

Assessment of natural radioactivity in soil around Khetri copper belt of Rajasthan, India

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Received: 30 August 2023 / Accepted: 26 November 2023 / Published online: 4 January 2024 © Akadémiai Kiadó, Budapest, Hungary 2024

Abstract

The concentration of natural radionuclides $(^{226}Ra, ^{232}Th, ^{40}K)$ in the fifty soil samples around the Khetri copper belt is measured using an HPGe detector. The values of ²²⁶Ra, ²³²Th, and ⁴⁰K are found to be lying in the range of 5.2 ± 0.3 to 27.5 ± 0.6 Bq kg⁻¹, 12.9 ± 0.4 to 38.8 ± 1.3 Bq kg⁻¹, and 113.3 ± 27.8 to 308.5 ± 31.2 Bq kg⁻¹ respectively. The radium equivalent activity ranged from 38.63 to 93.35 Bq kg⁻¹ with a mean value of 56.21 Bq kg⁻¹. The values of absorbed dose rate, annual efective dose, and annual gonadal dose rate are also estimated, which comes out to be less than their corresponding world average values. Hazard indices (internal and external) and level indices (alpha and gamma) are also observed to be less than unity. The soil of the study area can be considered safe for people living there because of its low radiological risk.

Keywords Natural radioactivity · Khetri copper belt · HPGe · Radium equivalent activity · Radiological hazards

Introduction

Natural radioactivity has existed since the beginning of the earth. Humans are subjected to natural radiation from inside and outside the Earth's atmosphere. Extraterritorial radiation is caused by cosmic radiation from space, whereas earth-based terrestrial radiation is caused mainly by ionising radiation emitting from the rocks, soil, and water around us. These radiations emanate naturally in the environment due to the presence of radioactive series such as the 238 U series, 232 Th series, and 40 K [[1\]](#page-8-0). Humans are also exposed to man-made radioactivity due to human events such as nuclear fallout and medical applications [[2\]](#page-8-1). According to the IAEA, natural radionuclides contribute 80% of the time to absorbed dosage in the environment, while cosmic rays contribute the remaining 20% [[3\]](#page-8-2).

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Natural radioactivity can be found in soil, water, rocks, and the atmosphere [[4\]](#page-8-3). The soil is one of the major sources of human radiation exposure [[5\]](#page-8-4). Radioactivity in the soil is caused mainly by primordial radionuclides and decay products in uranium, actinium, and thorium radioactive series [\[6–](#page-8-5)[9\]](#page-8-6). As a result, most radioactivity measurement studies concentrate on natural background radiations from primordial radionuclides, which account for around 80% of an individual's annual radiation dose [\[10](#page-8-7)]. The radiation exposure varies in strength from region to region due to geological and radiochemical variations [[11,](#page-8-8) [12](#page-8-9)]. Diferent types of rocks contain diferent amounts of radioactivity; for example, high amounts of radioactive radiation are found in volcanic and granitic rocks. In contrast, metamorphic rocks and sedimentary rocks are generally observed to have lower levels of radioactivity [\[13](#page-8-10)]. There has been an upsurge in the exploration of radioactivity in soils and parent rocks worldwide in the last ten to twenty years (both on an individual and organisational level) $[13-15]$ $[13-15]$. As soil transfers radionuclides to biological systems, it is an important component of radioactive pollution $[16]$ $[16]$. Thus, measuring the radioactivity $(^{238}U, ^{232}Th, ^{40}K)$ levels in the soil becomes imperative. Radioactivity in the soil is also utilised to investigate radiation dangers, nuclear safety, and exploration [[17\]](#page-8-13).

