference to 661 keV energy photo peaks of ^{137}Cs.

Each sample, as well as the background, was counted for 3 h; emitted gamma rays were detected by the detector and observed through a spectrum on the screen. The spectral analysis was performed using the gamma radiation computer software package SPTR-ATC (AT-1315). The estimation of activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K has been done using 1764 keV gamma peaks of ^{214}Bi,

2610 keV emission line of ^{208}Tl and 1460 keV gamma rays respectively. The minimum detectable concentration (MDC) limit of the detector for the radionuclides ^{226}Ra, ^{232}Th and ^{40}K were 3 Bq kg^{-1}, 3 Bq kg^{-1}, and 30 Bq kg^{-1} respectively.

The mass exhalation rate of Radon in soil samples has been measured using smart RnDuo monitor [27]. Each soil samples (400–500 g) were dried before enclosing in a leak-tight stainless steel chamber that coupled to the monitor. The SMART RnDuo monitor detects the emitted alpha particles from radon and its decay products formed inside the ZnS(Ag) based scintillation detector of volume 150 cm^3. ^{222}Rn is sampled into the scintillation cell through a progeny filter and "pinhole plate" eliminating radon progenies and thoron. At the end of every cycle obtained total alpha counts converted into ^{222}Rn activity concentration (Bq m^{-3}) using a smart microprocessor based algorithm. Every sample has been measured for a longer duration (25–30 h) to achieve proper ^{222}Rn growth and saturation. The monitor operates in the calculation range of 8–10 mBq m^{-3} with a sensitivity factor of 1.2 counts h^{-1}Bq m^{-3}.

To calculate the ^{222}Rn mass exhalation rate (J_m), growth data has been fitted in the following equation [28]:

$$C(t) = \frac{J_m M}{\lambda_e V}\left(1-e^{-\lambda_e t}\right) + C_R e^{-\lambda_e t} \quad (1)$$

where $C(t)$ is the radon build up concentration in chamber at given interval (t), C_R is radon concentration (Bq m^{-3}), J_m is the radon mass exhalation rate (Bq kg^{-1}h^{-1}), M is the total mass of the sample (kg), V is the volume of the chamber (m^3), and λ_e is disintegration constant for ^{222}Rn and t is the time for measurement.

In order to measure the ^{220}Rn surface exhalation rate, dried soil samples were kept in the exhalation chamber and scintillation- based smart RnDuo monitor was set in thoron mode to estimate thoron concentration [27]. The exhalation chamber was attached to the scintillation thoron monitor by an airflow pump inlet. Thoron decays before entering into the scintillation detector from soil chamber because of its short half-life (55 s). Therefore the pump was used with flow mode to get all thoron in a scintillation detector.

The ^{220}Rn surface exhalation rate (J_S) (Bq m^{-2} s^{-1}) in soil samples has been given by the following formula [29, 30]:

$$J_S = \frac{C_T V_T \lambda}{A} \quad (2)$$

where C_T is the average concentration of thoron inside the chamber (Bq m^{-3}), V is the residual air volume (m^3), λ is effective decay constant for ^{220}Rn, and A is the surface area of the chamber (m^2).

Theoretical formulation for evaluation of radiological hazard indicators

Contamination of soil due to radionuclides content contributes to various radiological health hazards including hazard index, level indices, and dose rate. These radiological hazards have been described below:

Radium equivalent activity

Natural occurring radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) are not uniformly distributed in the soil. Therefore the radiological hazards arising from the utilization of soil has been assessed through a common index containing different radioactive nuclides activity. The combined radiation exposure due to radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) is estimated in terms of common index i.e. radium equivalent specific activity (Ra_{eq}) in Bq kg^{-1} [31]. It is calculated using the following equation [7]:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (3)$$

where, C_{Ra}, C_{Th}, and C_K is the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K (Bq kg^{-1}) respectively. Equation (3) is based on the assumption that 370 Bq kg^{-1} of ^{226}Ra, 259 Bq kg^{-1} of ^{232}Th and 4810 Bq kg^{-1} of ^{40}K produce the same amount of gamma radiation dose rate [7].

