Vertical distribution and radiological risk assessment of natural radionuclides in the alluvial soil profile of south-west Punjab, India

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Abstract
Natural radionuclide levels are studied in alluvial sediments up to the depth of 900 cm. Eighteen profiles are selected from agricultural and undisturbed areas and analysed by gamma-ray spectrophotometer. Levels were found to vary significantly with depth and higher concentration is observed in agricultural samples till the depth of 600 cm and values are comparable after this depth, indicating role of anthropogenic activities on radionuclide enrichment in agricultural areas. However, value is higher than global average at both sites, indicating their geogenic presence. Elemental ratio and multi-statistical analysis are utilized to assess the behaviour of radionuclides involved. Radiological hazard risk assessment is also carried out.

Keywords Natural radionuclides · Soil profile · Radiological risk assessment · Geogenic and Anthropogenic contaminants · South-west Punjab

Introduction
Radioactivity either natural or man-made is ubiquitous in the Earth’s crust though in different amount. Human beings are continuously exposed to natural radiation from air, food, ground, building materials and the universe [1]. Natural radionuclides in the soil form significant component of radioactivity and originates mainly from the uranium (238U) - thorium (232Th) decay series and potassium (40K). This component of natural radioactivity depends primarily on mineralogy of the bedrock and also secondary products resulting from weathering [2]. The change in distribution patterns of 238U, 232Th and 40K in the soil involves process of soil formation, weathering and various chemical and biological interactions that influence their levels [3].

Mineral content of the sediment and rocks is closely related to the uranium, thorium and potassium content. Th/K elemental ratio is utilized to know about the mineral assemblage of sediments (light or heavy minerals). When source rocks disintegrate, potassium is rich in the light or rock forming minerals (feldspars and quartz) whereas most of the uranium and thorium remain associated with the dark or heavy mineral assemblage, that is in accessory minerals. Radionuclides are incorporated in the crystal lattice of minerals during their formation and are transported as such in various heavy minerals [4].

Uranium and thorium migrate under the surface soil conditions and the ability of hydrogenous migration is highest for uranium as it remains soluble for long time and also gets migrated along with the water to long distances [5]. Due to the contrasting behaviour of U and Th, elemental ratio of eTh/eU is used as indicator of the element oxidation or reduction in the sediments. Thorium exists in its insoluble tetravalent form, unaffected by the changing redox conditions and is geochemically associated with uranium. Uranium on the other hand occurs in its insoluble tetravalent form under reducing conditions in the sediments and as soluble hexavalent form in oxidising conditions where it gets mobilized in the solution [6]. Due to the high biological mobility, U-series nuclides have been of particular interest always. Radionuclides with significant mobility are considered as a special group of contaminants in the terrestrial media as they accumulate in the media and enter the food chain [7]. Fast downward moving radionuclides can reach down to the water level contaminating the groundwater.
resources. Thus understanding radionuclide distribution and migration is important part of environmental studies [8].

Distribution of radionuclides in soil also depend on the soil texture, physico-chemical properties of radionuclides, irrigation of land and fertilizer application [9]. Alluvial soil of Punjab is nitrogen deficient and the deficient nutrients are supplied by the common practice of chemical fertilizers application. Radionuclide content is present naturally in the phosphatic fertilisers, derived mainly from the phosophatic rocks [9–11]. Thus cultivation contaminates the agricultural land by inducing large concentration of natural radionuclides present in phosphate fertilizers. Most commonly used compounds in the study region are urea, di ammonium phosphate (DAP), murate of potash (MAP) and complex and single super phosphate (SSP) [12]. The activity concentration is much larger in the MAP fertilizer [13]. Possibility of change in radionuclide levels with land-use and soil heterogeneity is considered in the present study. Thus comparison have been made of agricultural sites with the undisturbed (uncultivated) areas. In recent years, many studies have been carried out to study the behaviour of natural radionuclides in surface soil [14–20]. However, studies are quite scarce on the vertical distribution of natural radionuclides. The study region of Punjab is of interest because of the high uranium levels detected in the aquifer system [21–23]. To better understand uranium distribution patterns in the groundwater of the region, it is highly important to look at the vertical distribution of radionuclides in the soil.

Main objective of the current study is the investigation of vertical distribution of natural radionuclide levels ($^{238}$U, $^{232}$Th and $^{40}$K) from agricultural and undisturbed areas of Punjab region. Radiological hazard risk levels has been estimated from the obtained data and are presented. An attempt has been made to understand the leaching and migration of radionuclides by utilizing elemental ratios of nuclides. The study is expected to provide the background data on natural radioactive isotopes for monitoring possible environmental radioactivity pollution in the future.

