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Paper

Assessment of radiation dose due to ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po in groundwater of Kodagu district, India

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Abstract

Natural radionuclides are universally spread and can be found in varying levels in rock, soil and water depending on the geology. A potential health threat may be caused by them to humans on consumption of water, food and inhalation of air due to the presence of radionuclides. In the present study, an attempt has been made to study the distribution of ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po in groundwater samples of Kodagu district, India. The activity concentrations of ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po were found to vary from 0.44 to 8.81 μg L^{-1}, 0.71 to 7.66 mBq L^{-1}, 1.54 to 9.61 Bq L^{-1} and 0.47 to 4.35 mBq L^{-1}, respectively. The associated dose due to radiation was assessed and was observed to be below the recommended standards. The total effective dose to the population was calculated and was found to be less than the recommended WHO standard of 100 mSv.

Introduction

Radioactivity is universal and radionuclides can be from terrestrial, cosmic or anthropogenic origin. The natural radioactive elements are found in soil, water, rocks and air in varying levels. Uranium may be found in almost every form of rock and soil in varying levels and its concentration is generally high in igneous rocks. The primary mineral source of uranium, Pitchblende, is a main component of igneous, granites and pegmatites rocks. The chemical composition of the underground water, its residence duration in the soil and bedrock, the lithological, hydrogeological and geochemical composition of the soil and rock all affect the amount of uranium in the groundwater[(1)].

Ingestion of water-soluble uranium compounds are found to cause renal dysfunction than exposure to insoluble uranium compounds at lower concentrations[(2)]. Uranium and its daughter products ^{226}Ra, ^{222}Rn and ^{210}Po may cause adverse and potential health risk to the inhabitants. The primary aspect of uranium toxicity to humans is chemical toxicity, which is affected by parameters such as uranium concentration, duration of exposure, pH, temperature, etc[(3)].

Among the naturally occurring radioactive elements, ^{226}Ra and its daughter products are important contributors to the radiation dose to the public[(4)]. In contrast to uranium, radium is water-soluble and can leach into groundwater from the surrounding rocks. Nearly 20% of the consumed radium is absorbed, transported to the bloodstream, and is primarily accumulated in the bones and may cause bone cancer[(5)].

^{222}Rn with a half life of 3.82 d is produced by the alpha decay of ^{226}Ra and contributes >50% of the radiation dose received by the population[(6)]. Radon typically enters groundwater from rock by diffusion through crystalline lattices, fissures and crystal boundaries and is used as a tracer in various geological and hydrogeological applications[(7)]. Radon is known to

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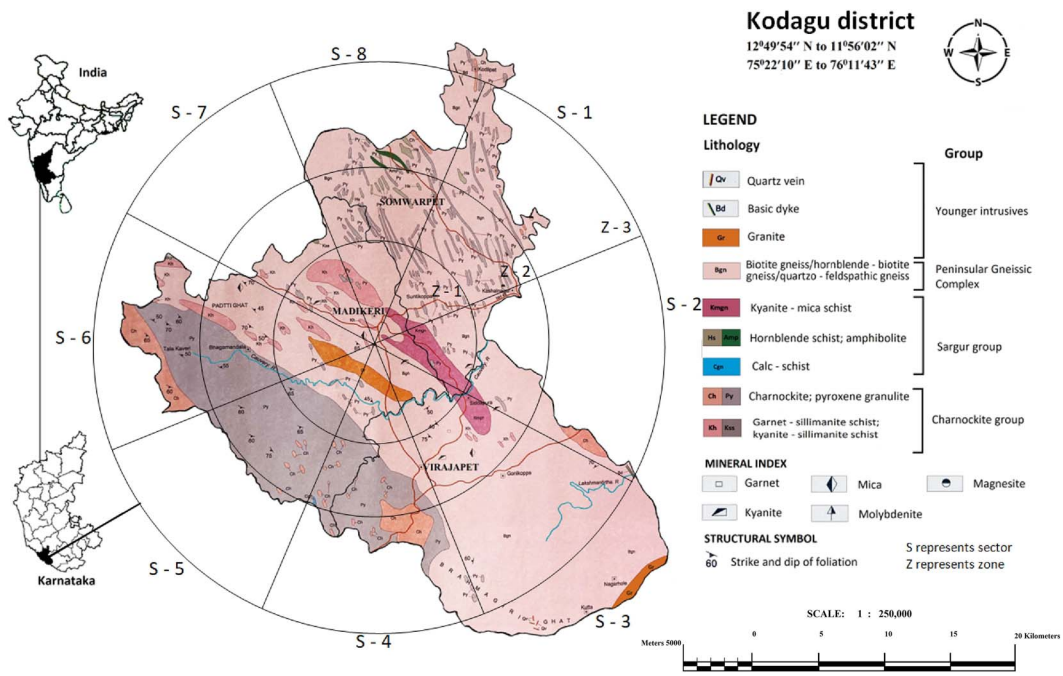

