Concentration of uranium in groundwater and its correlation with the gamma activity of primordial radionuclides in the bedrock samples: A study from northeastern part of Bengaluru city, India

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INTRODUCTION

Natural radiations originate from many sources, including more than 60 naturally occurring radioactive materials found in rock, soil, water, and air. The maximum contributions to these invisible radiations originate from the decay series of $^{238}$U and $^{232}$Th and the singly occurring isotopes like $^{40}$K.[1] The spatial distributions of these radionuclides show a varying pattern which depends on the nature of the parent rock and soil. Radionuclides can transfer from soil to the human system through various pathways, out of which ingestion plays a major role. Groundwater when it reacts with the soil and bedrock, the latter releases the dissolved components which in turn depends on the geochemical composition of the soil and the rock, chemical composition of water, degree of weathering of rock, redox conditions, and the residence time of groundwater in the soil and the bedrock.[2] Hence, one can presume that the activity concentration of natural radionuclides in groundwater has a direct bearing with the concentrations of uranium and thorium and their decay products in the soil and the bedrock.[3]

The present study has a two-fold approach. In the first part, bedrock samples collected from the borewells of the

Abstract

The present investigation aims to study the incorporation of uranium from the bedrock to groundwater through leaching. In view of this, rock powder samples were collected in the form of slurry from the bedrock of freshly drilled borewells of the study area. The rock powder and the supernatant were separated. The gamma activity concentrations of primordial radionuclides $^{238}$U ($^{226}$Ra), $^{232}$Th, and $^{40}$K in the rock powder samples were measured using HP-Ge gamma-ray spectrometer. The supernatant was analyzed for the concentration of $U_{nat}$ using laser-induced fluorimeter. A strong positive correlation was observed between the concentration of uranium in the rock powder and the corresponding concentration in the water samples indicating the possible leaching of uranium from the bedrock to groundwater under favorable conditions.

Keywords: Bengaluru, bedrock, hyperpure-ge detector, laser fluorimeter, radiation dose, radioisotopes

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study area were analyzed for the gamma activity of the primordial radionuclides $^{238}\text{U}$ ($^{226}\text{Ra}$), $^{232}\text{Th}$, and $^{40}\text{K}$) and the mass elemental concentration of these elements was estimated. In the second part, the concentration of natural uranium in the first spring up water of the same freshly dug borewells was measured. To find the dependence of the concentration of $U_{nat}$ in the water samples on the mass elemental concentration of uranium ($^{238}\text{U}$) in the bedrock, the correlation study was carried out.

Study area
The study area [Figure 1] of the present investigation covers an area of approximately 20 km$^2$ in the northeast region of Bengaluru urban district. As this area is a newly developed residential layout, treated water supply system of the Bengaluru City Corporation is not accessible to the residents and hence borewells are dug and people rely on the groundwater for drinking and other purposes.

Experimental Methods
In the present study, samples were collected in the form of slurry from the bedrock of the freshly drilled borewells during the time of drilling along with the first spring up water. The slurry was allowed to settle completely. The settled rock powder was used to measure the gamma activity of $^{226}\text{Ra}$ ($^{238}\text{U}$), $^{232}\text{Th}$, and $^{40}\text{K}$ whereas the supernatant was used for measuring the concentration of uranium. The depth of the borewells ranged from 183 to 381 m bgl (meters below ground level).

Gamma activity of radionuclides in bedrock samples
The rock powder samples obtained from the residue of the slurry were dried in the sunlight to remove the water content and then kept in hot air oven at 110°C overnight. The dried samples were then filled in 250 ml airtight plastic containers having threaded lids. The lids were tightened so as to avoid the leakage of Rn gas. The samples were then stored for over a month to achieve the radioactive equilibrium between the parent and the daughter nuclides. Since it is difficult to measure the concentration of $^{238}\text{U}$ directly, as the gamma lines of this element are weak, the gamma lines of energy 609 keV and 1120 keV of $^{214}\text{Bi}$ were used to measure the same by assuming secular equilibrium between the parent ($^{238}\text{U}$) and the daughter nuclide ($^{226}\text{Ra}$). In the present study, as the samples were collected from the highly undisturbed environment, the above consideration is more valid. To determine the gamma activities of $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ in the natural environmental samples, the hyperpure germanium (HP-Ge) gamma ray spectrometer is used. The HP-Ge gamma ray spectrometer used in the current study is an n-type closed end coaxial detector (Model GR 4021, Canberra USA) with a relative efficiency of 42%. Before the measurement of activity of radionuclides in the samples, the HP-Ge gamma ray spectrometer was calibrated for the energy and efficiency. Because of the low activity of the samples, the data were accumulated in each case for a period of 60000 s. The background count was also recorded for the same counting period. The gamma lines of 911 keV ($^{228}\text{Ac}$) and 2618 keV ($^{208}\text{Tl}$) were used to determine the activity of $^{232}\text{Th}$. The activity of $^{40}\text{K}$ was determined from its gamma line of energy 1460.8 keV. The activity (Bq/kg) of each radionuclide was calculated using the standard relation (IAEA, 1989).

