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Paper

Distribution of U and ^{210}Po in groundwater of Kodagu district, Karnataka, India

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Abstract

Trace amounts of uranium along with its decay products are found in varying levels in natural soil, rocks, water and air. They are a matter of significant concern due to their carcinogenic nature. In the present work, the distribution of U and ^{210}Po in groundwater of Kodagu District, Karnataka, India, was studied. The concentration of total U in groundwater samples was estimated using LASER and LED fluorimeter, and the activity of ^{210}Po in groundwater was studied using electrochemical deposition followed by alpha counting method. The concentration of U and ^{210}Po varied from 0.4 to 8.8 μgl^{-1} and 0.47 to 4.35 mBql^{-1}, respectively. The ingestion dose due to U and ^{210}Po in groundwater varied from 0.33 to 7.17 and 0.41 to 3.81 μSv y^{-1}, respectively. The estimated U activity was found to be well below the standard safe limits of 30 μgl^{-1} in drinking water, as recommended by WHO and USEPA. The ^{210}Po activity was low compared with the recommended 100 mBql^{-1} standard of WHO.

Introduction

Soil, rocks and water in the environment contain trace amount of natural radioactive elements. The living organisms on being exposed to ionising radiations may experience potential health risk. Human exposure to ionising radiation from natural sources is continuous and inevitable process on Earth[(1)]. Terrestrial sources of radioactive elements are the major contributors to the effective radiation dose received by the population, and 2.4 mSv per year remains the worldwide average exposure from natural radiation sources to mankind[(1)].

The average concentration of uranium in the earth's crust is 0.0003% and it is found in water, soil and rocks in varying level. The activity of ^{226}Ra and ^{232}Th are found to be higher in fractured granitic-gneisses, gneisses and hornblende schists type of rocks[(2)]. In groundwater, uranium is present both in dissolved and particulate forms due to minerals like uraninite, pitchblende and cornalite or as secondary mineral in the form of complex oxides of silicates, phosphates, vanadates, lignite and monazite sands. Apart from natural sources, phosphate fertilisers used in agricultural land is another important source of contamination of uranium[(3)].

Nearly 85% of uranium enters living beings through water and is nephrotoxic, primarily causing renal dysfunction[(4)]. Also, ^{238}U can deposit and accumulate in bones for about a year.

^{210}Po, a decay product of ^{238}U series is released to the atmosphere and can enter into living organism through food and water pathway. Alpha particle emitted by ^{210}Po causes direct damage to the tissues. Despite the short range of alpha particles of about 40–50 μm in tissues, doses from ^{210}Po are generally assumed to be delivered uniformly to the organs/tissues in which the radionuclide is retained[(5)]. Toxicity of ^{210}Po relies upon its chemical characteristics and is about 250 000 times more than that of hydrogen cyanide and can be carcinogenic on ingestion[(6)]. Ingestion of micrograms of ^{210}Po will likely to be fatal to all exposed persons, which damages the bone marrow and other internal organs[(6)].

In different parts of India, several researchers have carried out studies on concentration of uranium and ^{210}Po in ground water samples during the last few decades. A higher concentration of uranium in water was found in mining areas. In Bagjata, uranium mining area its concentration was in the range < 12.6–693 mBql^{-1} [(7)]. The concentration of ^{238}U in water samples

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of Haryana State, India, showed variations from 0.10 to 223.16 μgl^{-1}[8]. A lower concentration of ^{238}U in water samples was observed in places like Chamarajanagar district, Karnataka, India, which showed variations from 0.03 to 4.63 μgl^{-1}[9]. The average ^{210}Po activity of 0.75 $mBql^{-1}$ was reported in the water samples of Koraiyar river[10]. In ground water of Cauvery river basin of Karnataka State ^{210}Po activity concentration was reported in the range from 1.89 to 4.18 $mBql^{-1}$[11].