External and internal hazard indices

External (H_{ex}) and internal (H_{in}) radiation hazards due to naturally occurring radionuclides can be calculated through the following equations [32]:

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (4)$$

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \leq 1 \quad (5)$$

External hazard index is used to measure the indoor radiation dose rate due to the external exposure to gamma radiation, assuming that the soil of the study area is used for the construction purpose. However, the internal hazard index assesses the impact of radionuclides (radon and its decay products) on lungs and other respiratory organs. The values of these indices must be less than unity to ensure the safe use of building material for construction purpose of dwellings [33].

Gamma radioactivity level index

Activity level index address the amount of gamma radiation associated with the radionuclides content in soil and other building materials. $I\gamma$ is calculated using the following equation [34]:

$$I_{\gamma} = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \leq 1 \quad (6)$$

The value of I_{γ} should always be less than unity for the building materials and considering the highest recommended dose criterion of 1 mSv yr^{-1} for the inhabitants.

Alpha index

The amount of alpha radiation due to the released radon from building material can be described by alpha index (I_{α}), it can be calculated using Eq. (7) [26, 34]:

$$I_{\alpha} = \frac{C_{Ra}}{200} \leq 1 \quad (7)$$

An alpha index should be less than unity to reflect that ^{226}Ra concentration is less than the maximum recommended value of 200 Bq kg^{-1} [35]. The maximum gamma dose rate from ^{238}U series is delivered by ^{226}Ra series. Therefore, the disequilibrium between ^{238}U and ^{226} Ra does not affect the dose estimation from the measurement of ^{226}Ra.

Air absorbed dose rate

The external absorbed gamma dose rate (nGy h^{-1}) in outdoor air comes from terrestrial radiation. Absorbed dose rate in air at 1 m above the ground can be estimated from the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K by using Eq. (8) [1]:

$$D_A = 0.461C_{Ra} + 0.623C_{Th} + 0.0414C_k \quad (8)$$

where, D_A is the external absorbed gamma dose rate expressed in nGy h^{-1}.

Annual effective dose equivalent

The annual effective dose equivalent received by indoor $AEDE_{In}\left(mSvy^{-1}\right)$ and outdoor $AEDE_{out}\left(mSvy^{-1}\right)$ occupants, due to external exposure to terrestrial radiation was evaluated using conservation factor 0.7 $SvG\,y^{-1}$ with indoor (20%) and outdoor (80%) occupancy factors [1, 7]. The conversion factor is used to convert the absorbed dose rate in air to annual effective dose for indoor and outdoor occupancy. The annual effective dose equivalent is determined from absorbed dose values by following Eqs. (9) and (10) [7]:

For indoor:

$$AEDE_{In}\left(mSvy^{-1}\right) = \mathrm{D}\left(nGyh^{-1}\right) \times 8760h \times 0.7SvGy^{-1} \times 0.8 \times 10^{-6} \quad (9)$$

For outdoor:

$$AEDE_{out}\left(mSvy^{-1}\right) = \mathrm{D}\left(nGyh^{-1}\right) \times 8760h \times 0.7SvGy^{-1} \times 0.2 \times 10^{-6} \quad (10)$$

Doses for different organs and tissues

The annual effective dose rate to various organs and tissues can be calculated from absorbed dose rates using the following equation [36, 37]:

$$D_{organ}(mSv^{-1}) = AEDE\left(mSvy^{-1}\right) \times f \quad (11)$$

where, the conversion factor f is the ratio of the mass absorption coefficient of the organ to air. The annual dose equivalent rate in particular organs or in the whole body has been calculated using the average values of conversion factors of 0.64 for lungs, 0.58 for ovaries, 0.69 for red bone marrow, 0.82 for testes and 0.68 for whole body, respectively [36, 38, 39].