**Materials and methods**

**Study area**

The area selected for investigation is a part of south-west Punjab where high uranium levels are reported in the groundwater [21–23] (Fig. 1). Geologically, it is a part of Indo-Gangetic plain, formed by river sediments through weathering and erosion of igneous or metamorphic rocks from Himalayas. The river laid sediments has developed into the alluvial soil. The alluvium is mainly composed of sands of various grades, clays and silts and is heterogeneous in nature in accordance to the mode of deposition by constantly shifting river [24]. The area is intensely developed in agriculture, and is affecting the water and soil quality from quite long time.

**Soil sample collection for radionuclide measurement**

Soil samples were collected from soil cores of eighteen profiles by using a manually oriented hand auger. Auger is attached to the extension rod and lowered down the bore-hole by applying pressure. Auger was rotated to obtain the core of material and after reaching the desired depth, auger was carefully removed from the boring. Auger bucket and rod were cleaned to remove any accumulated soil and steps were repeated for sample collection from deeper depth. PVC (polyvinyl chloride) pipes were used for sampling at higher depth to avoid cross-contamination and any spillage from the upper layers. Sampling depth covered was limited to the depth at which sand began to flow. There are thirteen profiles selected from the agricultural soil (A1-13) and five from the undisturbed soil (U1-5), where there is no agricultural, industrial or urban land use. All the soil cores were drilled down to the depth of 900 cm and sample was collected from each core after an interval of 50 cm, resulting in a total of 300 samples. All the cores reached the depth of 900 cm except three due to the difficulty faced in field during drilling. Samples were packed in the polyethylene bags and coordinates were noted down by using the global positioning system. Samples were carried to the laboratory and dried in the oven at 60 °C for a day or two depending on their moisture content till all the moisture was driven out. Samples were sieved through 2 mm sieve and the texture was noted down.

For radionuclide measurement, each profile was divided into 3 sections on the basis of variation in soil texture, resulting in selection of 51 samples for analysis from all the cores. Sample are labeled as AiDh or UiDh where i refers to the number of soil profile and h (1–3) refers to the corresponding depth interval, 1 is for the depth range of 1–300 cm, 2 for 300–600 cm and 3 for 600–900 cm. A and U denotes the agricultural and undisturbed areas respectively. The depth interval is decided on the basis of texture variation. For the radionuclide measurement, known weights of samples were packed in Petri plates of 75 mm diameter. Containers were sealed tightly and kept aside for more than 30 days so as to attain the secular equilibrium between the parent radionuclides and their daughter products. $^{238}$U, $^{232}$Th and $^{40}$K determination was carried out by using gamma spectrometry at Saha Institute of Nuclear Physics in Kolkata, India. Activity concentration has been noted down in terms of Bq kg$^{-1}$.
Instrumentation or calibration and energy lines

All the samples were measured in Canberra model high purity germanium detector (HPGe) with 80% relative efficiency and resolution of 1.65 keV at 1.33 MeV energy. The detector was shielded from background radiation with cylindrical, laboratory lead shield series and was connected with DSA-LX analyser. Energy calibration was performed using $^{60}$Co, $^{133}$Ba and $^{137}$Cs single point sources. Each sample was counted for 60,000 s which was found optimum in the 80% HPGe detector [25, 26]. A Petri-plate filled with SiO$_2$ was also counted for 60,000 s, which was considered as background spectrum.

The background spectrum was stripped from the spectrum of each sample. $^{238}$U and $^{232}$Th standards were also prepared from the IAEA RGU-1 (uranium ore) and RGTh-1 (thorium ore) as described in Naskar et al. [26] kept aside to obtain secular equilibrium and were also subjected to measure in 80% HPGe. For measurement of $^{40}$K, array of KCl standards were prepared [27]. Gamma energy transition of 295.1, 351.9 keV of $^{214}$Pb and 609.1 keV of $^{214}$Bi for $^{238}$U activity measurement, 338.3, 911.2 keV of $^{228}$Ac and 583.2 keV of $^{208}$Tl for $^{232}$Th [28] and 1460.8 keV for $^{40}$K were used.

Activities of $^{238}$U, $^{232}$Th and $^{40}$K has been calculated in the measured samples using comparator method [29].