Major studies on the distribution of ^{210}Po has been carried out in marine ecosystem and dietary sources, whereas relatively less attempts have been made in studying the distribution in fresh water ecosystem and in underground sources. In the present study, an attempt has been made to find the distribution of U and ^{210}Po in the groundwater of Kodagu District, Karnataka State, India. The water samples were collected from bore wells located in different terrain covering the entire district during all the seasons of a year. This type of extensive and systematic study was carried out for the first time in the Kodagu District, India.

Study area

The study region, Kodagu District, Karnataka, India, occupies an area of 4102 km^2 with a population of about 0.56 million (Figure 1). The district comprises of 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. Weathered zones of granites and gneisses occur between the depths of 2–25 m below the 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. The district is characterised by slight to high humidity and temperature ranging from 15 to 32 °C and the region receives an average annual rainfall of around 2800 mm. Groundwater is the main source of water for consumption, irrigation and other domestic activities throughout the study area. 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, the study area was divided into three sectors of 15-, 30- and 60-km radius and eight zones.

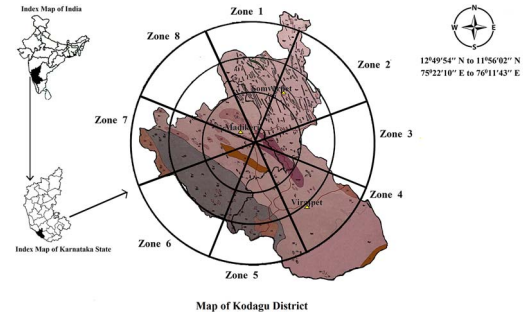

Figure 1. The study area.

Materials and methods

Uranium concentration in water

The activity concentration of uranium in ground water samples was measured employing LASER and LED Fluorimeter in the Department of Physics, University of Mysore, India. About 100 mL of groundwater sample was collected from different bore wells from the study area in clean polyethylene containers. The samples were acidified in order to avoid uranium precipitation and adsorption on the walls of the container during transportation and storage. About 2 mL of water sample was taken for analysis and 0.5 mL of fluran was added to it and shaken well. The sample was then fed into the fluorimeter and the number of counts was noted. A reagent blank was prepared likewise without the water sample and the background counts were noted. The instrument was calibrated using the uranium standard solution of known strength. Activity concentration of some samples was validated using a LASER Fluorimeter at CARER, Mangalore University and a LED Fluorimeter at RMP, BARC, Mysuru. The uranium concentration (A_U) in water samples was estimated using equation (1):

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

where F_1 is the total counts due to fluorescence of sample, F_2 is the total counts due to sample and added uranium standard, V_1 is the volume of added uranium standard (ml), V_2 is the volume of sample (ml) and C is the concentration of added uranium standard (μgl^{-1}).

Dose estimation

Ingestion dose due to uranium through drinking water pathway was estimated for different age groups according to the Water Intake (WI) rates of 0.8, 1.7, 2.7 and 3.7 L per day for infants, children, female adults and male adults, respectively. Dose coefficients of 3.4×10^{-7} for infants, 8×10^{-8} for children and 4.5×10^{-8} $SvBq^{-1}$ for female and male adults were used according to the IAEA safety standards[12]. Total effective radiation dose was calculated considering an average of 730 L of water consumption by an

individual per year. Dose conversion factor (DCF) equal to 4.5×10^{-8} $SvBq^{-1}$ was used. The annual radiation dose due to uranium intake through drinking water pathway was calculated using equation (2):

$$D\ \left(Svy^{-1}\right) = A_U\ \left(Bql^{-1}\right) \times WI\ \left(ly^{-1}\right) \times DCF\ \left(SvBq^{-1}\right) \quad (2)$$

Radiological risk assessment

The excess cancer risk (ECR) was calculated using the standard method adopted by EPA as shown in the equation (3)[13]:

$$ECR = A_U\ \left(Bql^{-1}\right) \times \text{Risk factor}\ \left(5.65 \times 10^{-5}/Bql^{-1}\right) \quad (3)$$

where A_U (Bql^{-1}) = Measured value (μgl^{-1}) × Conversion factor (0.0248 Bq μg^{-1}).