Annual gonadal dose equivalent

The reproductive organs like gonads, active bone marrow, and bone surface cells are considered as organs of interest for estimating dose equivalent [38]. The Annual Gonadal Dose Equivalent for the population of the study area due to the exposure caused by natural radionuclides in the soil was estimated using the Eq. (12) [40]:

$$AGDE = 3.09C_{Ra} + 4.18C_{Th} + 0.314C_K \quad (12)$$

Excess lifetime cancer risk

The possibility of cancer risk associated with radiation exposure from radionuclides by the residents of the study area, who will probably spend the majority or a lifespan in this environment at a particular exposure, can be evaluated using the excess lifetime cancer risk (ELCR). Calculated values of annual effective dose were used to estimate the ELCR using the following relationship [41]:

$$ELCR = AEDE \times DL \times RF \quad (13)$$

where AEDE, DL, and RF are the annual effective dose equivalent, the average duration of life (70 years) and RF (Sv^{-1}) is fatal risk factor per Sievert which is 0.05 for stochastic effects [41–43].

Results and discussion

Natural radionuclides content and exhalation rate in soil

The results of spectrometric analysis of soil samples in terms of specific activity concentration of natural radionuclides ^{226}Ra, ^{232}Th and ^{40}k in the studied area are presented in Table 1. The activity concentration of ^{226}Ra varied between 47 Bq kg^{-1} to 76 Bq kg^{-1} with an average value of 61 Bq kg^{-1} while activity concentration of ^{232}Th ranged from 40 Bq kg^{-1} to 69 Bq kg^{-1} with arithmetic mean of 52 Bq kg^{-1}. The specific activity concentration for ^{40}K varied from 431 Bq kg^{-1} to 549 Bq kg^{-1} with a mean of 506 Bq kg^{-1}. The variations of radionuclides content in different selected sites are presented in Fig. 2. The statistical parameters of radionuclides revealed that ^{226}Ra concentration is equal to or greater than that of ^{232}Th for all soil samples (Table 3). Besides, results showed that the ^{40}K activities in all samples were greater than the activities of ^{226}Ra and ^{232}Th which is normally expected for soil.

The reported world's average activity concentration for ^{226}Ra, ^{232}Th, and ^{40}K are 32 Bq kg^{-1}, 45 Bq kg^{-1} and 420 Bq kg^{-1} in natural soil [7]. It can be seen from the obtained data that the average activity concentration of ^{226}Ra, ^{232}Th, and ^{40}k is elevated from the reported average values. These measured average values agree well with similar studies performed in the Rajpur region along the MBT of the study area [44]. The high activity concentration of natural radionuclides is due to the presence of phyllite with interbedded sandstones and siltstone and further, the presence of quartzite beds in the studied region confirms it. The excess use of potassium-rich fertilizers for agriculture purposes and geological differences may be responsible for the higher concentration of potassium in the studied region.

Further, to check any synergistic behavior, correlations have been drawn between the activity concentrations of

Table 1 The activity concentration of radionuclide (^{226}Ra, ^{232}Th and ^{40}K) in soil

Location	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	Mass exhalation rate (mBq kg^{-1} h^{-1})	Surface exhalation rate (mBq m^{-2} s^{-1})
Shyampur	55 ± 9	54 ± 9	479 ± 62	19	1325
Jwalapur	70 ± 11	62 ± 9	467 ± 59	40	1369
Haridwar	70 ± 11	51 ± 9	507 ± 66	30	1771
Sitapur	76 ± 11	69 ± 10	431 ± 65	30	999
Bahadrabad	50 ± 9	42 ± 8	508 ± 67	60	1717
Roorkee	51 ± 9	43 ± 8	522 ± 63	36	1651
Bhagwanpur (H)	60 ± 10	55 ± 9	484 ± 63	16	836
Iqbalpur	52 ± 9	40 ± 8	486 ± 68	40	1740
Gurukula Narsen	68 ± 11	53 ± 9	527 ± 64	48	1390
Jhabiran	73 ± 11	64 ± 10	485 ± 67	53	1390
Laundhera	69 ± 11	59 ± 9	520 ± 69	60	2064
Koruba	47 ± 9	57 ± 9	538 ± 67	32	1254
Sahiya	57 ± 10	50 ± 9	514 ± 65	41	1633
Dumet	58 ± 10	60 ± 9	502 ± 64	70	2890
Dakpatthar	63 ± 10	53 ± 9	485 ± 65	45	1912
Herbtpur	56 ± 9	49 ± 8	501 ± 66	43	2271
Selaqui	60 ± 10	47 ± 8	512 ± 67	25	1336
Bhagwanpur (D)	56 ± 10	53 ± 9	518 ± 69	32	1782
Dehradun	56 ± 10	45 ± 8	535 ± 66	39	1662
Majra	60 ± 10	49 ± 8	512 ± 65	92	3172
Rajpur	61 ± 10	56 ± 9	497 ± 69	111	1608
Raipur	66 ± 10	48 ± 9	533 ± 70	42	1445
Doiwala	53 ± 9	47 ± 8	546 ± 62	52	1521
Raiwala	54 ± 9	44 ± 8	472 ± 71	28	2130
Shestradhara	76 ± 11	49 ± 9	549 ± 67	106	1912
Thakurpur	59 ± 10	58 ± 9	520 ± 66	56	1735
Jagjeetpur	70 ± 11	50 ± 9	507 ± 66	93	1635