$$\text{Activity (Bq/kg)} = (S \pm SD) \times \frac{100 \times 1000 \times 100}{E \times W \times A}$$  \hspace{1cm} (1)

where,

$S$ is the Compton corrected background subtracted counts per second under the photopeak used,

$SD$ is the standard deviation,

$E$ is the efficiency of the detector for the corresponding gamma energy (%),

$W$ is the weight of the sample taken for the analysis (g) and

$A$ is the branching intensity of gamma ray energy (%).

Mass concentration of radioisotopes in the bedrock samples
The mass concentration of uranium (in ppm) was calculated from the activity concentration of $^{238}\text{U}$ using the following equation (IAEA, 1989).

$$F_E = \frac{M_E C T_{1/2} A_E}{ln2 N_A}$$  \hspace{1cm} (2)

Where,
$F_e$ is the fraction of element in the sample (ppm),

$M_e$ is the atomic mass (kg/mol),

$N_A$ is the Avagadro’s number ($6.023 \times 10^{23}$ atom/mol),

$f_e$ is the fractional atomic abundance in nature,

$TV/2$ is the half-life of the radionuclide (s),

$C$ is a constant with a value $10^6$ for $^{238}$U ($^{226}$Ra) and $^{232}$Th for concentration in ppm and $10^2$ for concentration of $^{40}$K (%),

$A_i$ is the measured activity of the radionuclide (Bq/kg).

Measurement of uranium concentration in water samples

The water samples (supernatant) drained off from the slurry were filtered through Whatman 42 filter paper. Fluor (1 mL) was added to 6 mL of each water sample and analyzed for the concentration of uranium using pre-calibrated laser-induced fluorimeter having a minimum detection limit of 0.2 ppb.

Annual effective dose

The annual effective dose ($\mu$Sv/y) due to the ingestion of uranium through drinking water was calculated as the product of activity concentration (Bq/L) of the element in water, the annual intake of water (L/y), and the dose conversion factor (Sv/Bq). In the present study, the annual intake of water was taken as 730 L/y, at the rate of 2 L/day, and the dose conversion factor as $4.62 \times 10^{-8}$ Sv/Bq obtained as the average of the dose coefficients for $^{234}$U, $^{235}$U, and $^{238}$U isotopes based on ICRP publications.

RESULTS AND DISCUSSION

The results obtained in the present investigation reveal that the gamma activity concentrations of $^{238}$U ($^{226}$Ra) in the rock powder samples is found to vary from 4.8 ± 0.5 – 31.9 ± 1.2 Bq/kg with a mean of 16.04 ± 0.82 Bq/kg whereas that of $^{232}$Th and $^{40}$K are found to be in the range 13.2 ± 0.6–82 ± 1.4 Bq/kg and 499.1 ± 8.3–783.1 ± 10.3 Bq/kg, respectively, and are shown in Table 1.

The observed values are well within the global range for $^{238}$U ($^{226}$Ra). Three samples (S8, S10 and S11) show higher concentration of $^{232}$Th values compared to the global average. The average gamma activity of the $^{40}$K (669 ± 9.8 Bq/kg) is high compared to the global average of 420 Bq/kg. Among all the samples, sample no. S12 exhibits the lower activity of all the three radionuclides, whereas sample no. S8 shows the highest activity for $^{238}$U ($^{226}$Ra) and $^{232}$Th with quite high value (760.2 ± 10.4 Bq/kg) for $^{40}$K. The gamma activity of $^{232}$Th is considerably large compared to that of $^{238}$U ($^{226}$Ra) in all the samples and this may be due to the fact that uranium, which is the parent nuclide of radium is more susceptible to solubility compared to thorium which is less soluble and hence adsorbed to the rock system.