Figure 1. The study area.

be the second-largest cause of lung cancer, the first being smoking. The noble gas radon in drinking water enters living organisms via ingestion and inhalation. Therefore, ^{222}Rn poses a potential risk of radiation hazard on continuous exposure.

^{210}Po, a radioactive element with half life of 138.376 d decays by emitting alpha particle (5.407 MeV) having higher ionising power, causing chronic health issues upon ingestion. It is highly toxic causing direct damage to the tissues where it gets absorbed. In addition to being dissolved in groundwater, polonium is also found to be associated with colloids. This is due to its highly reactive behaviour with particles. Compounds of manganese and iron have high adsorption coefficients for polonium[8].

Radiation doses due to natural radionuclides in groundwater are higher compared with the doses from artificial sources[9]. The population of the present study area highly relies on groundwater for irrigation and all other domestic purposes. Hence, monitoring the levels of radioactive elements in water becomes crucial. Groundwater samples were collected from bore wells located in different terrain covering the entire district during all the seasons of a year.

Study area

Kodagu District, India, the study region spreads over 4102 km^2 and is a part of the Western Ghats of India (Figure 1). The district comprises archean base biotite, granites, gneisses, charnockites and amphibolites forming a part of hard rock terrain. A distinct litho assemblage of sargur group occurs as an array of enclaves of varied dimensions within peninsular gneissic complex. In the eastern and southern parts of the study area, pegmatite veins and dolerite dykes are common intrusive. Dolerite dykes appear as separated boulders and as intrusive in the granitic formations. Weathered zones of granites and gneisses occur between the depths of 2–25 m below ground level (bgl). Shallow aquifers and prolific deeper aquifers of jointed and fractured granite and gneisses occur between the depths of 25–150 m bgl. Recharge of groundwater takes place through the infiltration of rainwater. Due to the nature of rocks and soil in this region, the study of activity of radionuclides in groundwater samples becomes essential. For the purpose of analysis, study area was divided into three zones of 15, 30 and 55 km radius from the centre and eight sectors of 45°.

Materials and methods

Measurement of ^{238}U in water sample

Water samples were collected from various bore wells of the study locations in acid cleaned bottles to avoid uranium adsorption on walls. LED Fluorimeter (Figure 2) was employed to measure the activity concentration of uranium (A_U)[3, 10]. The U concentration

Figure 2. LED fluorimeter.

in groundwater (A_U) was calculated using equation (1).

$$A_U\ (\mu g\ L^{-1}) = \frac{F_1}{F_2 - F_1}\left(\frac{V_1 C}{V_2}\right) \quad (1)$$

Where,
F_1 = total counts due to fluorescence of the sample,
F_2 = total counts due to sample + added uranium standard, V_1 = volume of added uranium standard (mL),
V_2 = volume of sample (mL), C = concentration of added uranium standard ($\mu g\ L^{-1}$).

Estimation of radiation dose and cancer risk due to ^{238}U in water

Age—dependent ingestion dose was estimated due to water intake (WI) of uranium through drinking water pathway following the IAEA dose coefficients[11]. The total effective radiation dose was calculated for an individual who drinks an average of 730 L of water per year using the dose conversion factor (DCF) of 4.5×10^{-8} Sv Bq^{-1} for ^{238}U.

