Studies on $^{226}$Ra and $^{222}$Rn concentration in drinking water of Mandya region, Karnataka State, India

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Abstract

Radium is naturally occurring radioactive element in the earth's crust. It is chemically similar to calcium and absorbed from soil by plants, passed up the food chain to humans. The occurrence and the distribution of radioactivity in water depend on the local geological characteristics of the source, soil, rock and other factors that control the occurrence and distribution of radionuclides in ground water and the hydro geological condition and the geochemistry of radionuclides. Activities of the naturally occurring radionuclides $^{226}$Ra and $^{222}$Rn were determined in natural ground water of the Mandya district, Karnataka State, India. The concentration of $^{222}$Rn in borewell water varies from 6.44 ± 0.20 to 44.83 ± 0.54 Bq l$^{-1}$ with geometric mean 16.42 ± 0.31 Bq l$^{-1}$. Higher radon concentrations were observed at Yettaganahalli and Mandya city. $^{226}$Ra concentration varies from 14.26 ± 0.32 to 81.06 ± 0.99 mBq l$^{-1}$ with geometric mean 27.61 ± 0.43 mBq l$^{-1}$. It is observed that the radon concentration is high in ground water around the granitic rock exposures and similarly observation of high values of radon is reported in sheared gneiss which covers major portion of the Mandya district to phyllites and schists. The total dose due to ingestion and inhalation varies from 26.31 to 178.53 μSv y$^{-1}$ with a geometric mean of 65.94 μSv y$^{-1}$, which is below the prescribed dose limit of 100 μSv y$^{-1}$ by WHO.

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1. Introduction

Radium and radon concentration in ground water and its changeability with time and space have been studied frequently in recent years. The results of these investigations are very important for understanding radon migration processes in the lithosphere and the role of ground water as radon carrier fluid (Przylibski, Mamont-Cieśla, Kusyk, Dorda, & Kozlowska, 2004). It is also very important to recognize the role of geological structure and rock type as a source of radon dissolved in ground water. The underground water often moves through rock and soil containing radon and radon gets solubilized in the water (Duggal, Rani, Mehra, & Ramola, 2014). Water for human consumption should be free from chemical, microbiological and radiological contamination (UNSCEAR, 2000). Radium is more chemically active and it is similar to actinium. This can be absorbed from the soil by plants and
transported to the food chain to humans. The radiation emitted by radium will affect the tissues in the bone marrow that produces red blood cells and also can cause bone cancer. It undergoes natural disintegration and formed into stable isotope $^{206}$Pb. The radioactive gas radon is a decay product of $^{226}$Ra. It is very important from the point of view of health risk.

$^{222}$Rn is an inert gas whose concentrations in ground water are reportedly related to a number of factors including emission of $^{222}$Rn from surrounding rocks, temperature, pressure, rainfall and earthquake activities (Ilani, Minster, Kronfeld, & Even, 2006; Sannappa, Chandrashekara, & Paramesh, 2006).

Radon concentrations in water have been known to be high in most granite and in high-grade metamorphic rocks, whereas less metamorphosed rocks have somewhat less $^{226}$Ra. The highest $^{226}$Ra/$^{222}$Rn levels in ground water are found in granites, sillimanite or sillimanite/orthoclase grade rocks; the lowest levels are found in chlorite to staurolite grade rocks. Generally, higher radon concentration is observed in waters of low mineralization. Also, the radon activity decreases with the depth of the area where the water circulates (Kozlowska, Hetman, & Zipper, 1999).