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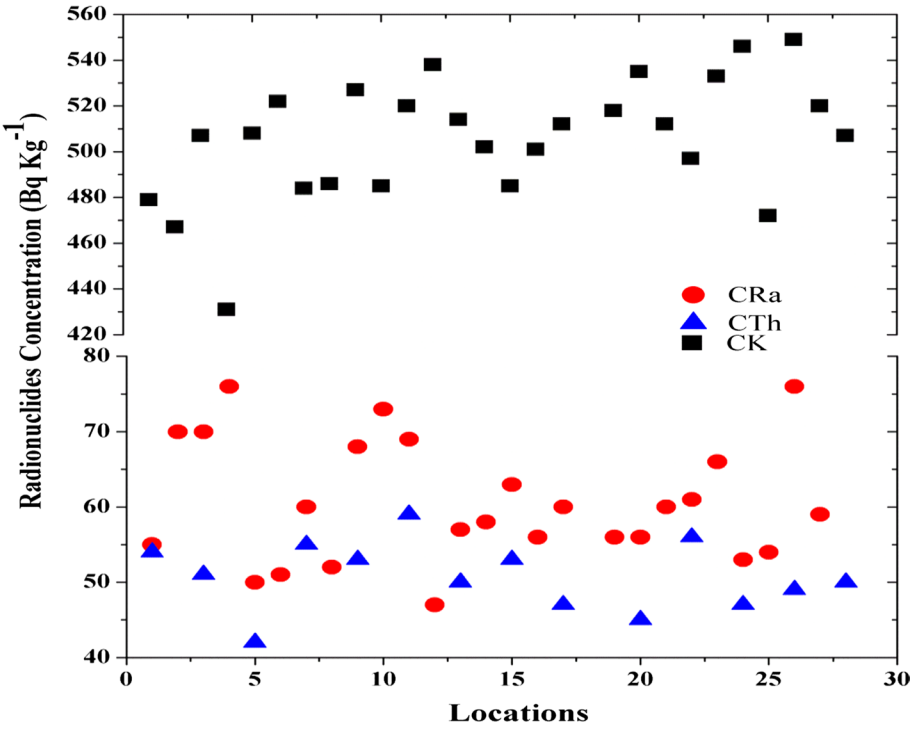

Fig. 2 Variation of radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) activity concentration in soil samples