Radiological hazard risk assessment

Absorbed dose rate in air (D)

By the use of measured radionuclide activity, absorbed gamma ray dose rate has been calculated in the outdoor air at height of 1 m using the conversion factors as per following equation [30]. It is assumed while calculation that contribution to the total dose rate from other radionuclides is insignificant such as $^{137}$Cs, $^{235}$U, $^{87}$Rb, $^{85}$Sr, $^{138}$La, $^{147}$Sm and $^{176}$Lu.

$$ D(nGy\ h^{-1}) = [0.462A_{Ra} + 0.621A_{Th} + 0.0417A_{K}] $$

where $A_{Ra}$, $A_{Th}$ and $A_{K}$ represent the activity concentration of $^{238}$U, $^{232}$Th and $^{40}$K in Bq kg$^{-1}$ respectively.
Annual effective dose equivalent (AEDE)

For conversion of absorbed dose rate to annual dose rate, conversion factor of 0.7 Sv G y\(^{-1}\) is applied. Considering 5 h of average time spent by people outdoor, 5/24 (0.2) is taken as outdoor occupancy factor (20%) and calculation is done using the following equation [31]:

\[
\text{AEDE - Outdoor}\ (\mu\text{Sv y}^{-1}) = D \times 8760 \times 0.7 \times 0.2 \times 10^{-3}
\]

Radium equivalent activity (Ra\(_{eq}\))

It is one of the widely used hazard index to describe the gamma output from samples with different amount of \(^{238}\text{U}, \ ^{232}\text{Th}\) and \(^{40}\text{K}\). Radium equivalent activity can be calculated by using the activity concentration of uranium (\(A_{\text{Ra}}\)), thorium (\(A_{\text{Th}}\)) and potassium (\(A_{\text{K}}\)) in Bq kg\(^{-1}\).

\[
\text{Ra}_{eq}\ (\text{Bq kg}^{-1}) = A_{\text{Ra}} + 1.43A_{\text{Th}} + 0.077A_{\text{K}}
\]

Hazard indices (\(H_{\text{in}}\) and \(H_{\text{ex}}\))

External hazard index (\(H_{\text{ex}}\)) gives the radiological suitability of material used in construction i.e. radiation dose given externally by the material used in construction [30]. Internal hazard index (\(H_{\text{in}}\)) is calculated to determine the internal exposure to gamma rays due to the short lived gaseous decay product of \(^{226}\text{Ra}\) i.e. \(^{222}\text{Rn}\) which is carcinogenic and is harmful to the respiratory organs [30].

The equations used for the calculation are given below:

\[
H_{\text{ex}} = \frac{A_{\text{Ra}}}{370} + \frac{A_{\text{Th}}}{259} + \frac{A_{\text{K}}}{4810}
\]

\[
H_{\text{in}} = \frac{A_{\text{Ra}}}{185} + \frac{A_{\text{Th}}}{259} + \frac{A_{\text{K}}}{4810}
\]

Here \(A_{\text{Ra}}\), \(A_{\text{Th}}\), and \(A_{\text{K}}\) represent the activity concentration of uranium, thorium and potassium respectively in Bq kg\(^{-1}\).

Annual gonadal dose equivalent (AGDE)

The gonads (reproductive organs), bone marrow cells and bone surface cells are actively reproducing cells and are more sensitive to radiation than the dormant cells. Thus AGDE due to the specific activity of \(^{238}\text{U}, ^{232}\text{Th}\) and \(^{40}\text{K}\) measures the yearly dose equivalent to reproductive organs of population receiving radiation. It can be calculated as described below [32]:

\[
\text{AGDE} (\text{mSv y}^{-1}) = 3.09A_{\text{Ra}} + 4.18A_{\text{Th}} + 0.314A_{\text{K}}
\]

Elemental ratios

Elemental ratios of eU/eTh, eU/K, eTh/K were calculated to visualise the relative concentration of the radioelements. Elemental activity concentrations are obtained for U, Th and K by using conversion factors as 1 ppm of \(^{238}\text{U}\) by weight is 12.35 Bq kg\(^{-1}\), 1 ppm of \(^{232}\text{Th}\) is 4.06 Bq kg\(^{-1}\) and 1% of K is 313 Bq kg\(^{-1}\) [33]. These give us an indication that whether the concerned radioisotope is enriched or depleted in the studied samples [34].

Statistical analysis

Descriptive statistical analysis was carried out to get single descriptive value for the large data set which makes it easy to look at the entire data set. Correlation analysis was applied to the chemical data to understand the strength of association and mutual relationships between the variables. All the statistical data processing was carried out with the help of SAS software.