It escapes from the earth’s crust through cracks and crevices in bedrock and either dissolves in ground water or seeps through foundation cracks into environment/human habitations (Ningappa, Sannappa, Chandrashekara, & Paramesh, 2009). The alpha radiation emitted by radon and its progeny polonium is considered a significant health hazard by the United States Environmental Protection Agency because at elevated levels it causes lungs cancer (Lubin et al., 1995; UNSCEAR, 2006). Radon present in rocks of the earth crust continuously diffuses through the water in the rocks leading to the presence of $^{222}$Rn in ground water. It has been determined that radon is the noble gas with higher solubility in water with of the noble gas mole fraction value of 0.00125 at 37 °C, half life of 3.8 days, which is 15 times higher than that of helium or neon. Radon in water is primarily a problem for water supplies which extract water from drill holes in rocks, which have somewhat higher uranium concentrations than the average bedrock. Some types of rocks which often have enhanced uranium concentrations, greater than 5 ppm of uranium include; granites, syenites, pegmatite, acid volcanic rocks and gneisses (Shashikumar, Chandrashekara, Nagaiah, & Paramesh, 2009; UNSCEAR, 2000).

1.1. Study area

The present study was carried out in Mandya district Karnataka, India. It lies between North latitude 12°13′ to 13°04′ and East longitudes 76°19′ to 77°20′ (Fig. 1). The district is located in the southern meridian region of the state. Ground water occurrence is limited to weathered zones and plains of foliation or weak fracture zones of hard rock formations like granites and gneisses. Geologically, the major portion of the district is covered by granite and granitic gneisses. However, a band of chlorite and mica schist along with intrusive like pegmatite, dolerites and porphyries occurs as isolated patches. The rocks are medium-to coarse-grained and composed of grayish or white feldspars, bluish grey or white translucent to opaque quartz, biotitic, and hornblende.

2. Methods of measurements

2.1. $^{226}$Ra concentration in water

About 20 l of water was collected from underground water and surface water sources in pre acid cleaned cans. Locations were marked using a GPS system and using portable pH meter pH was measured at the sampling Location. Analysis of radium in water generally involves pre-concentration of samples by co-
precipitation or evaporation (Iyengar, Kannan, & Rao, 1989; Reid, Key, & Schink, 1979).

We have adopted the method of adsorbing the radium in water on commercially available manganese dioxide powder, which can be used even for in-situ processing. About 20 l of water samples was filtered through Whatman 42 filter paper. To this filtered water in the container, 5 g of analytical grade MnO₂ powder was added. The mixture was stirred for a period of 1 h using a mechanical stirrer and then allowed to settle for 2 h. The solution was heated until the clear solution was obtained and then treated with 50 ml of concentrated HCl. The solution was evaporated to near dryness and treated with 30 ml of concentrated HNO₃ to convert it into nitric acid medium and heated 3 times to near dryness to evaporate the organic content present in the water. The system was then cooled and make the solution about 70 ml using 4 N HNO₃.

Radium being the parent of radon, its concentration can be obtained by measuring built up radon concentration in radon bubblers. Pre concentrated water sample of about 70 ml was transferred to the radon bubbler is shown in (Fig. 2). A vacuum pump was connected to the bubbler and air was sucked through the sample solution to scrub it for about 5 min. This would purge the solution of dissolved ⁰²²Rn. The solution in the blubber was then allowed to stand for a known period of about 21 days (about 3–5 half lives of ⁰²²Rn) for enough radon to build up.

At the end of this period, an evacuated scintillation cell was connected to the bubbler through the quick connector. Under the influence of the vacuum in the scintillation cell, air gets sucked through the solution and fills the scintillation cell. In the process the air carries the dissolved radon in the solution. By careful manipulation of the stopcocks the bubbling was controlled and adjusted to be uniform and steady to ensure complete transfer of radon into the scintillation cell (Fig. 3). The scintillation cell was kept for 3 h or more to allow radon daughters to reach equilibrium with radon. Then, the alpha activity was counted for a period of 1000 s. The activity of ²²⁶Ra in the sample was determined using the equation (1) (Raghavayya, Iyengar, & Markose, 1980).

\[
\frac{226\text{Ra (Bq l}^{-1})}{C_0} = \frac{6.97 \times 10^{-2} \times D}{V \times E \times (e^{-\lambda t}) \times (1 - e^{-\lambda t}) \times (1 - e^{-\lambda q})} \tag{1}
\]

where, 
- \( D \) = counts above background (Counts s⁻¹)
- \( V \) = volume of water (L),
- \( E \) = efficiency of the scintillation cell (74%),
- \( \lambda \) = decay constant for radon \((2.098 \times 10^{-6} \text{ s}^{-1})\),
- \( T \) = counting delay after sampling (s),
- \( t \) = counting duration (s) and
- \( q \) = build up time in the bubbler (s).

