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Study of Uranium Concentration in Drinking Water in Tiruvannamalai District, Tamil Nadu, India and Assessment of Health Risk

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Abstract: Natural uranium is found in various sources of water and its assessment in drinking water is important from viewpoint of public health. Intake of uranium through air and water is normally low, but in circumstances in which uranium is present in a drinking water source, the majority of intake can be through drinking water route. Present report is a preliminary study on evaluation of uranium in ground water of Tiruvannamalai district, Tamilnadu, India. Water samples were randomly collected from hand pumps around Tiruvannamalaidist, Tamilnadu. Concentrations of uranium in the samples were measured using the LED fluorimetric technique. Results showed variations in concentrations obtained from place to place and values ranged from1.9 to 23.4 ppm with the mean value of 6.94ppm.The measured values were found lower to permissible limits of AERB, USEPA, WHO, UNSCEAR and ICRP. Risk assessment of uranium in water is also calculated using life time cancer risk, life time average daily dose and hazard quotient. Finally it was concluded that there was no excess lifetime cancer risk and chemical risk to humans due to uranium concentration in water of the study area.
Keywords: Uranium concentration, Drinking Water**,** Lifetime Average Daily Dose (LADD), Hazard Quotient (HQ), Annual effective dose (D_E).

1 Introduction

Uranium is a naturally occurring radioactive metallic element. Small amounts of uranium are present in certain types of soils and rocks, especially granite. Natural uranium comprises of three important isotopes: uranium-234, uranium-235 and uranium-238. More than 99 percent of uranium found in nature is uranium-238. Uranium breaks down very slowly into other elements including radium and radon gas. [1-4].

Most natural waters contain detectable amounts of uranium. Uranium occurs more often in bedrock and deep bedrock wells are more likely than shallow wells to have elevated levels of uranium. The amount of uranium in well water will vary greatly from place to place. Testing is the only way to determine if water contains uranium. The concentration of uranium in a well water sample depends on factors such as the amount of uranium present in the rock through which the groundwater has passed and whether the water chemistry is favorable for uranium to remain dissolved. When ground water dissolves minerals that contain naturally occurring deposits of uranium, it can enter into drinking water. Another way is through human activity such as mining. Uranium and other elements are naturally occurring in ground water. The amount of uranium in drinking water depends on the natural levels in the area where the water system obtains its raw water.Intake of uranium through air is extremely low, and it appears that intake through food is between 1 and 4 µg/day. Intake through drinking-water is normally low; however, in circumstances in which uranium is present in a drinking-water source, the majority of intake can be through drinking-water.

Natural sources like leaching from natural deposits and anthropogenic sources like release in mill tailings, combustion of coal and other fuels, and the use of uranium containing phosphate fertilizers are responsible

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factors for uranium contamination of groundwater [5-6]. To prevent the adverse health effects of uranium in drinking water, it is recommended to monitor the uranium concentration in different drinking water resources

The first aim of the present work is to determine the uranium concentration in drinking water and effective dose to the public as a result of consumption of water in and around Tiruvannamalai city, Tamilnadu using LED fluorimetric technique.

2 Materials and Methods

2.1 Sampling

Samples were collected from 11 different locations from bore wells. Sampling sites are shown in the Figure 1. Before collecting the samples, water from the source was made to run out for 7-10 min to ensure that the fresh sample from the aquifer was taken. The collected subsurface water samples were filtered through 0.45 micron filter paper, acidified with 0.01M nitric acid (AR Grade, Merck) and stored in, acid washed, 200 ml capacity polypropylene bottles. Samples were collected in air-tight lab grade polypropylene bottles of 200 ml capacity. pH, TDS and Temperature of the samples were measured within 2 days of sampling. The longitude and latitude of study area is given in Table 1.