The present study evaluates the level of naturally occurring radioactivity in the soil around the copper mining area

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of Khetri, Rajasthan, India. The interest behind the selection of this region is due to the presence of a nearby mining area, which is having a signifcant impact on the soil of the surrounding area and the life of the people living nearby. The literature review reveals that several studies have been conducted around copper and uranium mines in India and worldwide. Nguyen et al. [[18\]](#page-8-14) measured the radioactivity around the copper mines region and found that the average values of ^{226}Ra , ²³²Th, and ⁴⁰K are 254 Bq kg⁻¹, 54 Bq kg⁻¹, 560 Bq kg⁻¹, respectively. Similarly, Atibu et al. [[19](#page-8-15)] discovered 226 Ra activity 5 to 10 times greater than the global average values in the vicinity of copper mining areas of the Democratic Republic of Congo. Patra et al. $[20]$ $[20]$ reported the mean values of 226 Ra, ²³²Th, and ⁴⁰K as 81 Bq kg⁻¹, 65 Bq kg⁻¹, and 517 Bq kg⁻¹, respectively, at the Singhbhum shear zone of Jharkhand, India. The natural radioactivity estimated in the artisanal mining sites of Anka in Nigeria is reported by Akpanowo et al. [[21](#page-8-17)]. It was observed that the radioactivity of 226 Ra and 232 Th are higher than the world average values, while the 40 K values were lower than the world average values. However, there is no systematic radiation evaluation in and around the Khetri mining area of Rajasthan, India.

To measure the background radioactivity in the region, we collected an extensive set of soil and groundwater samples. The uranium concentration in groundwater samples has already been published [\[22](#page-8-18)]. A higher amount of uranium concentration in groundwater is observed in many of the measured samples in the studied region, indicating the presence of higher uranium content in the region's bedrock. However, the soil of this arid region is continuously shifting due to widespread saltation and suspension processes. Due to this, the soil radioactivity would represent only an average radioactivity value of the region and may not represent the bedrock properties. In this study, the amount of ^{226}Ra , 232 Th, and 40 K radionuclides in the surface soil of the study area is estimated using a properly shielded HPGe detector system at IIT Ropar. The concentration of 226 Ra is measured instead of 238U as most of the infuence of the 238U series is estimated by 226 Ra [\[23](#page-8-19)]. The observed data can be taken as baseline data to notice any future changes in the radioactivity levels of the soil due to man-made activities. The radiological impact of these radionuclides on the inhabitants and the environment is estimated by computing the absorbed dose rate, Raeq (radium equivalent activity), yearly efective absorbed amount, and gamma and alpha radiation hazard indices.

Study area

The sampling area is primarily located in the districts of Jhunjhunu and Sikar in Rajasthan, India, and the sampling locations are depicted in Fig. [1.](#page-2-0) The soil samples were taken from the area surrounding the Khetri copper belt. The Khetri copper belt (KBC), located in the Aravalli hills, stretches for around 100 km from Singhana (28°05′: 75°49′) in Jhunjhunu District to Sangarva (27°34′: 75°18′) which is located in Sikar District. Khetri Nagar is wellknown for the Copper Project of Hindustan Copper Limited (HCL), a government-run public-sector enterprise that created and now operates the city. It has an average elevation of 485 m (1591 feet). There are primarily three mines in the Khetri copper belt: (a) the Madhan Kudhan mines, which are the largest underground metal mines in the country; (b) Chandmari, located 1 km northwest of Khetri town; and (c) Kolihan, located 10 km southwest of Nagar. The Khetri copper belt is divided in two by the Kantli Fault and a small seasonal river called Kantli. Khetri copper belt rocks are connected with mafc volcanic rocks derived from the Delhi Super Group. They are further classifed into (1) the Alwar Group and (2) the Ajabgarh Group. The Ajabgarh group of the Delhi super-group covers most of the research region. The Ajabgarh group comprises phyllites, biotite schists, calc gneisses, and other pre-Delhi intrusives such as amphibolites, granites, pegmatites, and epidiorites. Diorite, epidiorites, amphibolite, and other basic intrusives are examples. The district's climate is mostly desert.

The region has three seasons: summer, winter, and monsoon. The temperature can drop below 0 °C in the winter and reach 50 °C in the summer. The soil in the examined area primarily comprises lithosols and regisols of the hill type. These gravels are located just beneath the.

surface and are light-textured, relatively well-drained, and range from reddish to greyish brown.