2.2. ²²²Rn concentration in water

Radon in water is measured by bubbling the radon into evacuated ZnS(Ag) scintillation cells (Lucas cell) in a controlled way using specially built devices. The minimum detection limit for the measurement of radon is 0.05 Bq l⁻¹. The instruments are supplied by Polletec Instruments Pvt.
About 100 ml of water sample was collected from ground water and surface water sources using a pre acid cleaned airtight plastic bottles with minimum disturbance. In borewell water sampling, water should be pumped out for 5 min. In order to obtain fresh water from borewell, the water was allowed run fully at least 5 min before the sample was taken. Then is collected by allowing water to flow along the wide mouth bucket. About 100 ml of water samples were collected in airtight plastic bottles with minimum disturbance. The plastic bottles were filled completely in a gentle manner, so that zero head space was present. Care was taken to see that no air bubbles were seeing inside the container and also to avoid radon to attain equilibrium with its daughters and then it was coupled to a photomultiplier and alpha counting as-assembly. The concentration was calculated using the equation (2) given by Raghavayya et al. (1980).

\[
\frac{222\text{Rn}}{\text{Bq l}^{-1}} = \frac{6.97 \times 10^{-2} \times D}{V \times E \times (e^{-\lambda t}) \times (1 - e^{-\lambda t})}
\]

(2)

where, \( D \) = counts above background (Counts s\(^{-1} \)),

\( V \) = volume of water (l),

\( E \) = efficiency of the scintillation cell (74%),

\( \lambda \) = decay constant for radon (2.098 \times 10^{-9} \text{ s}^{-1}),

\( T \) = counting delay after sampling (s) and

\( t \) = counting duration (s).

### 2.3. Water quality parameters

The pH measurements were performed using a glass electrode. The instrument used was the ELICO LF120 by Elico Pvt. Ltd with an accuracy of 0.1 pH units. The electrical conductivity of the water samples was measured with a conductivity meter ELICO CM180 from Elico Pvt. Ltd with a relative accuracy of 1%.

### 2.4. Dose due to 222Rn and 226Ra concentration in water

The committed effective dose for the population of the region was estimated using the concentration of 222Rn and 226Ra in water samples. The parameters for the inhalation pathway were 222Rn concentration in water, air water concentration ratio of \( 10^{-4} \), indoor occupancy of 7000 h per year, equilibrium factor 0.4 and inhalation dose conversion coefficient 9 nSv (Bq h m\(^{-3} \))\(^{-1} \). The effective dose to the ingestion mainly depends upon the amount of water consumed by a human being in a day. The dose due to inhalation and ingestion are calculated by the equations (3)–(5) (UNSCEAR, 2000)

\[
\text{Inhalation dose} (\mu\text{Sv}) = 222\text{Rn conc. (Bq l}^{-1}) \times 10^{-4} \times 7000 \text{ h} \\
\times 0.4 \times 9 \text{nSv (Bq h m}^{-3} \text{)}^{-1} \tag{3}
\]

\[
\text{Ingestion dose} (\mu\text{Sv})=222\text{Rn conc. (Bq l}^{-1}) \times 365 \text{ l y}^{-1} \\
\times 3.5 \text{nSv Bq}^{-1} \tag{4}
\]

\[
\text{Ingestion dose} (\mu\text{Sv})=226\text{Ra conc. (Bq l}^{-1}) \times 365 \text{ l y}^{-1} \\
\times 280 \text{nSv Bq}^{-1} \tag{5}
\]

