Studies on concentration of ^{226}Ra and ^{222}Rn in drinking water around Bangalore Rural districts, Karnataka, India

G. Chandrashekaraiah¹, N Sivasankara Reddy², A Jayasheelan³

¹Department of Physics, Government First Grade College, Kunigal, Tumkur-572130, India

²Department of Physics, School of Engineering, Presidency University, Bangalore-560064, India.

³Department of Physics, Maharani Science College for Women, Bangalore-560001, India.

(Correspondingauthoremail: jayasheelanwillbe@gmail.com)

1. INTRODUCTION

Water is an imperative natural resource on the earth. Approximately 71% of the earth's floor is protected with water. Fresh water is located as underground water in massive reservoirs surrounded by means of rock referred to as aquifers and surface water. It's a vital and essential element for survival of all the living beings. There is a devastating call for pure water due to the fact the groundwater and surface water are highly polluted due to urbanization and industrialization. Out of the total quantity of worldwide water, only 2.4% is distributed on the main land, of which best a small component may be utilized as clean water. Ground water is the principal supply of drinking water in each rural and urban parts of India. Ground water is a susceptible electrolyte containing numerous elements and their associated radicals in ionic equilibrium. The fundamental cations are calcium, magnesium, sodium and potassium. The primary anions are carbonates, nitrate, bicarbonates, chlorides and sulphates. The opposite parameters decided are general dissolved solids, general hardness, PH, iron and radon, a natural radioactive gas. The ^{222}Rn is a obviously happening radioactive noble gas in the surroundings formed by radioactive decay of ^{226}Ra . It undergoes radioactive decay with the aid of emission of alpha particles with a feature half-life 2.82days. It decays by emission of alpha particle to form radon decay mechanism, which can be divided into quick-lived and long-lived progeny species. Radon is soluble in water and its solubility decreases with increase in temperature. The excessive degree of radon in groundwater results from radium affords within the rocks through which groundwater flows. Consequently, the high concentration of uranium in the surrounding rocks is the sources of high radon in groundwater [1]. Radon concentration ground water is affected by several factors, such as presence of shear zones, degree of metamorphism, soil porosity and uranium mineralization [2-4]. Radon dissolved in water can cause radiation exposure via inhalation and ingestion. The organ at maximum threat from radon dissolved in water is belly. While radon or its progeny are inhaled, lung cancer for most of the overall incremental cancer danger, even as ingestion of radon in water is suspected of being related to increased danger of tumors of several inner organs, typically the stomach [5-6]. The ability fitness dangers associated with radon and other certainly occurring radioactive factors present in water have extended in relevance in latest years. This is due to the attention directed toward the very excessive radiation doses which babies and youngsters can get hold of because of consumption of water containing radon and radon decay merchandise [7-9]. The modifications in high-quality of groundwater response to variant in physical, chemical and biological environments through which it passes [10]. Groundwater resource in the observe vicinity is extensively exploited for irrigation and other domestic purposes similarly to consuming reason. Concentration of radon and physiochemical parameters greater than their permissible limits in ingesting water ends in fitness issues together with water borne sicknesses, like fluorosis, typhoid, cholera, untimely child and other issues, mainly toddlers [11]. Not all drinking water contains Radon.

If drinking water arises from a surface water source, such as a river, lake, or reservoir, most Radon that might be in the water will be released into the air before reaching water supplier or home.

2. Study area

The study area is rural Bangalore, located in the south-eastern India. The flat topped lateritic hills are seen within the north-eastern part of the district. Geologically the region belonged to peninsular gneisses, more youthful and older granites, dolerite and amphibolite dykes, laterite, charnockite and amphibolite's and politic schist. Further to these lots of granite quarries and crushing vegetation are spread over the complete district, granite quarrying and crushing goes on for the past 3 decades and density of crushing vegetation inside the district is excessive. The quarrying activities motive air and water pollution. The crushing and mining activities are answerable for the emission of dirt main to the redistribution of associated radioactivity in distinct matrices of the environment