The annual radiation ingestion dose due to uranium was calculated using equation (2).

$$D_U = A_U\ (Bq\ L^{-1}) \times WI\ (L\ y^{-1}) \times DCF\ (Sv\ Bq^{-1}) \quad (2)$$

The lifetime cancer risk (CR) due to uranium in water was estimated using equation (3)[12].

$$CR = A_U\ (Bq\ L^{-1}) \times \text{Risk factor}\ (5.65 \times 10^{-5}/Bq\ L^{-1}) \quad (3)$$

Where,
A_U (Bq L^{-1}) = measured value of ^{238}U ($\mu g\ L^{-1}$) × Conversion factor (0.0248 Bq μg^{-1}).
Risk factor (per Bq L^{-1}) = Risk coefficient (4.40×10^{-11} per pCi) × WI (2 L d^{-1}) × Total exposure duration (23,725 d) × Conversion factor (27 pCi Bq^{-1})[12].

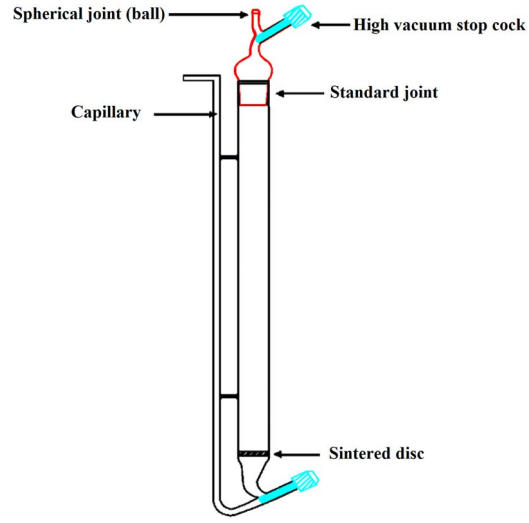

Figure 3. Schematic representation of radon bubbler unit.

Measurement of ^{226}Ra in water sample

The activity concentration of ^{226}Ra in groundwater was studied using emanometry method. A total of 20 L of water was collected from various bore wells from the study area. The collected water samples were pre-concentrated in the laboratory following the procedure available in literature[13–15]. A total of 70 mL of concentrated solution was transferred into a pre-evacuated radon bubbler (Figure 3), sealed and allowed to stand for ~21 d[14, 15]. The radon built up in the bubbler was transferred to an evacuated scintillation cell by agitating the solution. The alpha activity was measured using the alpha probe coupled with a programmable counter. Using equation (4) the activity concentration of ^{226}Ra dissolved in the sample was determined[14].

$$A_{Ra}\ (mBq\ L^{-1}) = \frac{6.97 \times 10^{-2} \times D}{V \times E \times (e^{-\lambda T}) \times (1 - e^{-\lambda \theta}) \times (1 - e^{-\lambda t})} \quad (4)$$

Where,
D = alpha counts due to sample – background counts, V = volume of water sample (20 L), E = efficiency of the alpha scintillation cell (74%), λ = radioactive decay constant of ^{222}Rn (2.098×10^{-6} s^{-1}), T = counting lag after collecting water sample (~1 d), t = duration of alpha counting (15 min) and θ = duration of ^{222}Rn build up in the bubbler (21 d).

Estimation of radiation dose due to ^{226}Ra in water

Annual ingestion dose due to consumption of ^{226}Ra through drinking water was calculated using equation (5)[6].

$$D_{Ra} = WI\ (L\ y^{-1}) \times DCF\ (Sv\ Bq^{-1}) \times A_{Ra} \quad (5)$$

Where,
WI = 730 L y^{-1}, DCF = 2.8×10^{-7} Sv Bq^{-1}, is the DCF for ^{226}Ra.

Measurement of ^{222}Rn in water sample

Water samples from different bore wells of the study area were collected in airtight bottles such that no air bubbles were formed. Sampling date and time was noted and the activity concentration of ^{222}Rn was measured employing a Smart Radon Monitor (SRM). The concentration of ^{222}Rn in water was measured using the equation (6) adopted from Raghavayya *et al.* with required modification[14].

$$A_{Rn}\left(\text{Bq L}^{-1}\right) = \frac{6.97 \times 10^{-2} \times D}{V \times E \times \left(e^{-\lambda T}\right) \times \left(1 - e^{-\lambda t}\right)} \quad (6)$$

Where,
D = sample counts—background counts, V = volume of water (60 mL), E = efficiency of the alpha scintillation cell (74%), t = counting duration, λ = radioactive decay constant of ^{222}Rn (2.098×10^{-6} s^{-1}), T = counting lag after collecting the sample.