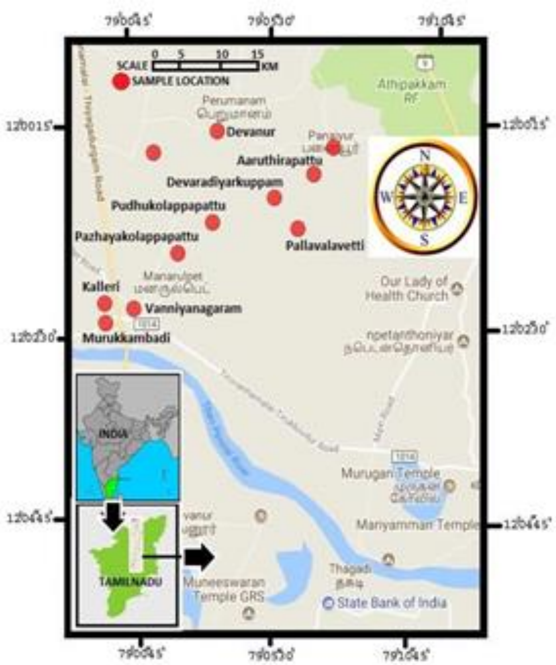

Fig. 1: Location map of the study area.

2.2 Measurement of Uranium in Ground Water Samples

Samples were analyzed for uranium content using LED fluorimeter (Quantalase LF-2a). Quality assurance of the data was made by the analysis of IAEA standard reference materials and by replicate analysis and spike recovery. Fluorescence yield varies for different complexes of uranium. Therefore an inorganic reagent Fluren (Fluorescence Enhancing Reagent) was added to the sample to convert all the complexes into a single form having same fluorescence yield. 6 ml of the sample with 10% fluren was taken in a cuvette made from ultra-low fluorescence fused silica and then they were analyzed for uranium in the fluorimeter.

3 Results and Discussion

3.1 Distribution of Uranium in Ground Water

Groundwater samples were collected from different villages in TiruvannamalaiDist, Tamilnadu analyzed for uranium concentration using LED Flourimeter together with location and radiological parameters are presented in Table 2. The concentration of uranium varied from 1.9 to 23.4µg L^{-1} with a mean value of 6.94 µg L^{-1}. The safe limit of uranium in groundwater is fixed to be 60 ppb by AERB in India, while other agencies fix it in much lower limits of 30 ppb [7]; 15 ppb [8]; 9 ppb [9] and 1.9 ppb [10].

In comparison of with WHO, UNSCEAR and ICRP safe limits, the values obtained for uranium in groundwater are very much lower than the safe limits suggested by USEPA, WHO, UNSCEAR and ICRP, AERB [7,10,11] Fig 2 shows locations with uranium concentrations. Table 3 lists the comparison of uranium concentration of present work with other cites of India.

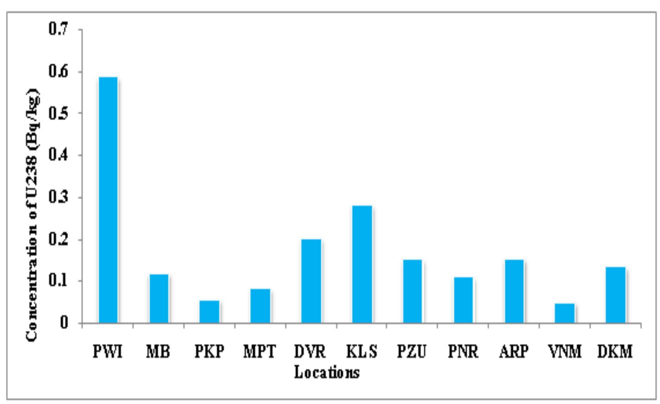

Fig. 2: Locations versus Uranium Concentration.

3.2 Radiological Risk Assessment

3.2.1 Risk Co-Efficient Mortality

Mortality is the term used for the number of people who died within a population. Morbidity refers to the

unhealthy state of an individual, while mortality refers to the state of being mortal. Both concepts can be applied at the individual level or across a population. For example, a morbidity rate looks at the incidence of a disease across a population and/or geographic location during a single year. Mortality rate is the rate of death in a population. The two are often used together to calculate the prevalence of a disease - e.g., measles - and how likely that disease is to be deadly, particularly for certain demographics.
Radiological risk which is also expressed as Excess cancer risk (ECR) is evaluated using the following equations [12].

