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Natural radioactivity (^{226}Ra, ^{232}Th &^{40}K) in soil samples from the Eastern Valley region of Manipur, India

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Abstract: The radioactivity concentration and radiological parameters have been assessed in soil samples collected from Thoubal and Imphal East districts in Manipur, India, using a High-Purity Germanium (HPGe) detector. The range of activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in the soil from the studied areas varies from 26 Bq/Kg (Heirok) to 132 BqKg^{-1}(Porompat), 22 Bq/Kg (Yairipok Khoirom) to 212 Bq/Kg (Porompat), and 451 Bq/Kg (Thoubal Bazar) to 1819 Bq/Kg (Lamlai Bazar) with overall average values of 70 Bq/Kg, 90 Bq/Kg, and 895 Bq/Kg, respectively. The Radium equivalent activities (Ra_{eq}) have been evaluated in soil samples to assess the radiation hazard. The corresponding absorbed dose rate, annual effective dose, and external radiation hazard index in the study area range from 56.9 to 264.9 nGy/h, 0.3 to 1.6 mSv, and 0.3 to 1.5, with an average value of 127.7 nGy/h, 0.8 mSv/y, and 0.7, respectively.

Keywords: Natural radioactivity, annual effective dose, Radiation hazard index, HPGe detector, Thoubal district, Imphal East district

Introduction

Natural background gamma radiation primarily originates from terrestrial radionuclides, which are ubiquitously present in trace concentrations within the Earth's soils. The specific concentration and distribution of these radionuclides are closely linked to the geological characteristics of the region, particularly the parent rock types from which the soils are derived. For instance, igneous rocks such as granite typically exhibit higher levels of radioactivity compared to most sedimentary rocks. However, certain sedimentary formations, such as shales and phosphate rocks, can also contain elevated radionuclide concentrations, constituting notable exceptions to this general trend [1].

Evaluating gamma radiation dose rates from natural sources is critical, as natural background radiation constitutes the predominant contributor to the global population's external exposure to ionizing radiation [2–4]. Numerous radiological assessments have been conducted worldwide to quantify the concentration of naturally occurring radionuclides in soil matrices [5–9]. These investigations have consistently identified three principal radionuclides ^{238}U and ^{232}Th series and ^{40}K as major contributors to external gamma dose rates both indoors and outdoors [4].

Research into the natural radioactivity levels in soils is vital not only due to the associated radiological health implications but also because such radionuclides serve as valuable tracers in geochemical and biochemical studies. Of particular interest are uranium-series radionuclides, which exhibit relatively high mobility in biological systems, thereby raising concerns regarding their environmental behaviour and potential uptake by living organisms [1]. While natural radioactivity is present across the Earth's crust, the extent of its concentration can vary spatially within relatively narrow limits depending on geological and environmental conditions [10].

The present study focuses on Thoubal (now, it is split into Thoubal district and Kakching district) and Imphal East districts, which are the eastern valley districts of Manipur, India. These districts lie approximately 300 km from Domiasiat in Meghalaya, a region recognized for its significant deposits of heavy minerals. Thoubal district encompasses an area of approximately 514 km² and had a recorded population of 420,517 according to the 2011 census. Imphal East district spans an area of about 709 km², with a population of 456,113 reported in the same census.

A radiological survey was carried out across ten selected sites in the Thoubal and Imphal East Valley regions to assess the levels of naturally occurring radioactivity. Measurements were performed using a High Purity Germanium (HPGe) gamma-ray spectrometer. Key radiological parameters, including the absorbed gamma dose rate, annual effective dose, radium equivalent activity, and external hazard index, were calculated. The findings were evaluated against the reference levels recommended by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2008) [4], and compared with data from similar international radiological assessments.

Materials and Methods

Sample collection and radionuclide measurement

A total of ten soil samples were collected from selected sites within Thoubal and Imphal East districts of Manipur, India, five from each district, as illustrated in Figure 1. Samples were obtained from a depth of approximately 30 cm below the ground surface to minimize the influence of surface contaminants. Each sample weighed around 1.5 kg and was considered representative of the respective sampling location. The collected samples were placed in labelled polyethylene bags and transported to the laboratory for further processing.

The soils in the study area are predominantly alluvial, characterized by their soft texture, which facilitates easy pulverization into fine powder. In the laboratory, each soil sample was oven-dried at 110°C for approximately 15 hours to remove moisture content. The dried samples were then homogenized, ground, and sieved using a 1 mm mesh to ensure uniformity. Subsequently, approximately 200 grams of the processed sample were packed into standardized Marinelli beakers, sealed tightly to prevent the escape of radon gas, and stored for a minimum of four weeks. This storage period allowed the attainment of secular equilibrium between parent radionuclides (^{234}U and ^{232}Th) and their short-lived progeny [11].