Estimation of total radiation dose due to ^{222}Rn in water

The inhalation and ingestion dose due to ^{222}Rn in drinking water was calculated using equations (7) and (8), respectively[6].

$$D_{inh} = A_{Rn} \times R_{aw} \times F \times I \times DCF \quad (7)$$

Where,
R_{aw} = ratio of ^{222}Rn in air to water [4–10], F = equilibrium factor between ^{222}Rn and its progeny (0.4), I = average indoor occupancy time per individual (7000 h y^{-1}), DCF = dose conversion factor for ^{222}Rn exposure (9 nSv (Bq h m^{-3}) $^{-1}$)[6].

$$D_{ing} = A_{Rn} \times WI \times EDC \quad (8)$$

Where,
EDC = effective dose coefficient for ingestion (3.5 nSv Bq^{-1} L^{-1}), and WI = weighted estimate of water intake (60 L y^{-1})[6].

Measurement of ^{210}Po in water sample

The water samples from different bore wells of the study area were collected in preconditioned containers. The activity of ^{210}Po in groundwater samples in the study area was below the detection level of the alpha counting system. Therefore, ~20 L of water samples were collected and pre-concentration of ^{210}Po was carried out using the method available in literature[16]. A silver planchet of 2.5 cm diameter was introduced into 500 mL of pre-concentrated solution. The ^{210}Po was electrochemically deposited on a silver planchet and the electroplated planchet was used to analyse the activity of ^{210}Po employing an alpha counting system. The activity concentration of ^{210}Po was calculated using equation (9)[17].

$$A_{Po}\left(\text{mBq L}^{-1}\right) = C \times \frac{100}{\varepsilon} \times \frac{100}{E_P} \times \frac{1000}{V} \quad (9)$$

Where,
C = net count rate (counts s^{-1}), ε = efficiency of the alpha counting system (%), E_P = deposition of ^{210}Po on silver planchet (%), V = volume of sample (L).

Estimation of radiation dose due to ^{210}Po in water

The annual effective dose due to the ingestion of ^{210}Po to an individual through drinking water was estimated using equation (10)[17].

$$D_{Po} = A_{Po} \times WI \times D_c \quad (10)$$

Where,
D_{Po} = annual effective dose, WI = annual intake of drinking water (730 L) and $D_c = 1.2 \times 10^{-3}$ mSv Bq^{-1} is the ingestion DCF for ^{210}Po, based on the report of ICRP[18, 19].

Results and discussion

Groundwater samples from various locations of the study area were collected to study the concentrations of ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po. About 15–30 samples were collected from each sector for the purpose. The concentration of ^{238}U was found to vary from 0.44 to 8.81 μg L^{-1} with an average of 3.28 μg L^{-1} (Table 1). The measured ^{238}U concentration was found to be lower compared with the recommended standard guideline level of 30and 60 μg L^{-1} by WHO and Atomic Energy Regulatory Board (AERB), respectively[19, 20]. The radiation dose due to ^{238}U in water for an individual consuming 730 L of water per year was calculated, and was found to vary from 0.33 to 7.17 μSv y^{-1} with an average of 2.64 μSv y^{-1}. The lifetime CR was calculated from the measured ^{238}U concentration and was found to vary from 0.62×10^{-6} to 12.65×10^{-6}, this was observed to be lower than 10^{-3}, the acceptable value of lifetime CR. The age-dependent ingestion dose due to uranium in water was estimated and is shown in Table 2. It was observed that the dose to infants of age 7–12 months was higher in comparison with children and adults of other age groups, and the dose to male adults was higher than female adults (Figure 4). This variation depends on the intake of water and also the dose conversion coefficient used. The average ^{238}U concentration was observed to be higher in sectors 1, 2 and 3 compared with other sectors where the major region of sectors 5, 6 and 7.