3. Materials and methods

Radon bubbler is a specially designed set up for the measurements of Radium as well as Radon in liquid samples. While Radon can be directly estimated by degassing from the sample, it is indirectly estimated through Radon build up by emanometric technique. Radon bubblers are made of corning glass with airtight joints and stop cocks. Due to leak proof couplings for gassing and degassing of samples as well Radon sampling to Lucas cells are provided in the system. The schematic diagram of the Radon bubbler is shown in Figure 1. The scintillation cells used for estimation of Radon collected from the bubbler were calibrated spending standard Radium solution (IAEA standard). The Radium solution of known strength ($Bq\,m^{-3}$) was kept stored in a Radon bubbler. The Radon built up all through a known period was quantitatively shifted to the scintillation cells and the form the identified Radon concentration the cell efficiencies were calculated. The efficiency of the cells used was of the order of 79.4 ± 2.25 percent.

Water samples from bore wells were collected from various locations of Bangalore rural district. The sample bottles were systematically cleaned with the water to be sampled before collection. Also before collecting bore well water, about 10-20 litre water was pushed out and then 1 litre sample was collected from which around 100ml were filled in airtight plastic bottles with minimum disturbance. Care was taken in filling the water in bottles to avoid any air bubble and to ensure zero headspace. This is because the existence of any airing during the sampling procedure, might lead to out gassing [12]. The samples were taken to the laboratory with nominal loss of time and subjected to the determination of dissolved Radon by means of bubbler.

4. Results and discussion.

4.1 Estimation of ^{222}Rn in water

Correctly 60 ml of the water sample was moved into the bubbler by the vacuum transfer technique. The water was shifted into a pre-evacuated and background counted scintillation cell. The scintillation cell was kept for 180 min to allow Radon to attain equilibrium with its daughters and then it was attached to a photomultiplier and alpha-counting assembly. The ^{222}Rn concentration was calculated using the equation 1 [13].

$$R_n^{222} (Bq\,l^{-1}) = \frac{6.97 \times 10^{-2} \times D}{V \times E \times (e^{-\lambda T}) \times (e^{-\lambda \theta}) \times (1 - e^{-\lambda t})} \quad (1)$$

Where, D = Counts per second, V = Volume of water (ml), E = Efficiency of the scintillation cell (74 %), λ = Decay constant of Radon ($2.098 \times 10^{-6} \text{ s}^{-1}$), T = Counting delay after sampling, t = Counting duration (s), θ = build-up time in the bubbler (s).

4.2 Estimation of ^{226}Ra in water

Nearly twenty liter of water from each sampling site was collected through Whatman 42 and the pH was made neutral. About 10 mg of MnO_2 powder was poured to the sample, stirred for an hour and permitted to settle for 24 hours. The supernatant was siphoned and the precipitate dissolved with 1N HCl. The solution was dried over hot plate and then converted to HNO_3 medium by treating with 4N HNO_3 . This process is called pre-concentration. The pre-concentrated water of nearly 60ml was transferred to the Radon bubbler (Figure 1). The air was sucked through the sample solution for nearly 5 minutes by means of a vacuum pump to brush it. This would eliminate the solution of dissolved ^{222}Rn . The solution in the bubbler was then permissible to position for a known period of about 3 - 5 half-lives (~21days) of ^{222}Rn , for enough Radon to build up.

At the end of the preferred buildup period, a displaced scintillation cell was associated to the bubbler through the fast connector valve. Under the effect of the vacuum in the scintillation cell, air gets sucked through the solution and fills the scintillation cell. In the process the air transports the liquefied Radon quantitatively. By careful handling of the stopcocks the bubbling was organized and adjusted to be constant and stable to confirm complete handover of Radon into the cell. The cell was kept for a known period of 180 minutes to allow Radon daughters to reach equilibrium with Radon. After the storage period, the cell was connected to the photomultiplier assembly and the alpha activity was counted for a period of 1000 s. The ^{226}Ra concentration in the sample was determined using the equation 4.9 [14].

$$R_a^{226} (\text{mBql}^{-1}) = \frac{6.97 \times 10^{-2} \times D}{V \times E \times (e^{-\lambda T}) \times (1 - e^{-\lambda \theta}) \times (1 - e^{-\lambda t})} \quad (2)$$

Where, D = Counts per second, V = Volume of water (20l), E = Efficiency of the scintillation cell (74 %), λ = Decay constant of Radon ($2.098 \times 10^{-6} \text{ s}^{-1}$), T = Counting delay after sampling, t = Counting duration (s) and θ = build-up time in the bubbler (s).