### 3. Results and discussions

The 222Rn concentration in natural drinking water in different types of water samples was estimated using emanometry method in the Mandya region. The major portion of the district is covered by granite and granitic gneisses. However, a band of chlorite and mica schist along with intrusives like pegmatites, dolerites and porphyrycs occurs as isolated patches. The rocks are medium-to coarse-grained and composed of grayish or white feldspars, bluish grey or white translucent to opaque quartz, biotitic, and hornblende. The variation of 222Rn and 226Ra activity in borewell water and the corresponding radiation dose is shown in Table 1. The concentration of 222Rn in borewell water varies from 6.44 ± 0.20 to 44.83 ± 0.54 Bq l\(^{-1} \) with geometric mean 16.42 ± 0.31 Bq l\(^{-1} \). Higher radon concentrations were observed at Yettaganahalli and Mandya city. 226Ra concentration varies from 14.26 ± 0.32 to 81.06 ± 0.99 mBq l\(^{-1} \) with geometric mean 27.61 ± 0.43 mBq l\(^{-1} \). Higher radium concentrations were observed at Mahadevapura, Saburayana Koppalu and Yetti- ganahalli. Higher radiation levels are associated with igneous rocks, such as granite, and lower levels with sedimentary rocks. There are exceptions, however, as some shales and phosphate rocks have relatively high content of radionuclides (Bonotto, 2014). It is observed that the radon concentration is high in ground water around the granitic rock exposures and similarly observation of high values of radon is reported in sheared gneiss which covers major portion of the Mandya district to phyllites and schists (Hunse, Najeeb, Rajarajan, & Muthukkannan, 2010).

The frequency distribution of 226Ra and 222Rn concentration in borewell water of Mandya region is shown in Figs. 4 and 5 respectively. The distribution is of log normal and about 18% of the samples were found to be less than permissible limit of 11 Bq l\(^{-1} \), which is proposed by USEPA for radon concentration through water intake. About 82% of the borewell water samples show slightly higher level of 222Rn concentrations, which exceeds the MCL 11 Bq l\(^{-1} \) as recommended by USEPA (2000).

The dose due to 222Rn is divided into two parts, namely the dose from ingestion and the dose from inhalation. For the
ingestion and inhalation part, $^{222}\text{Rn}$ and its progeny in water impart a radiation dose to the stomach and lung respectively. Computing from the radium and radon activity concentrations in borewell water samples, the total dose due to ingestion and inhalation varies from 26.31 to 178.53 μSv y$^{-1}$ with a geometric mean of 65.94 μSv y$^{-1}$, which is below the prescribed dose limit of 100 μSv y$^{-1}$ by WHO (2011). At Malavalli, Mandya and Yettaganahalli the total dose is above 100 μSv y$^{-1}$ due to higher concentration of radium and radon in borewell water (Eckerman et al., 2012; IAEA, 2011).

In Mandya region tap water facility is available for domestic use at few towns. The variation of $^{222}\text{Rn}$ and $^{226}\text{Ra}$ activity in tap water and the corresponding radiation dose is shown in Table 2. The total dose due to ingestion and inhalation varies from 26.54 to 33.01 μSv y$^{-1}$ with a geometric mean of 29.5 μSv y$^{-1}$, which is well below the prescribed dose limit of 100 μSv y$^{-1}$ by WHO.

The distribution of the radium and radon activity in different source of water samples is shown in Fig. 6. Borewell and open well contain higher concentration of $^{226}\text{Ra}$ and $^{222}\text{Rn}$. $^{226}\text{Ra}$ concentration in borehole water and open well water have the radium concentration with average value 27.61 ± 0.43 mBq l$^{-1}$ and 18.9 ± 0.90 mBq l$^{-1}$ respectively. $^{222}\text{Rn}$ concentration in borehole water and open well water with average value 19.79 ± 0.33 Bq l$^{-1}$ and 12.7 ± 0.56 Bq l$^{-1}$