Results and discussion

Distribution of radionuclides

Radionuclide levels of \(^{238}\text{U}, ^{232}\text{Th}\) and \(^{40}\text{K}\) for samples have been presented in Tables 1, 2 and Figs. 2, 3. Overall 51 samples were determined for radionuclides, with three samples selected from each profile. Only top samples were selected in the first 300 cm for differentiating the concentration in agricultural and undisturbed areas. However, samples were selected from different depth after this. Natural radionuclide presence is observed in both the agricultural and undisturbed soil samples. It is observed that the distribution is not homogenous throughout the region. From Table 2 and Fig. 2, we see that higher value of radionuclides is observed in the agricultural sites than the undisturbed areas, however, variation is observed only up to the depth interval of 600 cm. The values are comparable after this and are almost similar for D3, 600–900 cm depth. Reported higher levels of radionuclide in the agricultural land indicate role of anthropogenic activities on radionuclide accumulation particularly use of fertilizers. It is seen that in the top soil concentration of \(^{40}\text{K}\) is relatively higher than the other radionuclides, with an average of 718.50 and 580.38 for agricultural and undisturbed areas respectively (Table 2), representing largest contribution by \(^{40}\text{K}\) to the total activity of soil. This is due to the large amount of NPK fertilizers used. The continuous addition of chemicals have increased the original sufficient levels of \(^{40}\text{K}\) in the agricultural sediment samples (Figs. 2, 3).
Table 1 Sampling coordinates and activity concentration of radionuclides ($^{238}$U, $^{232}$Th, $^{40}$K)

| ID   | Latitude | Longitude | $^{238}$U Bq kg$^{-1}$ | $^{232}$Th Bq kg$^{-1}$ | $^{40}$K Bq kg$^{-1}$ |
|------|----------|-----------|------------------------|------------------------|------------------------|
| A1D1 | 30.53    | 75.64     | 44.0 ± 1.49            | 58.9 ± 3.85            | 785.6 ± 11.71          |
| A1D2 | 30.51    | 75.64     | 42.2 ± 1.63            | 59.3 ± 4.01            | 566.7 ± 10.09          |
| A1D3 | 30.54    | 75.64     | 39.8 ± 0.88            | 57.2 ± 1.44            | 637.3 ± 10.52          |
| A2D1 | 30.53    | 75.64     | 41.0 ± 1.02            | 55.1 ± 1.49            | 702.9 ± 11.02          |
| A2D2 | 30.54    | 75.64     | 37.5 ± 0.90            | 44.4 ± 2.29            | 655.4 ± 10.75          |
| A2D3 | 30.55    | 75.64     | 39.8 ± 0.70            | 57.2 ± 4.01            | 566.7 ± 10.09          |
| A3D1 | 30.47    | 75.44     | 43.9 ± 3.13            | 72.5 ± 1.42            | 781.1 ± 12.03          |
| A3D2 | 30.54    | 75.64     | 39.5 ± 1.77            | 62.7 ± 4.40            | 560.6 ± 10.15          |
| A3D3 | 30.51    | 75.54     | 41.6 ± 1.78            | 63.4 ± 3.22            | 673.0 ± 10.97          |
| A3D4 | 30.48    | 75.34     | 49.5 ± 1.57            | 60.5 ± 3.80            | 812.2 ± 12.02          |
| A4D1 | 30.49    | 75.54     | 47.9 ± 2.30            | 70.1 ± 3.12            | 777.7 ± 11.67          |
| A4D2 | 30.52    | 75.53     | 45.2 ± 3.13            | 72.5 ± 1.42            | 781.1 ± 12.03          |
| A4D3 | 30.55    | 75.57     | 47.6 ± 2.01            | 73.9 ± 5.89            | 684.4 ± 11.09          |
| A5D1 | 30.46    | 75.54     | 34.9 ± 0.79            | 50.5 ± 2.94            | 756.0 ± 11.64          |
| A5D2 | 30.51    | 75.59     | 33.0 ± 1.97            | 48.8 ± 4.03            | 529.5 ± 9.85           |
| A5D3 | 30.48    | 75.34     | 39.5 ± 1.77            | 62.7 ± 4.40            | 560.6 ± 10.15          |
| A6D1 | 30.54    | 75.54     | 41.6 ± 1.78            | 63.4 ± 3.22            | 673.0 ± 10.97          |
| A6D2 | 30.49    | 75.54     | 47.9 ± 2.30            | 70.1 ± 3.12            | 777.7 ± 11.67          |
| A6D3 | 30.52    | 75.53     | 45.2 ± 3.13            | 72.5 ± 1.42            | 781.1 ± 12.03          |
| A7D1 | 30.47    | 75.44     | 43.9 ± 3.13            | 72.5 ± 1.42            | 781.1 ± 12.03          |
| A7D2 | 30.54    | 75.54     | 41.6 ± 1.78            | 63.4 ± 3.22            | 673.0 ± 10.97          |
| A7D3 | 30.51    | 75.59     | 39.5 ± 1.77            | 62.7 ± 4.40            | 560.6 ± 10.15          |
| A8D1 | 30.49    | 75.54     | 41.6 ± 1.78            | 63.4 ± 3.22            | 673.0 ± 10.97          |
| A8D2 | 30.54    | 75.54     | 43.9 ± 3.13            | 72.5 ± 1.42            | 781.1 ± 12.03          |
| A8D3 | 30.51    | 75.59     | 39.5 ± 1.77            | 62.7 ± 4.40            | 560.6 ± 10.15          |