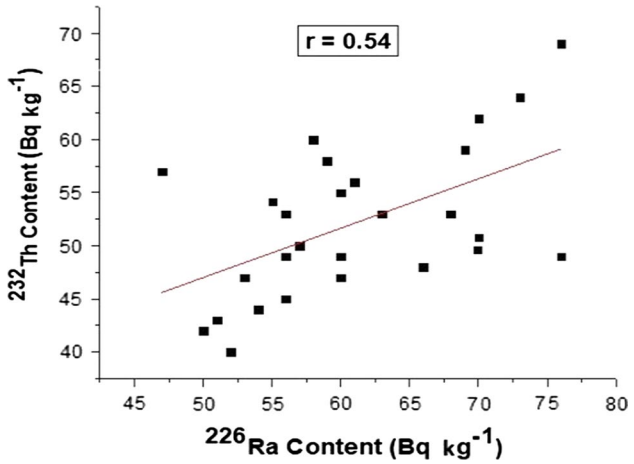

Fig. 3 Correlation between activity concentration of ^{226}Ra and ^{232}Th

^{226}Ra and ^{232}Th (Fig. 3), ^{232}Th and ^{40}K. The correlation coefficient (r) between ^{226}Ra and ^{232}Th was found positive and linear (0.54) whereas no correlation has been observed between ^{232}Th and ^{40}K. The positive correlation strongly indicates that the source of soil contamination might be the same and the radionuclide concentration in pair is a good predictor of the concentration of the other. ^{226}Ra and ^{232}Th come from natural decay series of ^{238}U and ^{232}Th whereas ^{40}K, a naturally occurring radionuclide, is not the part of any such decay series. However, a positive correlation may still be attributed to the property of the soil in retaining these radionuclides under varying weather conditions.

Additionally, a comparison of radionuclides activity concentration in sediment samples with the determined values in neighboring states of India and countries around the world as reported in the literature are shown in Table 2. It has been observed that the average activity concentration of ^{226}Ra in soil samples of the studied province was higher than the reported average values from nearby states; Rajasthan [45], Haryana [19] and Punjab [20]. However, the activity concentration of ^{232}Th falls on the lower side of the reported value from Haryana and Punjab [19, 20] but higher than the reported value from Rajasthan [45]. The activity concentration of ^{40}K was found lower than the reported activity concentration in Haryana and Punjab [19, 20] but equal with the reported mean value of Rajasthan [45]. By comparing the estimated average of activity concentration of ^{226}Ra, ^{232}Th and ^{40}K with other countries, it has been found that estimated data was lower than the reported value from Japan [46] but higher than the reported value from United States [47] and Vietnam [48].

In the present study, Radon exhalation in the soil samples has been also accounted for health hazards. The ^{222}Rn mass and ^{220}Rn surface exhalation rate in a natural soil sample of the examined province has been measured and the results are presented in Table 1. The ^{222}Rn mass and ^{220}Rn surface exhalation rate varied from 15.86 to 110.81 mBq kg^{-1} h^{-1} and from 836.34 to 3172.37 mBq m^{-2} s^{-1}, respectively. The overall average of ^{222}Rn mass and ^{220}Rn surface

exhalation rate for all samples was 47.88 mBq kg^{-1} h^{-1} and 1712.12 mBq m^{-2} s^{-1} respectively (Table 3). The variation of ^{222}Rn mass and ^{220}Rn surface exhalation rate for all soil samples is depicted in Fig. 4.

No significant correlation has been observed between ^{222}Rn mass exhalation and ^{226}Ra concentration in the soil samples of the studied area. A similar result has been observed for the ^{220}Rn surface exhalation with ^{232}Th

Table 2 Comparison of activity concentrations of radionuclides in soil with different areas around the world

Locations	Radioactivity in soil			References
	^{226}Ra (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})	
Japan	320	200	1100	[46]
United States	40	35	370	[47]
Iran	45.4	57.1	768.5	[50]
Vietnam	42.7	59.8	411.9	[48]
Italy	25.1	28.1	809.8	[51]
China	33.1	21.8	833.3	[52]
Pakistan	25.8	49.2	561.6	[53]
Rajasthan	24	55	549	[45]
Haryana	63	78	630	[19]
Punjab	46	98	756	[20]
Uttarakhand (Dehradun and Haridwar)	76	69	549	Present study

Table 3 Statistical data of soil samples for activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, and hazard indices