It is observed from the present study that, the outdoor gamma absorbed dose varies widely from 43.5 to 269.7 nGyh^{-1} with a geometric mean of 130.5 nGyh^{-1} . The average annual effective dose due to gamma radiations was 160.0 μSv . The indoor absorbed dose varied from 113.1 to 278.4 nGyh^{-1} with a mean of 179.4 and the average annual effective dose was found to be 879.9 μSv . Both outdoor and indoor lowest belong to the location Chandapura and Thattanahalli Anekal taluk with 49 and 545.8 μSvy^{-1} respectively. The nature of soil in this area is mixed red and black. The highest outdoor and indoor annual effective doses were same location Hosakote 330.8 and 1299.5 μSvy^{-1} . These places are surrounded by granitic hills. The presence of granitic formations may be the reason for the elevated radiation dose in these areas. The gamma levels in all the regions were found to be within the normal range [15].

The measured values of absorbed and annual effective gamma dose in different parts of the world were shown in Table 3.

4.3 ^{222}Rn and ^{226}Ra in water using Radon bubbler

The ^{222}Rn and ^{226}Ra concentration in water samples from bore wells collected from different locations of Bangalore rural district were measured by Radon emanometry using a Radon bubbler. The measured parameters of water in different locations were tabulated in Table 1. The Radon concentration varied from 9.2 to 47.2 Bql^{-1} with a geometric mean of 21.3 Bql^{-1} . The ^{226}Ra concentration in water varied from 17.2 to 97.3 mBql^{-1} with an average of 50.22 mBql^{-1} (Figure 3). The recorded Radium concentrations in all the water samples lie below the internationally recommended safe limit of 555 mBql^{-1} and hence safe for drinking purposes [16]. Both ^{226}Ra and ^{222}Rn concentration were found to be highest in Hosakote due to the presence of granite geology in that area. Radon activity concentration in water samples

shows good correlation with Radium activity concentration in water with a correlation coefficient of 0.73 (Figure 4). This suggests that, the concentration of Radon in water is mainly due the presence ^{226}Ra in ground water during percolation.

It has been proved from the earlier research that, the concentration of Radium in the underground water depends on the geochemical properties. In order to check the correlation between Radium concentration with the geochemical properties, the parameters such as, pH, conductivity and Total Dissolved Salt (TDS) were also determined using suitable instruments. The results were tabulated in Table 4 and correlation of Radium concentration in water with pH, Conductivity and TDS were plotted in figure 5, 6 and 7 respectively.

The pH of water in the study area ranged from 8.07 to 8.95 with a mean of 8.56 and a standard deviation of 0.21. At all the locations of the study area, the pH of water exceeded the safe limit recommended by WHO which ranges from 7.02 to 7.25 [17].

There was a poor correlation between the pH and the content of ^{226}Ra , that the concentrations decreased with increasing pH, with correlation coefficients of $r = -0.17$. But the negative correlation suggests that, water with low pH is associated with higher concentration of ^{226}Ra . Similar work carried out by earlier researcher showed better correlation [18]. This result can be ascribed to acid leaching and to the effect of pH on the different chemical behavior of Uranium and Thorium. As pH increases, fewer minerals including Uranium and Thorium were leached from the neighboring rock or sediment [19].

The correlation between conductivity and TDS with ^{226}Ra concentration was low, with correlation coefficient of 0.26 and 0.06 respectively. This indicates that, the variation of conductivity and TDS hardly affect the ^{226}Ra concentration in water. The concentrations of Radon and Radium in water were low in comparison with data available in India and world scenario.