A. Agricultural land; U, Undisturbed areas; D1, 1–300 cm depth; D2, 300–600 cm depth; D3, 600–900 cm depth
There is significant variation of radionuclide levels with depth (Figs. 2, 3; Table 2). Radionuclide content in the soil samples is affected by the number of factors and processes affecting its variability [35]. Variation in concentration with depth in the same profile is due to the different physical, chemical and geochemical properties of the sediments [36]. The soil texture is expected to control the distribution of natural radionuclides [37]. In the study area, clay percentage is found to be higher in the sediments at top and decreases with increasing depth. Radionuclide levels are higher in the finer fraction (clay sediments) due to adsorption of radionuclides in the crystal lattice or on the grain boundaries [38].

Thus high radionuclide concentration points towards more clay content and lower at depth is due to higher sand content. However, exceptionally high concentration of radionuclides at few of the sites does not correlate with the soil texture and marks the uranium accumulation zones in the soil.

It is seen that $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ concentration is higher than that of $^{238}\text{U}$ at all the sites (Fig. 3). $^{40}\text{K}$ value is higher because of its chemical composition in clay minerals and thorium by adsorption to clay sediments [39]. Obtained levels from the study area were compared with the data collected by various authors from Punjab, different parts of India and with

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**Table 2** Comparison of mean activity concentration (Bq kg$^{-1}$) of radionuclides in soil with those from similar investigations in other areas

|                | $^{238}\text{U}$ | $^{232}\text{Th}$ | $^{40}\text{K}$ | References |
|----------------|------------------|------------------|----------------|-----------|
| Jharkhand, India | 53.8             | 44.2             | 464.2          | [40]      |
| Haryana, India  | 27.9             | 34.0             | 306.9          | [41]      |
| Punjab, India   | 15–27            | 16–57            | 266–799        | [19]      |
| Ludhiana, Punjab| 28.58            | 50.95            | 569.59         | [14]      |
| World average   | 35               | 45               | 420            | [30]      |
| Barnala, Punjab |                  |                  |                |           |
| Agriculture D1  | 42.55            | 62.72            | 718.50         | Present Study |
| D2              | 49.12            | 74.09            | 692.94         |           |
| D3              | 34.60            | 51.32            | 661.45         |           |
| Undisturbed D1  | 33.12            | 50.92            | 580.38         |           |
| D2              | 46.66            | 71.27            | 667.42         |           |
| D3              | 35.86            | 54.06            | 667.93         |           |

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**Fig. 2** Vertical distribution of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ radionuclides along the soil profile in both agriculture and undisturbed areas
international standards (Table 2). It has been observed that more than 60% of samples have levels higher than the Indian average.

**Elemental ratios**

**Th and K distribution**

eTh/K ratio shows relative potassium enrichment as an indicator of clay mineral species because both Th (by adsorption) and K (chemical composition) are bound to clay minerals [42]. It is expected that sediment processes do not affect the enrichment or depletion of potassium and thus K acts as radioactive label of the source rock [43]. Calculated Th/K ratio presented in Fig. 4a ranges from 0.0598 to 0.397. High value of K and low eTh indicates that the light mineral assemblage dominates in the soil samples and it can be interpreted as quartz-feldspar system. However, few samples (A6D2) shows high eTh value and low K, and thus suggests the presence of heavy thorium bearing minerals in the system.