Risk factor (per Bql^{-1}) = Risk coefficient (4.40×10^{-11} per pCi) × WI (2 ld^{-1}) × Total exposure duration (23,725 days) × Conversion factor (27 pCi Bq^{-1}).

^{210}Po concentration in water

About 10 L of samples were collected from different bore wells of the study area in preconditioned polyethylene containers and brought to the laboratory. Samples were filtered through Whatman 42 filter paper and acidified below pH 2 using HCl. For pre-concentration of ^{210}Po, Ferric Chloride as Fe(III) carrier was added to 10 L of filtered and acidified water sample and agitated for an hour for uniform mixing. The pH of the sample was adjusted to 9 using 25% liquid ammonia solution to precipitate Fe as $Fe(OH)_3$. The precipitate solution was stirred for about 6 hours and was left to settle, the supernatant solution was discarded and the settled precipitate was dissolved in HCl (Conc). A 30% H_2O_2 solution was added to it in order to remove the organic matter. The sample was stirred continuously at 90 °C to completely evaporate it to dryness. The obtained residue was dissolved in 0.5 N HCl and a silver disc of 25 mm diameter and 2 mm thickness was introduced in to the solution. Ascorbic acid was added to restrict the interference due to Ferric ion deposition on silver disc. The solution is agitated for a period of 6 hours to get ^{210}Po deposited on the silver disc[15]. The electroplated Ag disc (Figure 2) was used to analyse the activity of ^{210}Po using an alpha counting system. Activity concentration of ^{210}Po was calculated using

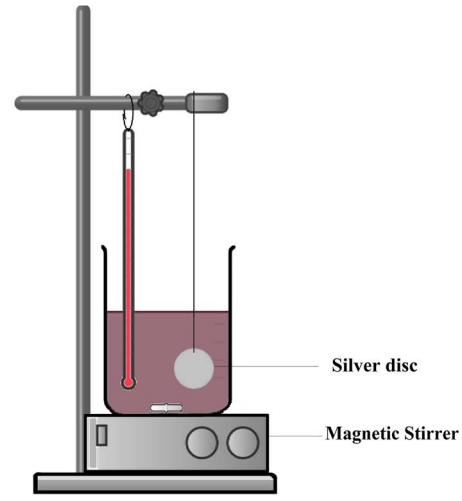

Figure 2. Schematic representation of ^{210}Po electro-deposition.

equation (4):

$$A_{Po}\left(mBql^{-1}\right) = C \times \frac{100}{E} \times \frac{100}{E_P} \times \frac{1000}{V} \quad (4)$$

where A_{Po} is Activity concentration of ^{210}Po ($mBql^{-1}$), C is the net count rate (Counts s^{-1}), E (%) is the efficiency of the alpha counting system, E_P is the % deposition of ^{210}Po on Ag disc, V is the volume of sample (l).

Annual effective dose due to ^{210}Po

The annual effective dose to an individual due to the ingestion of ^{210}Po through drinking water pathway was estimated using equation (5)[15]:

$$D_w = A_{Po} \times WI \times D_c \quad (5)$$

where D_w = Annual effective dose ($mSv\ y^{-1}$), A_{Po} = Activity of ^{210}Po (Bql^{-1}), WI = Annual intake of drinking water (730 l/year) and D_c = Ingestion dose conversion factor for ^{210}Po ($1.2 \times 10^{-3} mSvBq^{-1}$) based on the report of ICRP[14, 16].

Results and discussion

Groundwater samples from various sectors and zones of the study area were collected and concentration of U and ^{210}Po were analyzed. Around 30–50 ground water samples from each zone was collected to analyse the uranium activity in water and about 20 samples from each zone was collected for studying the ^{210}Po activity in water. The activity concentration, age

Table 1. Activity of U and ^{210}Po and dose rate.