<table>
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<tr>
<th>Location</th>
<th>Number of samples</th>
<th>pH</th>
<th>EC (μS cm$^{-1}$)</th>
<th>$^{222}\text{Rn}$ (Bq l$^{-1}$)</th>
<th>$^{226}\text{Ra}$ (mBq l$^{-1}$)</th>
<th>Inhalation dose due to $^{222}\text{Rn}$ (μSv y$^{-1}$)</th>
<th>Ingestion dose due to $^{222}\text{Rn}$ (μSv y$^{-1}$)</th>
<th>Ingestion dose due to $^{226}\text{Ra}$ (μSv y$^{-1}$)</th>
<th>Total dose (μSv y$^{-1}$)</th>
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<td>Mahadevapura</td>
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<td>68.09 ± 0.60</td>
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<td>14.73</td>
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<td>8.0</td>
<td>1123</td>
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<td>14.26 ± 0.32</td>
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<td>41.38</td>
<td>20.98</td>
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Fig. 4 – Distribution of $^{226}\text{Ra}$ in water samples of Mandya region.

Fig. 5 – Distribution of $^{222}\text{Rn}$ in water samples of Mandya region.
respectively. Tap water, Lake water and River water have relatively lower concentration of both $^{226}$Ra and $^{222}$Rn. Low $^{226}$Ra and $^{222}$Rn concentrations were observed in the tap water, Lake water and river water is due to the lack of major contact with radon emanating mineral material and also the aeration of radon gas to the atmosphere. The concentrations of radon in water may range over several orders of magnitude, generally being highest in well water, intermediate in ground water, and lowest in surface water (Chandrashekara, Veda, & Paramesh, 2012; Kitto, Parekh, Torres, & Schneider, 2005).

The scatter plot of variation between radon and radium concentration in water samples is shown in Fig. 7. There is a good correlation between radium and radon in water with a Pearson’s $r$ value of 0.76. The correlation between the $^{226}$Ra concentration and electrical conductivity in natural water samples was studied and it is shown in Fig. 8. There is a good correlation between electrical conductivity and $^{226}$Ra concentration in water with a Pearson’s $r$ value of 0.61. In general at places where there is higher electrical conductivity, the radium concentration is found to be high. But there is no significant correlation between pH and the concentration of radium or radon in water.

The results of the $^{222}$Rn concentration in the ground water of the study area are compared with the literature values reported for other environs and are shown in Table 3. The overall mean value of 8.22 Bq l$^{-1}$ observed in the present study is low compared to the literature value reported in Brazil, Israel, Italy South America, Korea, China, SW Poland, Finland, Himalay, Mysore, Bangalore, Tumkur. It is found to be high compared coastal Kerala Rajasthan, Punjab, and Coonoor Tamilnadu (Bonotto, 2014; Cho, Ahn, Kim, & Lee, 2004; Choubey & Ramola, 1979; D’Cunha, Narayana, Karunakara, Yashodhara, & Kumara, 2011; Duggal et al., 2014; Godoy & Godoy, 2006; Ilani et al., 2006; Kozłowska et al., 2009; Najeeb, Vinayachandran, Jose, & Vashistha, 2014; Przylibski et al., 2004; Rajesh, Chandrashekara, Nagaraja, & Paramesh, 2012; Najeeb, Vinayachandran, Jose, & Vashistha, 2014; Przylibski et al., 2004; Rajesh, Chandrashekara, Nagaraja, & Paramesh, 2012;...
Selvasekarapandian et al., 2002; Shiva Prasad, Nagaiah, Ashok, & Mahesh, 2007; Singh, Singh, Singh, Bajwa, & Sonkawade, 2009; Vesterbacka, Makelainen, & Arvela, 2005; Zhuo, lida, & Yang, 2001). The distribution of the $^{226}$Ra and $^{222}$Rn may range over several orders of magnitude in different types of water samples. It is evident that higher activity concentration observed in borewells water intermediated in ground water and lowest in surface water and tap water. The concentrations of radon in water generally being highest in well water, intermediate in ground water, and lowest in surface water.