Material & measurement steps

Sample collection and preparation

A total of 50 soil samples were taken at various locations around the study area. Each soil sample carries approximately 1 kg weight and is taken from a depth of 15 cm of the surface to avoid any contamination from the surface soil. The soil samples are collected with GPS coordinates and sealed in polythene bags. Each soil sample was dried for 24 h in an oven at 110 °C to remove moisture. After proper grinding, these samples are passed through a sieve of 0.5 mm to make them homogeneous. Soil samples thus processed are sealed in air-tight taped 250 cc plastic containers. They are stored for 30 days to achieve secular equilibrium between the natural radionuclides and their respective radioactive progenies. The samples were properly coded according to the sampling location; these codes are listed in Table [1](#page-3-0).

Fig. 1 Study Area Map along with sample locations

Calculation of activity concentration

The gamma spectroscopy of soil samples was carried out using an n-type coaxial HPGe detector (ORTEC, model GMX20) at IIT Ropar. For ${}^{60}Co$ at 1.33 MeV, the detector has a relative efficiency of 20% and an energy resolution of 1.8 keV. The minimum detection limit of the detector is 2.74 Bq kg⁻¹ for ²²⁶Ra, 1.17 Bq kg⁻¹ for ²³⁴Th, and 7.08 Bq kg−1 for 40K. A 10 cm thick lead shielding around the detector and sample setup is utilised [[24](#page-8-20)] to reduce the background radiation. The detector was linked to a PC-based MCA and ADC for data collecting. Since the decay of 226 Ra and 232Th do not produce gamma rays, their activity values are determined by calculating the activities of their decay products.

The estimation of 226 Ra activity can be carried out using the gamma measurements of its subsequent decay products, namely, ²¹⁴Pb (295 keV), ²¹⁴Bi (609 keV), and ²¹⁴Bi (1120 keV). Similarly, the decay products of the 232 Th series, namely, ²¹²Pb (238 keV), ²²⁸Ac (911 keV), and ²²⁸Ac (968 keV), with the assumption of the decay series being in equilibrium $[25]$ $[25]$, are used. The activity of 40 K is measured using the 1460 keV peak directly. The samples and the background were counted over a period of 43,200 s. The LAMPS software was used for spectrum analysis.

Calculation of activity (Bq kg−1)

The activity of radioisotopes was estimated using the relation [[26\]](#page-8-22)

Activity
$$
(Bq \text{ kg}^{-1}) = \frac{cps}{W * \eta * Py}
$$
 (1)

where cps is net counts per second after subtraction of background counts, η is the efficiency of the detector, W is the weight of the sample in kg, and Pγ is the photon emission probability for that energy.

Health risk assessment

Absorbed dose rate

Using the activity concentration of radionuclides ^{226}Ra , 232 Th, and 40 K in the soil, it is possible to determine the

absorbed dose rate and annual effective dose in the study area mtrotion Table 1 Radionuclides

air-absorbed gamma dose rates from gamma radiations at 1 m above the earth's surface. [\[27](#page-8-23), [28](#page-8-24)]

$$
Indoor(nGyh^{-1}) = 0.92A_{Ra} + 1.1A_{Th} + 0.08A_K
$$
 (2)

$$
Outdoor(nGyh^{-1}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K}
$$
\n(3)

where equation no (2) and (3) are the indoor and outdoor absorbed dose rates, 226 Ra, 232 Th and 40 K activity concentration, respectively.

Annual efective dose (AED)

The annual effective dose can be estimated from the values of air-absorbed dose using the relation [[27,](#page-8-23) [28\]](#page-8-24)

$$
Indoor (mSvy^{-1}) = AD(nGyh^{-1}) \times 8760h \times 0.8 \times 0.7SvGy^{-1} \times 10^{-6}
$$
\n(4)

$$
\begin{aligned} \text{Outdoor}\left(\text{mSvy}^{-1}\right) &= \text{AD}\left(\text{nGyh}^{-1}\right) \times 8760\text{h} \\ &\times 0.2 \times 0.7 \text{SvGy}^{-1} \times 10^{-6} \end{aligned} \tag{5}
$$

where AD stands for the absorbed dose. A conversion factor of 0.7 Sv Gy−1 is used, and 0.8 and 0.2 are considered indoor and outdoor occupancy factors, respectively.