ECR = U Conc. In Ground Water (Bq/L) x Risk

Factor (Mortality) (Per Bq/L) ------(1)

U Conc (Bq/L) = Measured Value of U (μg/L) x

Conversion Factor (0.025 Bq/L)

Risk Factor = Risk Co-eff. (Bq^{-1}) x Water

Ingestion Rate (L/Day) x T.E.D (Days) ----- (2)
Where,

T.E.D - Total Exposure Duration.
Risk Coefficient for mortality in equation (2) was taken as 1.19×10^{-6} Bq^{-1} Water ingestion rate was taken as 1.38 L/Day and total exposure duration was taken as 25509 days.
The calculated values of the ECR Mortality for the Studied samples are presented in Table 2. The cancer risk for mortality found to be varying in the range from 0.17×10^{-6} to 8.36×10^{-6} with the mean value of 4.08×10^{-6}.

3.2.2 RiskCo-Efficient Morbidity

Morbidity refers to the state of being diseased or unhealthy within a population. Radiological risk which is also expressed as Excess cancer risk is evaluated using the following equations [12].
ECR = U Conc. In Ground Water (Bq/L) x Risk

Factor (Morbidity) (Per Bq/L) -----(3)

Where , ECR is Excess Cancer Risk

U Conc. (Bq/L) = Measured Value of U (μg/L) x

Conversion Factor (0.025 Bq/L

Risk Factor = Risk Co-eff. (Bq-1) x Water Ingestion

Rate (L/Day) x T.E.D (Days) ----(4)

Where, TED is Total Exposure Duration.Risk Coefficient for mortality in equation (4) was taken as 1.84×10^{-6} Bq^{-1} Water ingestion rate was taken as 1.38 L/Day and total exposure duration was taken as 25509 days. The calculated values of the ECR Morbidity for the studied samples are presented in Table 2.

Table 1: Geographical data of Study Area.

S No.	Sample ID	Location	Latitude (N)	Longitude (E)
1	DVR	Devanur	12° 2' 16.5624" N	79° 6' 18.4212" E
2	PNR	Panaiyur	12° 2' 6.8928" N	79° 7' 35.0508" E
3	MPT	Manalurpet	12° 0' 31.9032" N	79° 5' 23.73" E
4	KLI	Kalleri	12° 2' 3.966" N	79° 5' 36.3984" E
5	PKP	Pudhukolappapattu	12° 1' 4.7136" N	79° 5' 52.4652" E
6	PZA	Pazhayakolappapattu	12° 1' 22.8648" N	79° 6' 15.3288" E
7	DKM	Devaradiyarkuppam	12° 1' 37.3728" N	79° 6' 56.1168" E
8	PVI	Pallavalavetti	12° 1' 51.2724" N	79° 7' 22.0728" E
9	ARP	Aaruthirapattu	12° 1' 19.2396" N	79° 7' 11.5644" E
10	MBD	Murukkampadi	12° 0' 35.1144" N	79° 5' 4.2612" E
11	VNM	Vanniyanagaram	12° 0' 23.6304" N	79° 5' 4.8804" E

Table 2: Uranium Concentration, ECR Mortality, ECR Morbidity of Study area, Lifetime Average Daily Dose (LADD), Hazard Quotient (HQ), Annual effective dose (DE) (μSv/year), Cumulative Dose.

S. No	1	2	3	4	5	6	7	8	9	10	11	**average**
Location ID	**DVR**	**PNR**	**MPT**	**KLI**	**PKP**	**PZA**	**DKM**	**PVI**	**ARP**	**MBD**	**VNM**	
U^{238}	8	4.3	3.2	11.2	2.2	6	5.3	23.4	6.1	4.7	1.9	**6.94**
μg/L	8	4.3	3.2	11.2	2.2	6	5.3	23.4	6.1	4.7	1.9	**6.94**
Bq/L	0.2	0.1075	0.08	0.28	0.055	0.15	0.1325	0.585	0.1525	0.1175	0.0475	**0.17**
Risk Coefficient of Mortality × 10^{-5}	4.18	4.18	4.18	4.18	4.18	4.18	4.18	4.18	4.18	4.18	4.18	4.18
Risk Coefficient of Morbidity × 10^{-5}	6.47	6.47	6.47	6.47	6.47	6.47	6.47	6.47	6.47	6.47	6.47	6.47
ECR Mortality × 10^{-6}	8.36	4.49	3.34	1.17	2.30	6.27	5.54	0.24	6.37	4.91	1.99	**4.08**
ECR Morbidity × 10^{-6}	0.12	6.96	5.18	0.181	3.56	9.71	8.57	0.37	9.87	7.60	3.07	**5.01**
Lifetime Average Daily Dose (LADD)	0.1577	0.0848	0.0631	0.2208	0.0434	0.8311	0.1045	0.4613	0.1203	0.0927	0.0375	**0.1367**
Hazard Quotient (HQ)	0.2629	0.14.13	0.1051	0.3680	0.0723	0.1971	0.1741	0.7689	0.2004	0.1544	0.0624	**0.2279**
Annual effective dose DE (μSv/year) × 10^{-6}	4.5333	2.4366	1.8133	6.3466	1.2466	3.3999	3.0033	0.1325	3.4566	2.6633	1.0766	**2.7371**
DE	2.2099	5.3832	6.2899	0.2833	5.6666	3.9099	0.5099	1.3599	8.6699	7.0266	9.6699	**3.0600**
Cummulative Dose	154.693	376.824	440.293	19.831	396.662	273.693	35.693	95.193	606.893	491.862	676.893	**324.4**