It is well established that approximately 98.5% of natural gamma radiation arises from ^{226}Ra (decay from ^{238}U series) and its decay products, underscoring the radiological significance of this radionuclide chain [12]. The specific activities of ^{226}Ra, ^{232}Th and ^{40}K in each soil sample were determined using a high-purity germanium (HPGe) gamma-ray spectrometer, featuring a relative efficiency of 35% and an energy resolution of 1.8 keV at the 1333 keV gamma line of ^{60}Co.

For energy calibration of the spectrometry system, a mixed point source containing ^{241}Am (59.5 keV), ^{137}Cs (661.7 keV), and ^{60}Co (1173.2 and 1332.5 keV) was employed. Efficiency calibration was performed using a uranium-ore reference standard (IAEA-RGU-1) with a certified activity of 4940 Bq/kg, which was measured after equilibrium had been established with its daughter nuclides. The gamma energies and corresponding daughter radionuclides used for calibration are listed in Table 1.

A fourth-order polynomial function was fitted to the energy-efficiency data to derive a calibration curve, which was subsequently used to determine detection efficiencies for the radionuclides of interest. The activity of ^{40}K was estimated using the 1460 keV gamma peak, while the activity of ^{226}Ra was assessed using the 352 keV peak of ^{214}Pb and the 1766 keV peak of ^{214}Bi. The specific activity of ^{232}Th was calculated from the prominent gamma emissions of ^{208}Tl (510.6, 583.2, and 2610 keV), ^{212}Bi (727.2 keV), and ^{212}Pb (238.6 keV).

Assessment of Radiological parameters:

Radium equivalent activity (Ra_{eq})

The environmental distribution and concentration of naturally occurring radionuclides such as ^{226}Ra, ^{232}Th, and ^{40}K are inherently non-uniform, owing to variations in geological and geochemical conditions. To assess the overall radiological impact of these radionuclides in a unified manner, the concept of radium equivalent activity (Ra_{eq}) is commonly employed. This parameter provides a single index that accounts for the combined gamma radiation hazard posed by ^{226}Ra, ^{232}Th, and ^{40}K. Ra_{eq} is calculated based on the assumption that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th, and 4810 Bq/kg of ^{40}K produce equivalent gamma dose rates in the environment [13–14] and is given as, and

$$Ra_{eq} = {}^{226}Ra + 1.43\ {}^{232}Th + 0.077\ {}^{40}K \quad - - - \quad (1)$$

A value of 370 Bq /Kg gives an annual effective dose of 1 mSv/y.

External Hazard index (H_{ex}):

The external hazard index (H_{ex}) is used to evaluate the potential external exposure risk associated with natural gamma radiation from the radionuclides present in the samples. It is calculated using the empirical formula proposed by Beretka and Mathew [13] as

$$H_{ex} = {}^{226}Ra/370 + {}^{232}Th/259 + {}^{40}K/4810 \quad - - - \quad (2)$$

The recommended upper limit for the external hazard index (H_{ex}) is less than unity, which corresponds to the maximum permissible value of radium equivalent activity (Ra_{eq}) of 370 Bq/kg. This threshold ensures that the annual effective external gamma dose remains below 1.5 mGy/year [15].

Absorbed dose rate (D):

The absorbed gamma dose rate in air, D (nGy/h), resulting from the presence of naturally occurring radionuclides (^{226}Ra, ^{232}Th, and ^{40}K) in soil, was calculated following the guidelines provided by the International Commission on Radiological Protection (ICRP) [16], using the following equation:

$$D(nGy/h) = 0.427C_{Ra} + 0.662C_{Th} + 0.043C_k \quad - - - \quad (3)$$

Where C_{Ra}, C_{Th} and C_k are the activity concentrations (Bq/Kg) of ^{226}Ra, ^{232}Th, and ^{40}K, respectively.

Annual effective dose (D_{eff}):

The evaluation of annual effective dose (D_{eff}), a conversion coefficient of adult as 0.7Sv/Gy [17] is considered from the absorbed dose in air to the effective dose.

Results and Discussion:

The measured activity concentrations of naturally occurring radionuclides ^{226}Ra, ^{232}Th, and ^{40}K from ten sampling sites (as shown in Figure 1) are presented in Table 1. The specific activity values ranged from 26 to 132 Bq/kg for ^{226}Ra, 22 to 212 Bq/kg for ^{232}Th, and 451 to 1819 Bq/kg for ^{40}K, with corresponding mean values of 70 Bq/kg, 90 Bq/kg, and 895 Bq/kg, respectively. For comparative purposes, specific activity values reported from other countries are provided in Table 2.