The correlation analysis (Pearson correlation coefficient) between the gamma activity of the three radionuclides was carried out, and the positive and strong correlation coefficient ($r = 0.66$, 0.57, and 0.56 for $^{232}$Th and $^{226}$Ra, $^{40}$K and $^{226}$Ra and $^{40}$K and $^{232}$Th, respectively) obtained among the three radionuclides indicate that the rock samples belong to a bedrock system that are geochemically coherent.

The mass concentration of $^{238}$U, $^{232}$Th, and $^{40}$K(%) calculated using Eq (2) are presented in Table 2. The concentration of uranium (ppb) in the water samples (obtained from the supernatant) measured using laser fluorimeter and the estimated annual effective dose ($\mu$Sv/y) is also included along with the concentration of uranium in the water samples collected from the same borewells when they are put into regular use.

In the present investigation, the mass concentration of $^{238}$U in the bedrock samples collected from the study area was found to lie in the range 0.39 ± 0.04–2.58 ± 0.09 with a mean of 1.3 ± 0.07 which is well within the average concentration of uranium (2.64 ppm) in the earth’s crust. It can also be seen from the table that the sample S8 exhibits the highest

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**Table 1: Gamma activity of $^{238}$Ra, $^{232}$Th, and $^{40}$K (Bq/kg) in the rock powder samples**

| Sample code | $^{238}$U ($^{226}$Ra) | $^{232}$Th | $^{40}$K |
|-------------|----------------------|-----------|---------|
| S1 (183)    | 15.4±0.5             | 33.9±0.8  | 549.6±8.9 |
| S2 (244)    | 16.2±0.9             | 36.8±0.9  | 687.9±10.2 |
| S3 (287)    | 14.4±0.8             | 39.3±1.0  | 641.5±9.7  |
| S4 (290)    | 14.7±0.8             | 37.4±0.9  | 731.7±10.3 |
| S5 (290)    | 8.4±0.7              | 17.3±0.6  | 611.0±9.2  |
| S6 (366)    | 27.4±1.2             | 38±0.9    | 698.8±10.2 |
| S7 (381)    | 17.8±0.9             | 26.8±0.8  | 700.2±10.1 |
| S8 (305)    | 31.9±1.2             | 82±1.4    | 760.2±10.4 |
| S9 (365)    | 11.9±0.7             | 35.3±0.9  | 783.1±10.3 |
| S10 (335)   | 18.9±0.9             | 66.0±1.2  | 728.6±10.1 |
| S11 (320)   | 15.6±0.8             | 65.4±1.2  | 661.6±9.6  |
| S12 (280)   | 4.8±0.5              | 13.2±0.6  | 499.1±8.3  |
| S13 (335)   | 11.1±0.8             | 33.2±0.9  | 655.4±9.7  |

Range: 4.8±0.5–31.9±1.2

Mean: 16.0±0.8

Global range: 17–60 (32)

and average: 4.6 ± 0.5

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"The values shown in the parenthesis are the depth of the borewells in m bgl"
Table 2: The mass concentration of $^{238}\text{U}$ and $^{232}\text{Th}$ (ppm) and the concentration of $^{40}\text{K}$ (%) in the bedrock samples along with the concentration of uranium in the water samples and the annual effective dose ($\mu\text{Sv}/\text{y}$) due to its ingestion