**Th and U distribution**

Cross-plot of eTh/eU and eTh/K shown in Fig. 4b gives interpretable patterns of the data by giving information about the change in redox potential and change in composition of K [4]. The value obtained for eTh/eU between 3 to 6 and eTh/K between 5 to 10 is consistent with alluvial sediments in oxidising terrestrial environment. This also suggests that this is a quartz-feldspar system with enrichment of clay minerals except few where eTh/K ratio is quite high,
suggesting quartz-feldspar system with enrichment of clay and heavy thorium bearing mineral association.

e\text{Th}/eU value in the sediment samples lies in range of 3.7 to 5.89 with an average value of 4.62, which is higher than the value of average continental crust of approx. 3.8 [44, 45]. This suggests that the samples are enriched in Th(IV) and U(VI). High value of e\text{Th}/eU indicates towards an oxidizing environment and thus suggests uranium mobilization through weathering or leaching.

Radiological hazard

Absorbed dose rate in air (D)

The absorbed dose rate in the present study ranges from 61.25 to 189.85 nGy h\(^{-1}\). Average absorbed dose rate value is 84.09 nGy h\(^{-1}\) and is comparable with the world average of 84 nGy h\(^{-1}\) [30]. The maximum average contribution to the total absorbed dose rate is by \(^{232}\text{Th}\) i.e. 44\% followed by \(^{40}\text{K}-34\%\) and \(^{238}\text{U}-22\%\). The change in absorbed rate is due to the spatial variation of mineral holding sediments of that particular element [39].

Annual effective dose equivalent (AEDE)

The calculated outdoor AEDE varies from 75.12 to 232.83 µSv y\(^{-1}\) with an average of 103.13 µSv y\(^{-1}\). Most of the locations in the region lie in the unsafe region as the value obtained is more than the world average value of 70 µSv y\(^{-1}\) [30].

Radium equivalent activity (Ra\(_{eq}\))

Ra\(_{eq}\) were found to range from 131.11 to 428.17 Bq kg\(^{-1}\) with an average value of 180.76 Bq kg\(^{-1}\). Thus the average value is lower than the world maximum value of 370 Bq kg\(^{-1}\) allowed for public dose and for safe use of material in the building construction [46].

Hazard indices (H\(_{in}\) and H\(_{ex}\))

Hazard indices are calculated to limit the annual external gamma radiation dose of materials to 1.5 mSv y\(^{-1}\); higher values are harmful to the people living in the region. External hazard index (H\(_{ex}\)) value ranges from 0.354 to 1.15 with an average value of 0.49 and Internal hazrad index (H\(_{in}\)) value for the soil samples lies in the range of 0.43 to 1.46 with an average of 0.59. H\(_{ex}\) and H\(_{in}\) value of all the samples in the current study lie within the permissible limit and thus poses negligible hazard to the people living in the region.

Annual gonadal dose equivalent (AGDE)

Calculated AGDE range from 0.43 to 1.31 mSv y\(^{-1}\) with an average value of 0.59 mSv y\(^{-1}\). All the sites have value less than unity, which is the limit recommended by International Commission on Radiological Protection for the public [31].

| 238U Bq kg\(^{-1}\) | 232Th Bq kg\(^{-1}\) | 40K Bq kg\(^{-1}\) | D nGy h\(^{-1}\) | AEDE µSv y\(^{-1}\) | Ra\(_{eq}\) Bq kg\(^{-1}\) | H\(_{in}\) mSv y\(^{-1}\) | H\(_{ex}\) mSv y\(^{-1}\) | AGDE mSv y\(^{-1}\) |
|-------------------|-------------------|-------------------|------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| **Agriculture**   |                   |                   |            |                   |                   |                   |                   |                   |
| Mean              | 42.57             | 63.57             | 694.88     | 87.04             | 106.75            | 186.98            | 0.51              | 0.62              | 0.62              |
| Median            | 39.16             | 58.44             | 681.17     | 81.75             | 100.25            | 174.39            | 0.47              | 0.58              | 0.58              |
| Std Dev           | 14.70             | 24.39             | 110.30     | 21.69             | 26.61             | 49.65             | 0.13              | 0.17              | 0.15              |
| Range             | 87.52             | 150.14            | 555.96     | 128.60            | 157.72            | 297.07            | 0.81              | 1.04              | 0.88              |
| Minimum           | 27.42             | 42.78             | 485.13     | 61.25             | 75.12             | 131.10            | 0.35              | 0.43              | 0.43              |
| Maximum           | 114.94            | 192.92            | 1041.09    | 189.85            | 232.84            | 428.17            | 1.16              | 1.47              | 1.31              |
| **Undisturbed**   |                   |                   |            |                   |                   |                   |                   |                   |                   |
| Mean              | 37.34             | 58.61             | 616.52     | 78.36             | 96.10             | 168.62            | 0.46              | 0.56              | 0.55              |
| Median            | 35.73             | 59.47             | 576.89     | 76.35             | 93.64             | 165.44            | 0.45              | 0.54              | 0.54              |
| Std Dev           | 6.70              | 11.61             | 89.52      | 12.10             | 14.84             | 26.69             | 0.07              | 0.09              | 0.09              |
| Range             | 25.96             | 39.62             | 305.92     | 42.10             | 51.64             | 93.35             | 0.25              | 0.32              | 0.30              |
| Minimum           | 27.97             | 42.33             | 529.48     | 62.62             | 76.79             | 133.73            | 0.36              | 0.44              | 0.44              |
| Maximum           | 53.93             | 81.95             | 835.40     | 104.72            | 128.43            | 227.08            | 0.61              | 0.76              | 0.74              |