Radiation hazards

Radium equivalent activity (Raeq)

The distribution of ²²⁶Ra, ²³²Th, and ⁴⁰K radionuclides in the soil is not homogeneous. To estimate the total effect of exposure from these radionuclides and for comparison of specifc activity of the soils having diferent concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K radionuclides, the Ra_{eq} (Bq kg.⁻¹) is calculated by using the following relation $[29, 30]$ $[29, 30]$ $[29, 30]$ $[29, 30]$

$$
Ra_{eq} = \left(\frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \times 370\tag{6}
$$

where A_{Ra} , A_K , and A_{Th} are the activity of ²²⁶Ra, ⁴⁰K, and ²³²Th in Bq kg⁻¹.

External and internal hazard indices (Hex and H_{in})

For the risk assessment of the effect of these radiations on living beings, the Hazard Indices are calculated. The External Hazard Index is calculated for external exposures using the relation [[27,](#page-8-23) [29\]](#page-9-0)

$$
H_{ex} = \frac{A_{Ra}}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}
$$
 (7)

where A_{Ra} , A_{Th} , and A_K are the activity of ²²⁶Ra, ²³²Th, and 40 K in units of Bq kg⁻¹.

The internal hazard index is calculated for internal exposure to radiation through inhalation to humans and estimated using the relation [[27](#page-8-23), [29](#page-9-0)]

$$
H_{in} = \frac{A_{Ra}}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}
$$
 (8)

where A_{Ra} , A_{Th} , and A_K are the activity of radionuclides ²²⁶Ra, ²³²Th, and ⁴⁰K in units of Bq kg⁻¹. The value of the external and internal hazard index is considered safe, having a value less than unity.

Gamma index (I_v) and alpha index (I_a)

Gamma exposure from rocks and soils is measured using the gamma index. It considers how much radiation exposure is possible when soil and rocks are used as building materials. The relationship shown below can be used to estimate it. [\[28,](#page-8-24) [30\]](#page-9-1)

$$
I_{\gamma} = \frac{A_{Ra}}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000}
$$
 (9)

The value of I_{ν} ≤ 0.5 corresponds to 0.3 mSv y⁻¹ dose rate, and for $I_{\gamma} \ge 0.5$, the value corresponds to 1 mSv y⁻¹ dose rate.

The exposure induced by inhaling 222 Rn, a decay product of.226Ra, can be assessed in terms of the alpha index, which is estimated by the relationship [\[30](#page-9-1), [31](#page-9-2)]

$$
I_{\alpha} = \frac{A_{Ra}}{200}
$$
 (10)

The value of I_α will be unity for 200 Bq kg⁻¹ activity concentration of ²²⁶Ra in the soil. The values of I_{α} exceeding unity signify the higher rates of radon exhalation from the soils.

The gonadal dose equivalent rate (AGDE)

The bone surface cells, bone marrow, and gonads are widely regarded as the most critical organs. AGDE was accomplished using the equation shown below [[13\]](#page-8-10).

$$
AGDE(\mu Svy^{-1}) = 3.09 \times A_{Ra} + 4.18 \times A_{Th} + 0.314 \times A_K
$$
\n(11)

The maximum safe value¹³ of AGED is 300 mSv v^{-1} .

Cancer risk

The equation is used to calculate the excess lifetime cancer risk [\[32](#page-9-3), [33\]](#page-9-4).

$$
ELCR = AEDE \times DL(65 \text{years}) \times RF(ICRP60, 1990 \text{uses} 0.05 \text{Sv}^{-1})
$$
\n(12)

Result and discussion

In this manuscript, the gamma radioactivity concentration in ffty soil samples is measured using an HPGe detector. The observed values of activity concentration of 226Ra, 232Th, 40 K, and Ra_{eq.} along with the sample locations and radiological doses, are summarised in Table [1](#page-3-0).