	C_{Ra}	C_{Th}	C_K	Mass exhalation rate (mBq kg^{-1} h^{-1})	Surface exhalation rate (mBq m^{-2} s^{-1})	Ra_{eq} (Bq kg^{-1})	Hex ≤ 1	Hin ≤ 1	Iα ≤ 1	Iγ ≤ 1
Minimum	47	40	431	15.86	836.34	147	0.40	0.54	0.23	0.54
Maximum	76	69	549	110.81	3172.37	207	0.56	0.76	0.38	0.74
A.M.	61	52	506	47.88	1712.12	174	0.47	0.63	0.30	0.63
G.M.	60	52	505	42.96	1646.19	173.36	0.47	0.63	0.30	0.63
Median	59	51	510	41.56	1656.4	173.56	0.47	0.63	0.30	0.63
S.D.	8	7	27	24.17	508.08	15.58	0.04	0.06	0.04	0.05

A.M. Arithmetic Average, *G.M.* Geometric Average, *S.D.* Standard Deviation

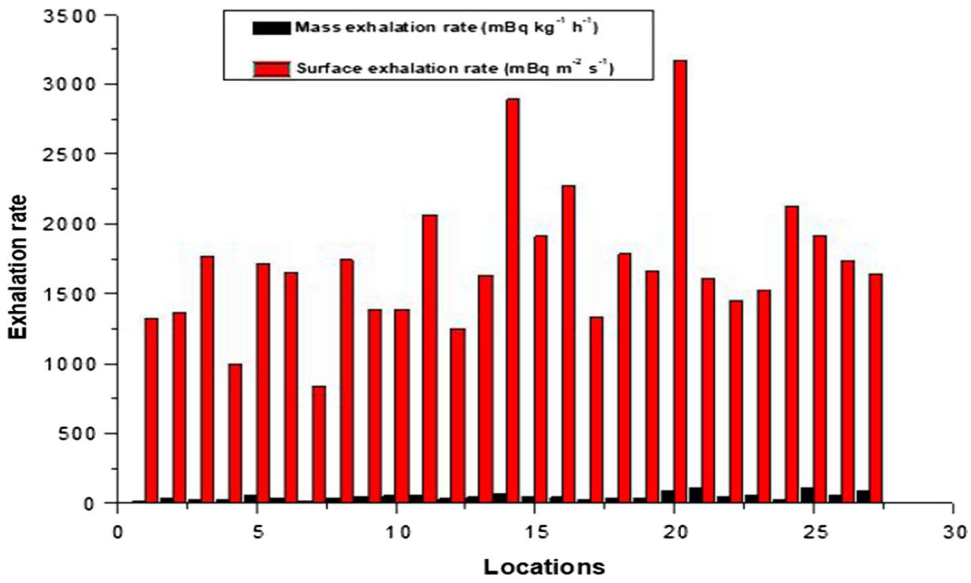

Fig. 4 Variation of ^{222}Rn mass and ^{220}Rn surface exhalation rate for all soil samples

concentration in the studied soil samples. It can be noted from some previous studies; both positive and negative correlations are possible due to the difference in soil quality, chemical and geological properties of the radionuclides [21–23, 49].

Radiological hazard indices

The statistical values of the calculated radiological health hazards parameters such as radium equivalent activity (Ra_{eq}), external (H_{ex}) and internal (H_{in}) hazard indices, gamma radioactivity level index ($I\gamma$), alpha Index ($I\alpha$), absorbed Dose Rate (D_A), annual effective dose equivalent ($AEDE$.), doses for different organs and tissues (D_{organ}), annual gonadal dose equivalent ($AGDE$), excess lifetime cancer risk ($ELCR$) respectively are given in Table 3, 4 and 5. Radium equivalent activity represents the radiation threats associated with the activities of ^{226}Ra, ^{232}Th, and ^{40}K and the suitability of the soil for agriculture and construction purpose. In the present work, Radium equivalent activity varied from 147 to 207 Bq kg^{-1} with an arithmetic mean of 174 Bq kg^{-1}. Ra_{eq} for all samples were lower than the threshold limit of 370 Bq kg^{-1} prescribed by the Organization for Economic Cooperation and Development (OECD) [31]. Hence, it can be concluded that the soil of the studied area with respect to Ra_{eq} activities do not pose any significant health hazards to the inhabitants. The external hazard index for all the soil samples were found within the safe limit of 1 and have been reported in Table 3. Additionally, internal exposure because of radon and its progeny is also hazardous to the respiratory system. Therefore, the internal hazard index is also calculated to assess internal exposure. The calculated values of Hex and Hin for all the soil samples is varied from 0.40 to 0.56 with an average value of 0.47 and from 0.54 to 0.76 with an average value of 0.63, respectively. The average values of both indices are well below the limiting value, indicating that the radiation hazard in the soil samples of the studied region is insignificant.