The ^{226}Ra concentration in the study area was found to vary from 0.71 to 7.66 mBq L^{-1} with an average of 3.04 mBq L^{-1} (Table 1). The ^{226}Ra activity concentration in the study region was found to be lower than the standard safe level of 1 Bq L^{-1} recommended by IAEA and WHO[19, 21]. The radiation dose associated with radium in water was assessed and was found to vary

Table 1. Activity concentration and the estimated radiation dose due to ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po in groundwater.

Sector	A_U	A_{Ra}	A_{Rn}	A_{Po}	Radiation dose (μSv y^{-1})				Total eff. dose
	μg L^{-1}	mBq L^{-1}	Bq L^{-1}	mBq L^{-1}	U	^{226}Ra	^{222}Rn	^{210}Po	μSv y^{-1}
1	8.81	7.66	8.45	3.64	7.17	1.56	23.06	3.19	34.99
2	6.54	5.41	5.33	4.35	5.29	1.10	14.55	3.82	24.79
3	5.37	4.37	9.61	3.41	4.32	0.89	26.23	2.99	34.48
4	1.61	1.78	1.75	1.16	1.3	0.36	4.77	1.02	7.46
5	0.89	0.71	1.61	0.47	0.65	0.14	4.39	0.42	5.67
6	0.44	0.88	1.54	1.35	0.33	0.17	4.20	1.19	5.91
7	0.85	0.94	2.56	0.52	0.65	0.19	6.99	0.46	8.32
8	1.76	2.56	5.69	0.85	1.38	0.52	15.53	0.75	18.23
Min.	0.44	0.71	1.54	0.47	0.33	0.14	4.20	0.42	5.67
Max.	8.81	7.66	9.61	4.35	7.17	1.56	26.23	3.82	34.99
GM	1.68	2.17	3.94	1.25	1.34	0.44	10.76	1.10	13.27
Average	3.28	3.04	4.57	1.97	2.64	0.61	12.46	1.73	17.48
SD	3.17	2.54	3.21	1.57	2.59	0.51	8.76	1.37	12.57

GM, geometric mean; SD, standard deviation.

Table 2. Age-dependent ingestion dose due to ^{238}U in groundwater.

Sector	Lifetime cancer risk	Age-dependent ingestion dose (μSv y^{-1})									
		Infants (months)		Children (years)		Adult female (years)			Adult male (years)		
	10^{-6}	0–6	7–12	1–3	4–8	9–13	14–18	>18	9–13	14–18	>18
1	12.35	18.99	21.70	12.45	10.85	11.39	12.29	9.69	13.02	17.64	13.28
2	9.17	14.09	16.11	9.24	8.06	8.46	9.13	7.20	9.67	13.09	9.86
3	7.53	11.57	13.23	7.59	6.62	6.95	7.50	5.91	7.94	10.75	8.10
4	2.26	3.47	3.97	2.28	1.99	2.09	2.25	1.78	2.38	3.23	2.43
5	1.25	1.92	2.20	1.26	1.10	1.16	1.25	0.98	1.32	1.79	1.35
6	0.62	0.95	1.09	0.63	0.55	0.57	0.62	0.49	0.66	0.89	0.67
7	1.20	1.84	2.10	1.21	1.05	1.10	1.19	0.94	1.26	1.71	1.29
8	2.47	3.80	4.34	2.49	2.17	2.28	2.46	1.94	2.61	3.53	2.66
Min.	0.62	0.95	1.09	0.63	0.55	0.57	0.62	0.49	0.66	0.890	0.670
Max.	12.35	18.99	21.70	12.45	10.85	11.39	12.29	9.69	13.02	17.64	13.28
GM	2.36	3.63	4.15	2.38	2.08	2.18	2.35	1.86	2.49	3.38	2.54
Average	4.61	7.08	8.09	4.64	4.04	4.25	4.58	3.61	4.85	6.57	4.95
SD	4.44	6.83	7.80	4.47	3.90	4.09	4.42	3.48	4.68	6.34	4.77

GM, geometric mean; SD, standard deviation.

from 0.14–1.56 μSv y^{-1} with an average of 0.61 μSv y^{-1} and was within the WHO recommended standard guideline of 100 mSv[9].