The ^{222}Rn dose to the population can be of two types, viz. the dose from ingestion and from inhalation. The dose due to ingestion mainly depends on the amount of water consumed by human being an year [20]. It is estimated that, on an average an adult, children and infant consume 100, 75 and 50l per annum [21]. Assuming the proportion of the groups to be 0.05, 0.3 and 0.65, the weighted estimate of consumption is 60l per annum.

For inhalation dose, air-water concentration ratio of 10^{-4} is to be taken. The indoor occupancy of 7000 hours per annum and the inhalation dose coefficient of 9nSv and 3.5nSv per Bqhm^{-3} to be taken [22].

The annual effective dose, due to the inhalation of Radon, resulting from the Radon concentration in domestic water and ingestion were calculated using the equations 5.8 and 5.9 respectively [23]. The results were tabulated in Table 5.

$$\text{Inhalation dose (nSvy}^{-1}) = C_{\text{Rn}} \times E_q \times T \times 10^{-4} \times F \quad (3)$$

Where, C_{Rn} = Radon concentration (Bqm^{-3}), E_q = Equilibrium between Radon and its progeny (0.4), T = Indoor occupancy time (7000h/year), F = Dose conversion factor (9nSv per Bqhm^{-3})

$$\text{Ingestion dose (nSvy}^{-1}) = C_{\text{Rn}} \times V \times F \quad (4)$$

Where, C_{Rn} = Radon concentration (Bql^{-1}), V = volume of water consumed per annum (60 l per year), F = Dose conversion factor (3.5nSv per Bq)

The annual inhalation dose resulting from the Radon concentration in domestic water varied from 22.9 to 113.7 μSv with an average of 68.3 μSv . The corresponding annual ingestion dose of Radon ranged from 1.8 to 9.3 μSv with an average of 5.55 μSv .

Conclusion

Radon is water-soluble radioactive noble gas produced from the decay of ^{226}Ra in Uranium series. its presence in drinking water and open air increases the risk of lung and intestinal cancers in human beings. In view of thus Radon concentration in groundwater and its dose due to inhalation and ingestion to the population of Bangalore rural district in Karnataka state. In the present study, the ambient gamma radiation measured using in situ hand held gamma ray meter showed highest value of $331.8 \mu\text{Svy}^{-1}$ in Hosakote and a minimum value of $190.55 \mu\text{Svy}^{-1}$ in chandapura, with an average of $190.55 \mu\text{Svy}^{-1}$ throughout the area under study. The radon concentration in drinking water in the study area showed a minimum value in Sulibele area and a maximum value in devalahalli area with 9.2 and 46.3 Bqm^{-3} respectively. The average value of radon in drinking water is found to be 21.3 Bqm^{-3} . The ^{226}Ra concentration in water was found to be highest in Devanahalli and minimum in thattanahalli with 91.3 and 15.3 mBq/l respectively. The average value of ^{226}Ra concentration in water was 53.25 mBq/l . The higher value of radium concentration may be due to the presence of granitic underground and the lower value may be due to the presence of loamy soil.

The annual inhalation dose resulting from the Radon concentration in domestic water varied from 23.4 to 113.7 μSv with an average of 68.3 μSv . The corresponding annual ingestion dose of Radon ranged from 1.8 to 9.3 μSv with an average of 5.55 μSv . The increase in radon concentration in water may due to the presence of increased amount of radium concentration in the underground rocks. The percolation of water through the cracks of these rocks may result in dissolving small fraction of radium in water. The decay of radium results in the production of radon concentrations in water. The study also revealed that 90% of the 20 sample studied showed higher Radon concentration compared to the UNSCEAR recommendation. The sample have concentration above the maximum permissible level prescribed by WHO. The ground water sample are found to be slightly alkaline within limit of Indian standards values.

Table 1: Ambient Gamma Radiation Level (Monsoon).