Table 5 Statistical values of annual effective dose rate to different body organs and tissues

	Effective dose equivalent (mSv y^{-1})				
	Lungs	Ovaries	Red bone marrow	Testes	Whole body
Minimum	0.22	0.20	0.23	0.28	0.23
Maximum	0.30	0.27	0.32	0.38	0.32
A.M.	0.26	0.23	0.28	0.33	0.27
G.M.	0.25	0.23	0.27	0.33	0.27
Median	0.25	0.23	0.27	0.33	0.27
S.D.	0.02	0.02	0.02	0.03	0.02

Further, the gamma index is also calculated and varied from 0.54 to 0.74 in the studied region. It has been observed that, $I\gamma \leq 1$ for all soil samples (Table 3). Alpha index in the studied soils samples varied from 0.23 to 0.38 with an average value of 0.30 as tabulated in Table 3. The values of the alpha index for all samples were less than unity, which means that the soils are safe from the radiation hazards. Hence, it confirms that the soil of this region is suitable for use as construction material in dwellings.

Radionuclides are the main contributor to gamma radiation in the natural environment. The absorbed dose rate in the air at a level of 1 m from the ground has been estimated using the conversion factors prescribed by UNSCEAR 2000 [7]. The total absorbed dose rates in the studied area varied from 69 to 96 nGy h^{-1}, with an average of 81 nGy h^{-1} (Table 4). The average absorbed dose rate was lower than the reported value of 90 and 86 nGy h^{-1} for Indian and global, respectively [7]. The recorded value of the absorbed dose rate for all samples was below the safe limit except one, which is important from health hazard effects point of view to the inhabitants. The annual indoor effective gamma dose rates were varied in the range of 0.34 to 0.47 mSvy^{-1}, with an average of 0.40, while the annual outdoor effective gamma dose was in the range of 0.08 to 0.12 mSv y^{-1} with

Table 4 Statistical values of gamma radiation dose and excess lung cancer risk

	Air absorbed dose rate (nGy h^{-1})	Effective dose equivalent (mSv y^{-1})			AGDE (μSv y^{-1})	ELCR
		Indoor	Outdoor	Total		
Minimum	69	0.34	0.08	0.42	481.19	0.28
Maximum	96	0.47	0.12	0.59	656.23	0.42
A.M.	81	0.40	0.10	0.50	565.12	0.35
G.M.	81	0.40	0.10	0.50	563.36	0.35
Median	81	0.40	0.10	0.50	566.08	0.35
S.D.	7	0.03	0.01	0.04	45.28	0.03
The adapted global average value	86 nGy h^{-1} (UNSCEAR 2008)	0.46 mSv y^{-1} ICRP (1990)	0.06 mSv y^{-1} ICRP (1990)	0.52 mSv y^{-1} ICRP (1990)	300 μSv y^{-1} (UNSCEAR 2000)	0.29×10^{-3} (UNSCEAR 2000).