The activity concentrations observed in the present study are moderately elevated relative to values reported in several international studies and are nearly twice the global average levels. The results follow the trend $^{226}Ra < ^{232}Th < ^{40}K$ in terms of mean activity concentrations.

The radium equivalent activity (Ra_{eq}), which provides a single index accounting for the combined radiological effects of ^{226}Ra, ^{232}Th, and ^{40}K, ranged from 118.1 to 552.8 Bq/kg across all sampling sites, with an average of 267.1 Bq/kg. This average value remains below the recommended safety limit of 370 Bq/kg established by the Organization for Economic Co-operation and Development (OECD) [28]. However, two sites, namely, Lamlai Bazar (552.8 Bq/kg) and Porompat (513.1 Bq/kg) exceeded this threshold, indicating localized radiological concerns.

The absorbed gamma dose rate in air (D), annual effective dose rate (D_{eff}), and the external hazard index (H_{ex}) calculated from the measured activity concentrations varied from 56.9 to 264.9 nGy/h, 0.3 to 1.6 mSv/y, and 0.3 to 1.5, respectively. The mean values were 127.7 nGy/h for absorbed dose rate, 0.8 mSv/y for annual effective dose, and 0.7 for the external hazard index.

The average absorbed dose rate (127.7 nGy/h) and annual effective dose (0.8 mSv/y) exceed the global average values of 59 nGy/h and 0.4 mSv/y, respectively, as reported by UNSCEAR (2000) [4] and UNSCEAR (1998) [10]. They also surpass findings from other regional studies, including 55 nGy/h by Butt et al. [29] and 52.8 nGy/h with 0.5 mSv/y by Alaamer et al. [25]. 9 out of 10 sampling locations exhibited annual effective dose rates equal to or exceeding the global average of approximately 0.5 mSv/y [24], suggesting elevated natural radioactivity likely associated with the presence of heavy mineral deposits in the region. Notably, two sites reached the critical value of unity for the external hazard index, indicating potential radiological risk in those areas [15].

Conclusions

The average values of key radiological parameters, namely, absorbed gamma dose rate, annual effective dose, radium equivalent activity, and external hazard index, were assessed for ten locations across Thoubal and Imphal East districts. The mean absorbed dose rate was found to be 127.7 nGy/h, while the corresponding annual effective dose rate, radium equivalent activity, and external hazard index were 0.8 mSv/y, 267.1 Bq/kg, and 0.7, respectively.

In most of the studied sites, the annual effective dose exceeded as compare to the global average of approximately 0.5 mSv/y. This elevated dose level is attributed to the naturally enhanced radioactivity, likely due to the presence of heavy mineral deposits in the region.

The data generated in this study serve as a valuable radiological baseline for the investigated area and can be used for future regional assessments, national comparisons, and evaluations against international radiological safety standards.

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Table 1: Radionuclide daughter products and their energies used for efficiency calibration

Radionuclides	**Energy (Kev)**
Th-234	63.34
	92.79
Ra-226	186.22
Pb-214	352.36
Bi-214	609
	935.21
	1121
	1766.8

Table 2: Specific Activity of ^{226}Ra, ^{232}Th, and ^{40}K in soil samples along with their corresponding radium equivalent activity (Ra_{eq}), annual effective dose (D_{eff}), radiation absorbed dose rate (D), external radiation hazard indices (H_{ex}).

Site	Activity concentration (Bq/Kg)			Ra_{eq} (Bq/Kg)	D_{eff} (mS/y)	D (nGy/h)	H_{ex}
	^{226}Ra	^{232}Th	^{40}K				
Thoubal Bazar	29 ± 7	38 ± 8	451 ± 111	118.1	0.3	56.9	0.3
Heirok	26 ± 3	56 ± 8	861 ± 112	172.4	0.5	85.2	0.5
Kakching Bazar	35 ± 4	72 ± 6	925 ± 98	209.2	0.6	102.4	0.6
Chairen	30 ± 3	69 ± 6	879 ± 118	196.4	0.6	96.3	0.5
Toupokpi	38 ± 6	71 ± 11	734 ± 79	196.0	0.6	94.8	0.5
Pukahao Bazar	58 ± 7	70 ± 12	491 ± 104	195.9	0.6	92.2	0.5
Lamlai Bazar	121 ± 3	204 ± 5	1819 ± 112	552.8	1.6	264.9	1.5
Porompat	132 ± 11	212 ± 20	1012 ± 245	513.1	1.5	240.2	1.4
Andro Park	100 ± 7	85 ± 8	887 ± 87	289.8	0.8	137.1	0.8
Yairipok Khoirom	127 ± 3	22 ± 7	891 ± 101	227.1	0.7	107.1	0.6
Min	26	22	451	118.1	0.3	56.9	0.3
Max	132	212	1819	552.8	1.6	264.9	1.5
Mean	70	90	895	267.1	0.8	127.7	0.7
Stdev	45	65	374	146.8	0.4	69.0	0.4

Table 3: A review of radiological survey in some countries.