Sl. No	Location	γ Exposure Level (μRh^{-1})			γ Absorbed Dose (nGyh^{-1})	γ Effective Dose (μSvy^{-1})
		Number of Readings	Range	Mean		
1	Nelamangala	10	6-12	9.0	77.4	94.0
2	Thyamagondlu	10	14-21	18.0	147.8	189.1
3	Dobbspet	10	15-23	21.0	180.6	221.5
4	Dasanapura	10	14-28	19.0	163.4	201.8
5	Doddaballapura	10	21-34	27.0	235.2	286.7
6	Thubagere	10	17-28	23.0	199.1	241.7
7	Rajankunte	10	12-24	16.0	138.2	169.7
8	Doddabelavangala	10	15-30	22.0	189.6	231.8
9	Devanahalli	10	15-32	23.0	198.98	241.8
10	Bagaluru	10	17-32	24.0	206.9	241.5
11	Southgowdanahalli	10	15-27	24.0	207.6	249.1
12	Yerthigahahalli	10	17-21	20.0	171.0	213.4
13	Hosakote	10	24-36	31.0	267.8	331.8
14	Hasigala	10	6-14	9.0	76.4	95.0
15	Sulibele	10	4-13	7.0	59.9	74.7
16	Mallasandra	10	6-13	10.0	84.0	103.9

17	Anekal	10	12-21	17.0	143.7	177.3
18	Chandapura	10	4-7	5.0	41.7	49.3
19	Thattanahalli	10	6-16	13.0	112.6	131.7
20	Jigani	10	5-10	7.0	59.3	73.7
Min				5.00	41.70	49.3
Max				31.0	267.80	331.8
Mean				18.0	154.75	190.55
STDEV				7.0	60.73	74.5

Table 2: Ambient Gamma Radiation Level (Summer).

Sl. No	Location	γ Exposure Level (μRh^{-1})			γ Absorbed Dose (nGyh^{-1})	γ Effective Dose (μSvy^{-1})
		Number of Readings	Range	Mean		
1	Nelamangala	10	12-25	19.0	162.3	805.9
2	Thyamagondlu	10	15-30	25.0	211.8	1023.0
3	Dobbspet	10	17-30	26.0	220.4	1098.6
4	Dasanapura	10	21-32	27.0	230.7	1123.7
5	Doddaballapura	10	24-33	29.0	249.8	1212.8
6	Thubagere	10	21-31	26.0	223.7	1099.7
7	Rajankunte	10	16-24	20.0	171	847.8
8	Doddabelavangala	10	25-35	32.0	275.6	1361.9
9	Devanahalli	10	23-34	31.0	267.8	1298.0
10	Bagaluru	10	21-32	27.0	229.9	1109.3
11	Southegowdanahalli	10	19-33	27.0	231.7	1103.6
12	Yerthigahahalli	10	19-32	26.0	221.7	1069.8
13	Hosakote	10	24-34	31.0	265.8	1299.5
14	Hasigala	10	10-20	15.0	128.8	623.6
15	Sulibele	10	10-20	14.0	119.6	588.4
16	Mallasandra	10	9-19	16.0	137.4	633.4
17	Anekal	10	16-23	21.0	179.8	856.8
18	Chandapura	10	9-19	13.0	111.7	543.9
19	Thattanahalli	10	9-15	14.0	119.8	545.8
20	Jigani	10	10-24	18.0	151.9	735.8
Min				13.0	111.7	543.9
Max				32.0	275.6	1299.5
Mean				22.5	193.65	952.9
STDEV				5.8	50.2	246.4

Table 3: The average absorbed gamma dose and effective dose in different environments.

Region	Absorbed dose (nGyh^{-1})		Effective dose (mSvy^{-1})		Reference
	Indoor	Outdoor	Indoor	Outdoor	
Bangalore rural	193.65	154.7	0.881	0.16	Present work
Bangalore	384.1	412.0	0.88	0.51	[24]
Mysore	75.7	40.3	0.36	0.06	[25]
Indian average	----	56	----	0.08	[26]

China	99	62	0.55	0.09	[26]
Bulgaria	75	70	0.45	0.10	[26]
Denmark	63	38	0.36	0.05	[26]
Finland	80	63	0.49	0.08	[26]
Germany	53	70	0.41	0.10	[26]
Japan	50	49	0.32	0.07	[26]
Norway	95	73	0.48	0.10	[26]
Spain	68	43	0.40	0.06	[26]
Sweden	110	56	0.65	0.08	[26]
U.K.	60	34	0.35	0.05	[26]
U.S.A.	38	47	0.28	0.07	[26]
Global average	83	57	0.45	0.08	[27]

Table 4: Radium, Radon, pH, Conductivity and TDS in water samples collected from Bangalore rural district.