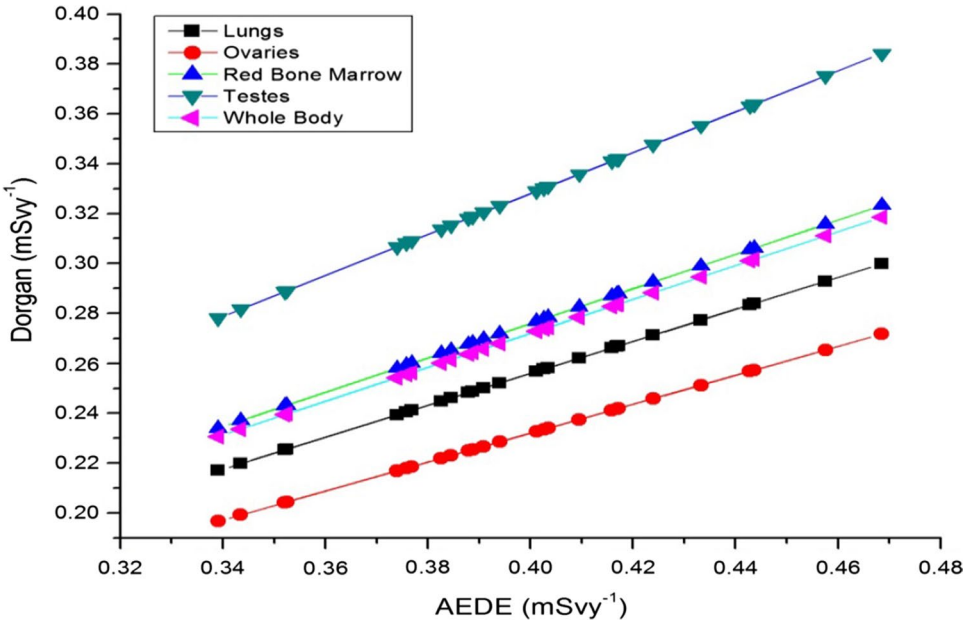

Fig. 5 Variation of D_{organ} with annual effective doses equivalent (AEDE)

an average of 0.10 (Table 4). The average annual indoor dose was below the permissible level of 0.46 mSv y^{-1} while average annual outdoor dose was relatively higher than the advocated level of 0.07 mSv y^{-1} [7]. These results are very useful from the radiation protection point of view.

The statistical variation of D_{organ} has been tabulated in Table 5. The variation of D_{organ} and corresponding absorbed dose is shown in Fig. (5). From Table 4, it can be clearly seen that the obtained results for the annual gonadal dose equivalent (μSv y^{-1}) at 1 m above the ground due to ^{232}Th, ^{238}U and ^{40}K has crossed the permissible limit of 300 μSv y^{-1} for all soil samples [7]. This implies that the gonadal values may pose some threat to the reproductive organs of the residents in the dwellings of the study area.

The calculated values of *ELCR* for outdoor exposure in the investigate area is given in Table 4, ranged from 0.28×10^{-3} to 0.42×10^{-3} with an average value of 0.35×10^{-3}. The average value of *ELCR* is found higher than the world's average of 0.29×10^{-3} based on the annual dose limit of 1 mSv for general civic [7].

Conclusions

The present study showed the natural radionuclide concentration (average value of ^{226}Ra, ^{232}Th and ^{40}K being 61 Bq kg^{-1}, 52 Bq kg^{-1} and 506 Bq kg^{-1}), radon and thoron exhalation rates as well as the radiological impact on various organs in human body from the soil of Haridwar and Dehradun district of Uttarakhand, India. The measurements of natural radioactivity in soil samples show that the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K is considerably greater than the worldwide average values. The higher ^{226}Ra concentration than ^{232}Th for all soil samples indicates the presence of the radium rich soil in the investigated area. No significant correlation has been observed between ^{222}Rn exhalation rate and ^{226}Ra concentration as well as between ^{232}Th concentration and ^{220}Rn exhalation rate. Based on the determined activity concentrations, various radiation hazards parameters, doses, and health risks were assessed. By comparing the estimated values of hazard parameter with the world averages, it can be said that this region is safe with respect to gamma radiation level, despite the higher activity of radionuclides in soil. The results of this survey suggest that the soil of the study area as building materials would not pose any significant health hazards to the inhabitants in the dwellings. The similar studies should be conducted periodically to measure the natural radiation in terms of monitoring the region for inhabitant's health. This study provides a base data for the evaluation of the further radiation hazard and can be adopted in a future study on natural radioactivity mapping.

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Authors contribution All authors contributed equally in the preparation of this manuscript.

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