### 4. Conclusion

The concentrations of $^{226}$Ra and $^{222}$Rn in natural ground water of Mandya districts the water samples were collected and analyzed by radon Emanometry technique. The concentration of $^{222}$Rn in borewell water varies from $0.44 \pm 0.20$ to $44.83 \pm 0.54$ Bq l$^{-1}$ with geometric mean $14.26 \pm 0.31$ Bq l$^{-1}$. Higher radon concentrations were observed at Yettaganahalli and Mandya city. $^{226}$Ra concentration varies from $14.26 \pm 0.32$ to $81.06 \pm 0.99$ mBq l$^{-1}$ with geometric mean $27.61 \pm 0.43$ mBq l$^{-1}$. It is observed that the radon concentration is high in ground water around the granitic rock exposures and similarly observation of high values of radon is reported in sheared gneiss which covers major portion of the Mandya district to phyllites and schists. The total dose due to ingestion and inhalation varies from $26.31$ to $178.53$ $\mu$Sv y$^{-1}$ with a geometric mean of $65.94$ $\mu$Sv y$^{-1}$, which is well below the prescribed dose limit of $100$ $\mu$Sv y$^{-1}$ by WHO.

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### References


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**Table 3 – $^{222}$Rn concentration (Bq l$^{-1}$) in ground water at different parts of the world.**

<table>
<thead>
<tr>
<th>Present work</th>
<th>Literature value</th>
<th>Region</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$6.44 \pm 0.20$–$44.83 \pm 0.54$</td>
<td>50–300</td>
<td>South Korea</td>
<td>(Cho et al., 2004)</td>
</tr>
<tr>
<td></td>
<td>61–274</td>
<td>Israel</td>
<td>(Ilani et al., 2006)</td>
</tr>
<tr>
<td></td>
<td>12.7</td>
<td>Italy</td>
<td>(Kozlowska et al., 2009)</td>
</tr>
<tr>
<td></td>
<td>$&lt;1.2$–$3542$ (57.7)</td>
<td>Brazil</td>
<td>(Godoy &amp; Godoy, 2006)</td>
</tr>
<tr>
<td></td>
<td>27–460</td>
<td>Finland</td>
<td>(Vesterbacka et al., 2005)</td>
</tr>
<tr>
<td></td>
<td>$0.71$–$3735$ (229.4)</td>
<td>China</td>
<td>(Zhuo et al., 2001)</td>
</tr>
<tr>
<td></td>
<td>14–64</td>
<td>South America</td>
<td>(Bonotto, 2014)</td>
</tr>
<tr>
<td></td>
<td>0.2–$1645$ (240)</td>
<td>SW Poland</td>
<td>(Przylibski et al., 2004)</td>
</tr>
<tr>
<td></td>
<td>5–887</td>
<td>Himalaya</td>
<td>(Choubuy &amp; Ramola, 1997)</td>
</tr>
<tr>
<td></td>
<td>0.87–32.10</td>
<td>Punjab</td>
<td>(Singh et al., 2009)</td>
</tr>
<tr>
<td></td>
<td>1.6–5.4 (3.3)</td>
<td>Rajasthan</td>
<td>(Duggal et al., 2014)</td>
</tr>
<tr>
<td></td>
<td>0.03–5.72</td>
<td>Coonoor Tamilnadu</td>
<td>(Selvasekarapandian et al., 2002)</td>
</tr>
<tr>
<td></td>
<td>0.3–1.31 (0.29)</td>
<td>Coastal Kerala</td>
<td>(D’Cunha et al., 2013)</td>
</tr>
<tr>
<td></td>
<td>5.3–283 (87.0)</td>
<td>Bangalore</td>
<td>(Shiva Prasad et al., 2007)</td>
</tr>
<tr>
<td></td>
<td>BDL–$643.9$</td>
<td>Mysore</td>
<td>(Rajesh et al., 2012)</td>
</tr>
<tr>
<td></td>
<td>5–250</td>
<td>Tumkur</td>
<td>(Najeeb et al., 2014)</td>
</tr>
</tbody>
</table>


Zhuo, W., Iida, T., & Yang, X. (2001). Occurrence of $^{222}$Rn, $^{226}$Ra, $^{228}$Ra and U in groundwater in Fujian Province, China. *Journal of Environmental Radioactivity*, 53, 111–120.