The radioactivity concentrations of 226 Ra in the study area vary from 5.2 ± 0.3 to 27.5 ± 0.6 Bq kg⁻¹. The average activity concentration of ²²⁶Ra in soil is 10.6 ± 0.6 Bq kg⁻¹ and is lower than the world average value for $226Ra$ in soil (32 Bq kg⁻¹) [\[29](#page-9-0)]. The activity concentration of ²³²Th varies between 12.9 ± 0.4 and 38.8 ± 1.3 Bq kg⁻¹ with an average value of 21.4 ± 1.0 Bq kg⁻¹. The highest value is found for the village 'Tihara' and the minimum value for the village 'Nagli Saledhri'. The ²³²Th concentration in all the soil sam-ples is below the world average value of 45 Bq kg⁻¹ [[29](#page-9-0)]. The activity concentration of ⁴⁰K lies between 113.3 ± 27.8 and 308.5 ± 31.2 Bq kg^{-1} , having an average value of 194.8 ± 22.4 Bq kg⁻¹. It is observed that the concentration of $40K$ lies below the world average value of 420 Bq kg⁻¹ [[29\]](#page-9-0). The activity concentrations of ²²⁶Ra, ²³²Th, and ⁴⁰K are comparable or lower than those reported in neighbouring states and other areas of Rajasthan [[34](#page-9-5)[–37](#page-9-6)]. The lower levels of natural radionuclide concentration (particularly 40 K) recorded in the research region can be linked to the geological and mineral composition of the Khetri copper belt. When compared to locations with diferent geological formations, the geology of the Khetri copper belt may not be conducive to the presence of higher concentrations of natural radionuclides. The region's soil is older alluvium, and the region's principal rock types are metamorphic and igneous, resulting in a low concentration of radionuclide 40 K [[38](#page-9-7)]. This study is the frst of its type in the studied region. The obtained data can be taken as baseline data for future reference. Spatial distribution maps of the region also indicate no region of higher radioactivity in the study area. The spatial distribution maps are shown in Fig. [2](#page-6-0).

Radium equivalent activity (Raeq.)

Radium equivalent activity is lying between 38.63 and 93.35 Bq kg⁻¹ with a mean value of 56.21 Bq kg⁻¹, as shown in Table [1.](#page-3-0) Maximum value of $Ra_{eq.}$ is found for the village

Indoor and outdoor absorbed dose rate

Indoor absorbed dose rate (D_i) for all the samples lies between 34.52 and 79.98 nGy h^{-1} with an average value of 48.89 nGy h^{-1} . The value for all the soil samples is less than the world average value of 84 nGy h^{-1} [[29\]](#page-9-0). Outdoor absorbed dose rate (D_0) varied between 18.30 and 42.58 nGy h⁻¹ with a mean value of 25.95 nGy h⁻¹. The world average value for outdoor absorbed dose rate is 58 nGy h^{-1} [[29](#page-9-0)], and all samples exhibited values less

than the recommended limit. The obtained values are displayed in Table [1](#page-3-0).

Indoor and outdoor annual efective dose (AED) and Annual gonadal dose (AGDE)

The values of indoor and outdoor annual effective doses are tabulated in Table [1.](#page-3-0) The indoor annual effective dose is observed between 0.17 and 0.39 μ Sv y⁻¹ with an average value of 0.24 μ Sv y⁻¹. The values of the indoor absorbed dose rate were found to be lying below the rec-ommended limit of 0.48 µSv y⁻¹ [[29\]](#page-9-0). Outdoor annual effective dose is observed to be varying in between 0.02 and 0.05 μ Sv y⁻¹. The average value for outdoor AED value comes out to be 0.03 μ Sv y⁻¹. The outdoor AED value for all samples is lying well below the recommended limit of 0.07 μ Sv y⁻¹ [[29](#page-9-0)].

Fig. 2 Spatial Distribution map of radionuclides present in the soil of the study area

The yearly gonadal dose value (AGDE) ranges from 130.84 to 299.64 µSv y−1, having an average value of 183.39 μ Sv y⁻¹, as shown in Table [2.](#page-7-0) However, the AGDE value for all samples lies below the prescribed limit of 300 μ Sv y⁻¹ [[29\]](#page-9-0). However, few samples indicated AGDE value very close to the recommended limit.