| Sample code | Concentration of radionuclides | Concentration of uranium in water samples (supernatant) (ppb) | Annual effective dose (µSv/y) | Concentration of uranium in water samples (ppb) |
|-------------|--------------------------------|---------------------------------------------------------------|-------------------------------|-----------------------------------------------|
| S1 (183)    | 1.25±0.04                      | 8.4±0.2                                                       | 1.80±0.03                     | 28.5±0.02                                     |
| S2 (244)    | 1.31±0.07                      | 9.1±0.2                                                       | 2.25±0.03                     | 36.2±0.03                                     |
| S3 (297)    | 1.17±0.07                      | 9.7±0.2                                                       | 2.10±0.03                     | 17.1±0.02                                     |
| S4 (290)    | 1.19±0.07                      | 9.3±0.2                                                       | 2.4±0.03                      | 16.8±0.02                                     |
| S5 (290)    | 0.68±0.06                      | 4.3±0.2                                                       | 2.00±0.03                     | 24.1±0.02                                     |
| S6 (366)    | 2.22±0.09                      | 9.4±0.2                                                       | 2.29±0.03                     | 165.0±0.03                                    |
| S7 (381)    | 1.44±0.07                      | 6.6±0.2                                                       | 2.29±0.03                     | 15.8±0.02                                     |
| S8 (305)    | 2.58±0.09                      | 20.3±0.3                                                      | 2.49±0.03                     | 172.4±0.03                                    |
| S9 (365)    | 0.96±0.06                      | 8.7±0.2                                                       | 2.57±0.03                     | 15.4±0.01                                     |
| S10 (335)   | 1.53±0.07                      | 16.3±0.3                                                      | 2.39±0.03                     | 20.3±0.02                                     |
| S11 (320)   | 1.26±0.06                      | 16.2±0.3                                                      | 2.17±0.03                     | 10.4±0.01                                     |
| S12 (280)   | 0.39±0.04                      | 3.3±0.1                                                       | 1.64±0.02                     | 1.0±0.01                                      |
| S13 (335)   | 0.89±0.07                      | 8.2±0.2                                                       | 2.15±0.03                     | 2.5±0.01                                      |
| Mean        | 1.3±0.07                       | 9.9±0.2                                                       | 2.19±0.03                     | 40.4±0.02                                     |

The values shown in the parenthesis are the depth of the borewells in m bgl. *After the borewells are put into regular use

The leaching of uranium from the rock system to the water body can be understood as follows. The flow of groundwater to a drilled well in crystalline rock normally follows through the fracture zones and other rock structures such as rock contacts. Due to the hydrogeochemical processes occurring in the deep subsurface, uranium and its decay products such as radium and radon may be enriched on the surface of the fractures. Uranium in rocks normally occurs in tetravalent (immobile) state. If the surrounding environment is oxidizing, uranium is easily oxidized to the more mobile hexavalent state. The leached uranium gets into solution and is transported together with groundwater. Under chemically reducing conditions, the dissolved uranium precipitates out of the solution. Thus, the observed trend of variation in the concentration of uranium in the groundwater samples can be correlated mainly with that in the bedrock system.

The typical concentration of uranium in granitic rocks is 3–5 ppm whereas in sedimentary rocks, it is 2–3 ppm. The observed high elemental concentration of uranium in few rock powder samples could suggest pockets of relatively

![Concentration of $^{238}\text{U}$ in rock samples (ppm)](image)

$y = 86.41x - 71.73$
$r = 0.87$
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higher concentration in the bedrocks. The permissible limit of concentration of uranium in drinking water as set by the World Health Organization is 30 ppb and the annual effective dose due to its ingestion is 100 µSv/y[8] whereas the Atomic Energy Regulatory Board, India has set 60 ppb as the safe limit for the Indian environment.[17] Thus, in the present investigation, the concentration of uranium in two water samples collected from the borewells (S6 and S8) is much higher compared to these permissible limits.

CONCLUSIONS

The data obtained in the present study reveal that the water originating from borewells drilled in uranium rich bedrock has a high concentration of uranium due to the leaching of the element under favorable conditions. A positive correlation ($r = 0.87$) observed between the concentration of $^{238}$U in rock powder samples and that of $\text{U}_{\text{sw}}$ in water samples collected from the same borewells substantiates this fact. Although the study area covers a ground area of about 20 km$^2$, the mass concentration of uranium in the rock powder samples shows considerably wide range (0.39 ± 0.04–2.58 ± 0.09 ppm) with an average of 1.3 ± 0.07 ppm. This is well within the world average of 2.64 ppm for concentration of uranium in the earth's crust. The concentration of uranium in the water samples lies in the range 1.0 ± 0.01–172.4 ± 0.03 ppb with an average of 40.4 ppb. This corresponds to the annual effective dose of 0.82–147.34 µSv/y with a mean of 34.54 µSv/y. The exact reason for the wide range of concentration of uranium in the water samples collected from the borewells of the study area require a better understanding of the factors such as the degree of leaching, the properties of the rock, the flow path, fracture system, pH, transport mechanism, and many others which influence the concentration of the element.

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Conflicts of interest

There are no conflicts of interest.

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