The activity concentration of ^{222}Rn was found to vary from 1.54 to 9.61 Bq L^{-1} with an average of 4.75 Bq L^{-1} (Table 1). The dose due to inhalation and ingestion due to ^{222}Rn was calculated and the total radiation dose in drinking water was observed to be varying from 4.20 to 26.23 μSv y^{-1}. However, the observed concentration and total effective dose was lower compared with 100 Bq L^{-1} and 0.1 mSv standard recommended by WHO[9].

The activity concentration of ^{210}Po was observed to vary from 0.47 to 4.35 mBq L^{-1} with an average of 1.97 mBq L^{-1} (Table 1). The concentration of ^{210}Po was lower compared with 100 mBq L^{-1}, prescribed safe level by WHO[9]. The radiation dose due to ingestion of ^{210}Po through the drinking water pathway was between 0.42 and 3.82 μSv y^{-1}.The annual effective dose due to ^{210}Po to the inhabitants of the study area was found to be lower compared with the standard limit of 0.01 mSv y^{-1} (WHO), 0.12 mSv y^{-1} (UNSCEAR) and 1.0 mSv y^{-1} (ICRP)[6, 9, 22].

Concentration of ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po in groundwater samples from different sectors of the study area is shown in Figure 5. The total effective radiation dose due ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po in each sector was estimated and is represented in Table 1. The effective radiation dose to the population was found to be well below the recommended standards

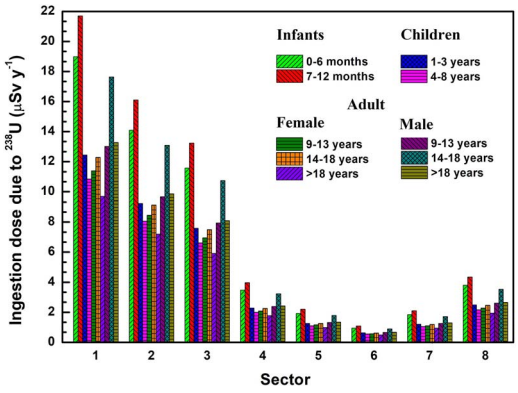

Figure 4. Age-dependent ingestion dose due to ^{238}U in groundwater.

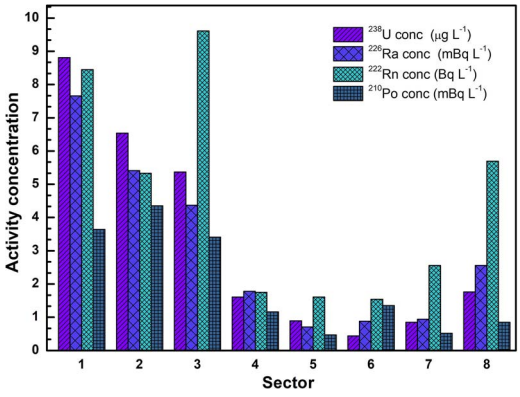

Figure 5. Activity concentration of ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po in different sectors of the study area.

of 100 mSv y^{-1}. The lithology of Kodagu district shows that the major area of sectors 5 and 6, and a few regions of 1 and 8 are comprised charnockite rock structure. The study confirms that the levels of radionuclides ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po in groundwater samples of Kodagu district, India are lower than the permissible limits and the population of the study region is minimally affected by them.

Conclusions

The activity concentration of radionuclides like ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po in groundwater samples was measured systematically in Kodagu district, India and their associated radiation dose was assessed. The concentrations of ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po varied from 0.44 to 8.81 μg L^{-1}, 0.71 to 7.66 mBq L^{-1}, 1.54 to 9.61 Bq L^{-1} and 0.47 to 4.35 mBq L^{-1}, respectively. The activity of all measured radioactive elements was found to be lower than the recommended standard levels. The radiation dose due to the distribution of ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po in groundwater was assessed and was found to be lower in comparison with the standard value. The investigation concludes that the inhabitants of the study area are very less prone to radiation dose from ^{238}U, ^{226}Ra, ^{222}Rn and ^{210}Po, and are safe from radiological hazard.

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