Std Dev, Standard Deviation; D, Absorbed Dose Rate in Air; AEDE, Annual Effective Dose Equivalent; Ra\(_{eq}\), Radium Equivalent activity; H\(_{in}\), Internal Hazard Index; H\(_{ex}\), External Hazrad Index; AGDE, Annual Gonadal Dose Equivalent
Multivariate statistical analysis

Descriptive statistics

The list of statistical data (mean, median, standard deviation, range, minimum, maximum) of radionuclides and all the radiological parameters is given in Table 3 for both the agricultural and undisturbed soil samples. The data presented shows variation in the concentration of radionuclides in agricultural and undisturbed areas with minimum value comparable in both the areas.

Pearson correlation coefficient

Study of correlation analysis has been carried out to understand the strength of association and mutual relationships between the variables. Obtained correlation coefficients are given in Table 4 as a linear correlation matrix. It has been observed that there is strong positive correlation coefficient between Th and U radionuclides in both site sediments and radiological parameters. This strong positive correlation is due to Th and U decay series occurring together in nature [47] and it shows that U and Th contribute to the gamma radiation in the soil of the region at both the sites. However, in case of K contrasting behaviour is observed in the agricultural and undisturbed areas. Weak correlation has been observed in case of K and other radionuclides as well as radiological parameters in the agricultural soil samples and is moderately correlated with radionuclides and strongly with radiological parameters in the undisturbed areas. Thus correlation analysis indicates that the mobility of radionuclides is affected by the sedimentation processes in a different way.

Principal component analysis (PCA)

PCA is a multivariate technique used to identify variables by applying varimax rotation with Kaiser Normalization. By extracting the Eigen values and Eigen vectors from the correlation matrix, number of significant factors and the percent of variance explained by each of them were calculated. It is observed from the PCA that PC1 accounts for 89.59% of total variance in agricultural areas and all the variables have positive projection on PC1. As seen from Fig. 5a, PC1 is heavily loaded on U and Th series associated with all the radiological parameters. PC2 is strongly correlated with the potassium and shows contrast between U and Th (negative projection) and K (positive projection) accounting for 10.03% of total variance. These projections show the authentic behaviour of K and U-Th elements due to their enrichment or low influence of K and can be related to the pre-dominance of K-bearing mineral leaching, because K is geochemically mobile under most weathering conditions [48]. However, in case of undisturbed

|  | 238U | 232Th | 40K | D  | AEDE | Ra_{eq} | H_{ex} | H_{in} | AGDE |
|---|------|------|-----|----|------|--------|-------|-------|------|
| Agriculture |      |      |     |    |      |        |       |       |      |
| 238U | 1.000 |      |     |    |      |        |       |       |      |
| 232Th | 0.972 | 1.000 |     |    |      |        |       |       |      |
| 40K | 0.031 | -0.074 | 1.000 |    |      |        |       |       |      |
| D | 0.979 | 0.968 | 0.172 | 1.000 |    |        |       |       |      |
| AEDE | 0.979 | 0.968 | 0.171 | 0.999 | 1.000 |        |       |       |      |
| Ra_{eq} | 0.984 | 0.978 | 0.128 | 0.999 | 0.999 | 1.000 |        |       |      |
| H_{ex} | 0.984 | 0.977 | 0.129 | 0.999 | 0.999 | 0.999 | 1.000 |        |      |
| H_{in} | 0.990 | 0.979 | 0.107 | 0.997 | 0.997 | 0.999 | 0.999 | 1.000 |      |
| AGDE | 0.976 | 0.961 | 0.194 | 0.999 | 0.999 | 0.998 | 0.997 | 0.996 | 1.000 |
| Undisturbed |      |      |     |    |      |        |       |       |      |
| 238U | 1.000 |      |     |    |      |        |       |       |      |
| 232Th | 0.844 | 1.000 |     |    |      |        |       |       |      |
| 40K | 0.661 | 0.418 | 1.000 |    |      |        |       |       |      |
| D | 0.948 | 0.924 | 0.720 | 1.000 |    |        |       |       |      |
| AEDE | 0.948 | 0.924 | 0.720 | 0.999 | 1.000 |        |       |       |      |
| Ra_{eq} | 0.946 | 0.942 | 0.684 | 0.999 | 0.999 | 1.000 |        |       |      |
| H_{ex} | 0.943 | 0.945 | 0.675 | 0.997 | 0.997 | 0.999 | 1.000 |        |      |
| H_{in} | 0.966 | 0.929 | 0.684 | 0.996 | 0.996 | 0.997 | 0.996 | 1.000 |      |
| AGDE | 0.945 | 0.918 | 0.731 | 0.999 | 0.999 | 0.997 | 0.997 | 0.994 | 1.000 |