Location	Conc. of U	Effective ingestion dose due to U	Age dependent dose to public due to U				ECR	Conc. of ^{210}Po	Ingestion dose due to ^{210}Po	Effective dose due to U and ^{210}Po
			Infants	Children	Adult female	Adult male				
	(μgl $^{-1}$)	(μSv y $^{-1}$)	(μSv y $^{-1}$)				(10^{-06})	(m Bql $^{-1}$)	(μSv y $^{-1}$)	(μSv y $^{-1}$)
Zone 1	8.8	7.17	21.67	10.84	9.68	13.27	12.34	3.64	3.18	10.36
Zone 2	6.5	5.30	16.01	8.01	7.15	9.80	9.11	4.35	3.81	9.12
Zone 3	5.3	4.32	13.05	6.53	5.83	7.99	7.43	3.41	2.98	7.31
Zone 4	1.6	1.31	3.94	1.97	1.76	2.42	2.25	1.16	1.01	2.33
Zone 5	0.8	0.66	1.97	0.99	0.88	1.21	1.13	0.47	0.41	1.08
Zone 6	0.4	0.33	0.99	0.50	0.44	0.61	0.57	1.35	1.18	1.52
Zone 7	0.8	0.66	1.97	0.99	0.88	1.21	1.13	0.52	0.45	1.12
Zone 8	1.7	1.39	4.19	2.10	1.87	2.57	2.39	0.85	0.74	2.14
Min.	0.4	0.33	0.99	0.50	0.44	0.61	0.57	0.47	0.41	1.08
Max.	8.8	7.17	21.67	10.84	9.68	13.27	12.34	4.35	3.81	10.36
Avg.	3.24	2.64	7.97	3.99	3.56	4.89	4.54	1.96	1.72	4.37
GM	1.95	1.60	4.81	2.41	2.15	2.95	2.75	1.43	1.25	2.98
SD	3.18	2.59	7.83	3.92	3.50	4.79	4.45	1.56	1.37	3.89

dependent radiation dose and excess cancer risk due to U in groundwater of Kodagu District are shown in Table 1. Activity concentration of U varied from 0.4 to 8.8 μgl^{-1} with a geometric mean of 1.95 μgl^{-1}. The average U activity in groundwater collected from various zones showed very low concentrations compared with the standards recommended by WHO (30 μgl^{-1}), USEPA (30 μgl^{-1}) and AERB (60 μgl^{-1}) [16–18].

Water consumption level varies with factors like age, body weight, sex, environmental condition, physical activities, etc. Age-dependent radiation dose was estimated according to the IAEA standard dose coefficients and excess cancer risk to the public due to U in groundwater was calculated from the measured U concentration. The age dependent radiation dose due to U in groundwater was found to vary from 0.99 to 21.67 μSv y^{-1} with a geometric mean of 4.81 μSv y^{-1} in infants, 0.50 to 10.84 μSv y^{-1} with a geometric mean of 2.41 μSv y^{-1} in children, 0.44 to 9.68 μSv y^{-1} with a geometric mean of 2.15 μSvy^{-1} in adult female and 0.61 to 13.27 μSv y^{-1} with a geometric mean of 2.95 μSv y^{-1} in adult male.

It is observed that the radiation dose is higher in infants over children and also higher in adult males when compared with adult females, due to the variation in volume of water consumption and different dose conversion factors used. The estimated ECR was found to be varying from 0.57×10^{-6} to 12.34×10^{-6} and is very low compared with the recommended standards of 8.4×10^{-5} and 1.68×10^{-4} by WHO and AERB, respectively. Therefore, the population of the study area is at a very minimal level of carcinogenic risk due to uranium content in groundwater. Effective radiation dose due to U in water who drinks an average of 730 L of water per year was estimated and was found to vary from 0.33 to 7.17 μSv y^{-1}. However, the estimated effective dose per year is found to be less than the WHO standard of 0.1 mSv[16].

Activity concentration of ^{210}Po in the present study region varied from 0.47 to 4.35 mBql^{-1} with a geometric mean of 1.43 mBql^{-1} (Table 1). Radiation dose due to ingestion of ^{210}Po was estimated and it showed variations from 0.41 to 3.81 μSv y^{-1} with a geometric mean of 1.25 μSv y^{-1}. The effective annual dose due to ^{210}Po to the population was found to be low compared with standards of UNSCEAR (0.12 mSv y^{-1}), WHO (0.01 mSv y^{-1}) and ICRP (1.0 mSv y^{-1}). The total effective dose due to ingestion of U and ^{210}Po through drinking water pathway was found to vary from 1.08 to 10.36 μSv y^{-1} with a geometric mean of 2.98 μSv y^{-1}.