Region	Activity concentration (Bq/Kg)				Ref.
	^{226}Ra	^{232}Th	^{40}K	D (nGy/h)	
India	41(14-160)	29(7-81)	400(38-760)	53.14*	[4]
China	32(2-440)	41(1-360)	440(9-1800)	57.90*	[4]
Pakistan(north western area)	24.5	43.2	508.8	58.63*	[18]
Bangladesh(Building materials)	14	25	158	28.16*	[19]
Hong Kong (Building materials)	24	27	842	62.51*	[20]
Malaysia(Johor – H. Lunas soil)	78.1 ± 8.9	410.5 ± 55.4	56.4 ± 8.8	286.38*	[21]
Malaysia (Johor – Pontian district)	68 ± 6	113 ± 9	683 ± 29	128.15*	[22]
Malaysia (Kedah – agricultural soil	102.1 ± 4.0	134.0 ± 2.9	325.9 ± 9.8	141.6	[23]
Saudi Arabia (Al-Rakkah)	23 ± 1.6	20 ± 1.4	233 ± 12	32.69	[24]
Saudi Arabia (Ad-Dahna Desert)	23.4 ± 4.3	29.7 ± 5.9	380 ± 65	44.60*	[25]
Saudi Arabia (Al-Qassim)	9.5 ± 2.8	12.6 ± 3.3	546 ± 23	35.2	[1]
Iran (Bushehr)	35	17	285	38.32*	[26]
Iran (N-W coast, Persian Gulf)	35	26	395	48.35*	[27]
Turkey (sand)	44	26	441	54.42*	[8]
United State	40(8-160)	35(4-130)	370(100-770)	55.05*	[4]
World average	32	45	420	59.48*	[4]
Present study	**70**	**90**	**895**	127.7	

*Evaluated the absorbed dose rate using eqn (3).

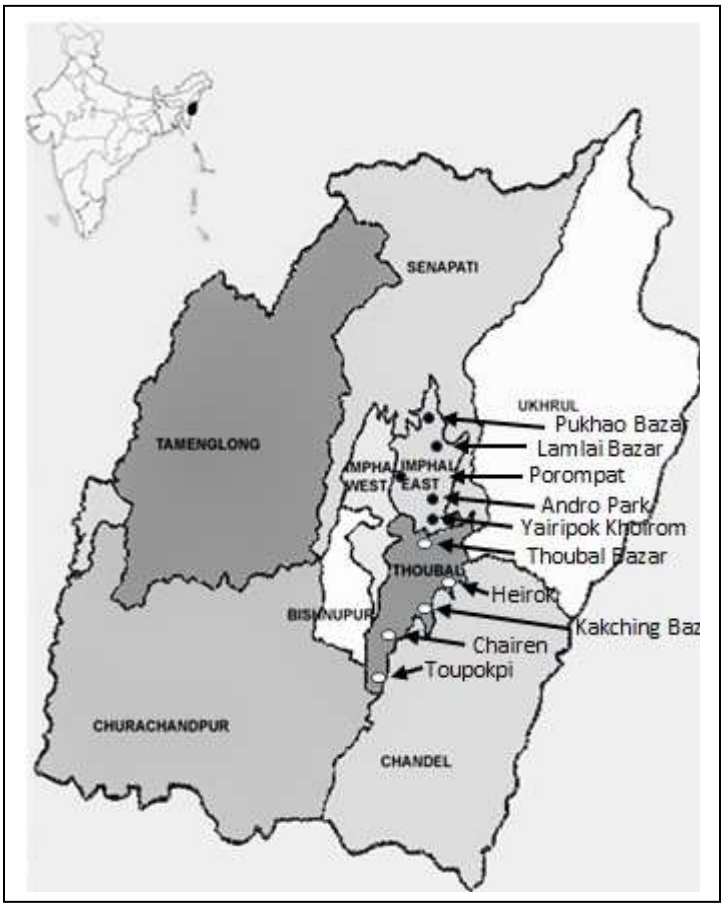

Figure 1: Map of Manipur and the radionuclide assessment districts

Solid circles – study sites in Imphal East district

Hollow circles – study sites in Thoubal district