D, Absorbed Dose Rate in Air (nGy h^{-1}); AEDE, Annual Effective Dose Equivalent (µSv y^{-1}); Ra_{eq}, Radium Equivalent activity (Bq kg^{-1}); H_{in}, Internal Hazard Index (mSv y^{-1}); H_{ex}, External Hazard Index (mSv y^{-1}); AGDE, Annual Gonadal Dose Equivalent (mSv y^{-1})
samples, only one component forms and no variation is observed. This shows that agricultural activities affect the correlation between radionuclides to a greater extent.

**Cluster analysis**

Cluster analysis was applied to identify and classify groups with similar characteristics among natural radioisotopes and radiological parameters. This also confirms the existing correlation between variables. Within same cluster, each observation is mostly like others, but all the clusters are dissimilar from each other. Similarity is measure of distance between clusters relative to the largest distance between any two individual variables. Zero percent similarity means clusters are as disparate as least similar region whereas similarity of one hundred percent means clusters are zero distance apart [32].

In the current dendrogram shown in Fig. 5b, all the variables are grouped into 3 significant clusters on the basis of similarity between them. Cluster-I shows correlation of $^{238}\text{U}$ with all the radiological parameters such as AGDE, Hex, Hin, Req, AEDE, D. Cluster-II consists of $^{232}\text{Th}$ and main radiological parameters distribution. Cluster-III accounts for $^{40}\text{K}$. Cluster analyses suggest that $^{238}\text{U}$ is the major contributor of radiation hazard and $^{40}\text{K}$ does not contribute to any radiological parameters in the agricultural samples of study area. Results corroborate with the PCA and Pearson correlation data as well.

**Conclusion**

$^{238}\text{U}$, $^{232}\text{Th}$ and $^{40}\text{K}$ radionuclide levels in the soil profile are found to be higher than the previously reported levels and the global average. Levels are higher in the agricultural regions than the undisturbed and the probable cause of difference is the use of high amount of fertilisers as levels are comparable after depth interval of 600 cm at both the lands. However, value is higher than global average even in the undisturbed areas and indicates the geogenic presence of radionuclides in the study region with anthropogenic enrichment. Few of the sites with enhanced uranium levels mark uranium accumulation zones in the soil from where it may get released into the groundwater and can be studied in detail in the future. It is observed that levels are higher in clay sediments at top than the sandy sediments at bottom. Vertical movement of uranium can be traced from the sub-surface sediments through weathering or leaching due to the prevailing oxidising conditions. It shows that soil of the region act as source and sink for uranium with changing redox conditions and counts to be the major factor for the high uranium detected in groundwater of the region.

Various radiological parameters have been calculated, keeping in view that the entire study area is a living place and agricultural region. Sand in the region is also used as construction material thus $\text{Ra}_{eq}$ and $\text{H}_{eq}$ were calculated to know the radiological hazard. These indices were found to be within the permissible limits, thus posing insignificant radiation hazard and soil can be used safely for construction purposes. However, Annual Effective Dose Equivalent (AEDE) is found to be higher than the world average and poses serious health risk as per its carcinogenic effects are
concerned. Thus there is a need to monitor the radioactivity levels in the future as well.

The data obtained is also important for natural radioactivity mapping and provides the baseline in future to access the environmental pollution due to change in radioactivity levels by any of the nuclear, industrial or human activity.

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Declarations

Conflicts of interest The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analysis, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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