Sl no.	Location	pH	Conductivity (mS/cm)	TDS (g/l)	²²² Rn in water (Bq/l)	²²⁶ Ra Conc. (mBq/l)
1	Nelamangala	8.01	0.33	0.18	10.9	26.3
2	Thyamagondlu	8.26	0.41	0.37	14.2	32.3
3	Dobbspet	8.29	0.44	0.38	33.5	43.3
4	Dasanapura	8.47	0.86	0.56	22.1	49.3
5	Doddaballapura	8.23	1.29	1.36	15.2	22.4
6	Thubagere	8.26	0.40	0.38	14.7	30.6
7	Rajankunte	8.51	0.68	0.18	16.4	51.2
8	Doddabelavangala	8.55	0.70	0.54	24.7	80.6
9	Devanahalli	8.18	0.75	1.57	44.2	91.3
10	Bagaluru	8.17	1.69	1.76	24.9	70.6
11	Southegowdanahalli	8.64	0.45	1.36	18.7	62.5
12	Yerthigahahalli	8.57	0.43	0.98	20.8	41.5
13	Hosakote	8.84	0.74	0.78	27.3	51.4
14	Hasigala	8.40	0.26	2.68	16.6	59.8
15	Sulibele	8.29	1.29	1.37	11.2	32.4
16	Mallasandra	8.41	0.61	2.11	24.4	68.2
17	Anekal	8.22	1.10	1.77	11.7	24.5
18	Chandapura	8.35	0.65	1.57	11.9	22.1
19	Thattanahalli	8.16	0.46	1.16	10.3	15.2
20	Jigani	8.62	0.36	0.78	16.2	33.8
Min		8.01	0.26	0.18	10.3	15.2
Max		8.64	1.69	2.68	44.2	91.3
Mean		8.32	0.975	1.43	27.25	53.25
STDEV		0.21	0.45	0.61	9.1	22.60

Table 5: Annual inhalation and ingestion dose due to Radon.

Sl no.	Location	^{222}R in water (Bqm^{-3}) $\times 10^3$	Annual dose (μSv)	
			Inhalation	Ingestion
1	Nelamangala	11.1	27.8	2.3
2	Thyamagondlu	17.2	42.7	3.3
3	Dobbspet	33.4	83.4	7.4
4	Dasanapura	23.4	59.9	5.3
5	Doddaballapura	13.3	34.3	3.3
6	Thubagere	9.8	26.3	2.4
7	Rajankunte	13.8	36.2	3.3
8	Doddabelavangala	27.9	73.1	6.3
9	Devanahalli	46.3	113.7	9.3
10	Bagaluru	26.9	67.8	5.4
11	Southegowdanahalli	19.7	49.6	4.3
12	Yerthigahahalli	21.8	54.9	4.1
13	Hosakote	28.3	71.3	5.3
14	Hasigala	17.6	44.4	3.4
15	Sulibele	9.2	23.2	1.8
16	Mallasandra	26.4	66.5	5.3
17	Anekal	10.7	27.0	2.0
18	Chandapura	11.6	29.2	2.3
19	Thattanahalli	9.3	23.4	1.8
20	Jigani	13.2	33.3	2.6
Min		9.2	22.9	1.8
Max		46.3	113.7	9.3
Mean		21.3	52.8	5.55
STDEV		9.1	22.9	1.9

Figure 1: Schematic diagram of Radon bubbler

Figure 2: Bangalore rural map

Figure 3. Activity concentration of ^{226}Ra in water samples from different locations of Bangalore rural. Figure 4: Correlation between ^{226}Ra and ^{222}Rn concentration in water.

Figure 5: Correlation between pH of water and ^{226}Ra content in water. Figure 6: Correlation between conductivity of water and ^{226}Ra content in water.

Figure 7: Correlation between TDS of water and ^{226}Ra content in water.

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