Hazards indices

The values of the internal hazard index (H_{in}) ranged from 0.12 to 0.30, with a mean value of 0.18. Similarly, the external hazard index (H_{ex}) values lie between 0.10 and 0.25, along with an average value of 0.15. The values of both internal and external hazard indices lie below the recommended limit of 1 [[29](#page-9-0)], which indicates no radiological danger due to the radionuclides present in the soil of the study area. The values are presented in Table [2.](#page-7-0)

Level indices

The value of the alpha index (I_{α}) is found to be between 0.03 and 0.14. The average value of I α is obtained as 0.05, which is very low from the prescribed limit of 1. The evaluated value of gamma index I_{γ} varies between 0.15 and 0.34 with an average value of 0.21, as shown in Table [2](#page-7-0). The values of I_{ν} lie below the prescribed limit of 1 for all the soil samples, indicating no radiation hazards to the people living in the study area. The obtained values are shown in Fig. [3](#page-7-1) in the form of the box plot.

Cancer risk

The possible cancer risk (CR) due to the presence of the radionuclide in the soil of the study area is also estimated. The value of the cancer risk area was found to be lying in the range of 0.07×10^{-3} – 0.17×10^{-3} with a mean value of 0.10×10^{-3} , as shown in Table [2](#page-7-0). The cancer risk value for all the soil samples is lying well below the

Fig. 3 Box plot for hazard indices and level indices in the studied region

prescribed limit of 0.29×10^{-3} [\[28\]](#page-8-24), indicating low cancer risk in the soil of the study region.

Conclusion

In this study, the concentration of primordial radionuclides $(^{226}Ra, ^{232}Th,$ and $^{40}K)$ is measured in fifty soil samples collected from the region around the Khetri copper belt, Rajasthan, India. The radionuclides are estimated using the gamma spectroscopy technique using the HPGe detector setup. It is observed that the concentration of radionuclides $(^{226}Ra, ^{232}Th,$ and $^{40}K)$ lies well within the corresponding world average values for all the soil samples. The mean values of ²²⁶Ra, ²³²Th, and ⁴⁰K are 10.6 ± 0.6 Bq Kg⁻¹, 21.4 ± 1.0 Bq kg^{-1} , and 194.8 ± 22.4 Bq kg^{-1} respectively. Radium equivalent activity is found to be lying between 38.63 and 93.35 Bq kg⁻¹ with a mean value of 56.21 Bq kg⁻¹, which is far less than the recommended value of 370 Bq Kg−1. The indoor and outdoor absorbed dose rate and annual efective dose values also lie below the world average values, indicating low radiological implications of the soil of the study area. Annual gonadal dose ranges from

	H_{in}	$H_{\rm ex}$	$\mathbf{I}_{\mathcal{N}}$	\mathbf{I}_{α}	AGDE $(\mu Sv y^{-1})$	$CR \times 10^{-3}$
Minimum	0.12	0.10	0.15	0.03	130.84	0.07
Maximum	0.30	0.25	0.34	0.14	299.62	0.17
Mean	0.18	0.15	0.21	0.05	183.39	0.10
Median	0.18	0.15	0.20	0.05	179.11	0.10
Standard deviation	0.04	0.03	0.04	0.02	36.76	0.02
Variance	0.002	0.001	0.002	0.00	1351.56	0.00
Geometric mean	0.18	0.12	0.20	0.05	179.90	0.10

Table 2 Statistical parameters for the Hazard indices, Level indices, gonadal dose, and cancer risk in the soil

130.84 to 299.64 μ Sv y⁻¹, which is slightly less than the world average value of 300 μSv y⁻¹. The value of hazard indices (H_{in} and H_{ex}) and level indices (I_{α} and I_{γ}) are less than unity for all the samples, which indicates that the soil of the study region has no radiological hazards. The soil of the region also indicated no cancer risk. The fndings of this study state that the soil around the Khetri copper belt has no signifcant radiological risk to the inhabitants living in the area.

Acknowledgements The research work of Naresh Kumar is supported by the research grant (Junior Research Fellowship) of the University Grant Commission (UGC) of India.

Declarations

Conflict of interest The authors declare that they do not have any potential confict of interest which may infuence the work reported in the present research paper. Most of the data from research work is included in the present paper, and any further information may be obtained from the corresponding author.

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