Comparison of uranium and ^{210}Po activity concentrations measured in different parts of the world is shown in Table 2. The present values are compared with the concentration of U from other parts India as reported in a nationwide survey conducted by Bhabha Atomic Research Centre (BARC), Mumbai, and is found to be low[19]. The activity concentration of ^{210}Po of the present study are comparable with the activity concentration of ^{210}Po in borewell water collected from Cauvery river basin region[11].

The average distribution of U and ^{210}Po in zones 1, 2 and 3 is found to be high compared with the other zones of the study area (Figure 3). This can be

Table 2. Comparison of U and ^{210}Po activity concentrations from different region of the world.

Region	Source		U	^{210}Po (mBql $^{-1}$)	References
Poland	Surface water	^{238}U (mBql^{-1})	2.75	0.12	(20)
	Deep water	^{238}U (mBql^{-1})	0.20	0.25	
Italy	Bottled water	^{238}U (mBql^{-1})	<0.17–89.00	<0.04–21.01	(21)
Southern Bulgaria	Drinking water	^{238}U (mBql^{-1})	1.4–1484	<0.3–13.6	(22)
Chamrajanagar, India	Borewell	U (μgl^{-1})	0.03–4.63	–	(9)
Bangalore, India	Groundwater	U (μgl^{-1})	0.136–2027.5	–	(23)
Cauvery river basin, India	Borewell	–	–	1.89–4.18	(11)
Nationwide survey, India	Surface water	U (μgl^{-1})	0.2–22	–	(19)
	Groundwater		0.2–4918		
Kodagu, India	Groundwater	U (μgl^{-1})	0.4–8.8	0.47–4.35	Present study

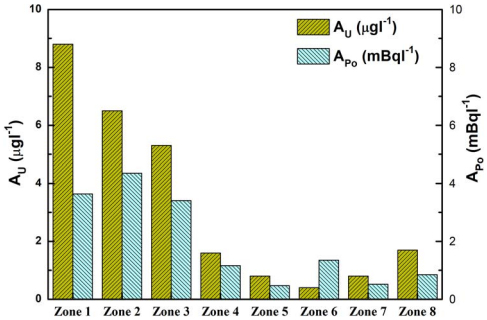

Figure 3. Variation of concentration of U and ^{210}Po in groundwater in different zones of study area.

attributed to the lithology of the zones which comprises of charnockite and pyroxene granulite type of rocks. Hence, varying concentration of U and ^{210}Po can be due to the composition of soil and rock of the study region. Although different types of rock and the soil structure make up the study area, the concentration of uranium in the groundwater also depends on the depth and the amount of uranium present in the host aquifer rock. Major regions of zones 1 and 2 are cultivation lands where various crops are grown seasonally; hence, the use of phosphate based fertilisers could also be one of the reasons for higher uranium activity compared with other parts of the study area. Other zones in the study area consist of either forest or coffee plantation. Rocks composed of phosphates contain higher concentrations of uranium and thorium and are attributed to show higher activity concentration of ^{210}Po.

Conclusions

A systematic study carried out on the distribution of the natural U and ^{210}Po activity in ground water samples of Kodagu District, Karnataka, India, revealed a wide distribution of radioactive elements. The concentration of uranium varied from 0.4 to 8.8 μgl^{-1}, which is found to be below the recommended standard limits of 60 μgl^{-1} by AERB and 30 μgl^{-1} by WHO and USEPA. The activity of ^{210}Po varied from 0.47 to 4.35 mBql^{-1}, which is much below the recommended 100 mBql^{-1} standards of WHO. The total effective dose due to U and ^{210}Po in groundwater varies from 1.08 to 10.36 μSv y^{-1}. The study shows that the inhabitants in the study region are less prone to radiation hazards due to the low activity of U and ^{210}Po in groundwater.

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