Table 3: Comparison of Uranium concentration of present work with other cites of India.

S. No.	Name of the Cites	Basic Source	Uranium concentration (µg/L)	References
1	Himachal Pradesh	Groundwater	0.56 – 10.11	[13]
2	ShriGanganagar (Rajasthan)	Groundwater	2.5 – 171	[14]
3	Churu (Rajasthan)	Groundwater	13 – 95	[14]
4	Khalilabad, Gorakhpur, Maharajganj, Kushinagar (Uttar Pradesh)	Bore well, River water Tap water, open well	0.02 – 64.00	[15]
5	Fatehabad (Haryana)	Groundwater	0.3 – 48	[16]
6	Western Haryana	Groundwater	6.37 – 43.31	[17]
7	Mansa (Punjab)	Groundwater	5.90 – 645.22	[18]
8	Bathinda (Punjab)	Groundwater	7.0 – 323.94	[18]
9	Amritsar (Punjab)	Groundwater	0.87 – 42.51	[18]
10	Hoshiarpur (punjab)	Groundwater	0.48 – 25.19	[18]
11	**Present study**	**Groundwater**	**1.9 - 23.4**	**-**

The cancer risk for mortality and morbidity was found to be varying in the range from 0.12 x 10^{-6} to 9.87 x 10^{-6} with the mean value of 5.01 x 10^{-6}. Fig 3.shows variation of ECR mortality and morbidity with different locations.

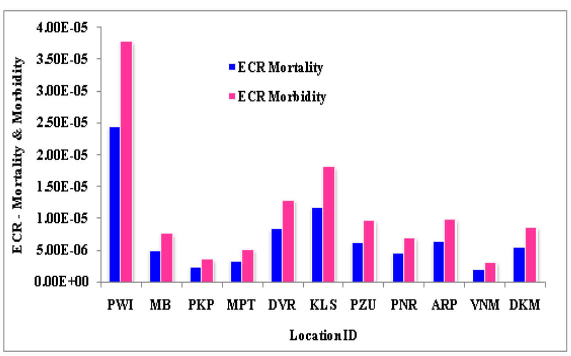

Fig.3: LocationsVs ECR Mortality and ECR Morbidity.

3.3 Chemical Risk Assessment

3.3.1 Life Time Average Daily dose

Chemical toxicity risk associated with any element is evaluated in terms of LADD (Lifetime Average Daily Dose). This can be estimated using the following equations [19].

$$\textbf{LADD } (\mu g / kg / Day) = [Ci \times IR \times EF \times LE] / [BW \times AT] \text{ ----- (5)}$$

Where, *Ci* in equation (5) is the conc. of U in groundwater (µg/L), IR is the ingestion rate (L/Day) which is taken to be 1.38 L/Day. EF is the exposure frequency (Days/year) which is taken to be 365 days per year. LE is the life expectancy (Years) which is taken as 69.89 years. BW isthebodyweight(kg) which is taken as 70 kg. AT is the average time (Days) which is taken as 25509 days. The calculated values of the Life time Average Daily dose for the studied samples are presented in Table 2. Chemical toxicity risk (LADD value) was varying in the range of 0.0375 to 0.8311 µg/Kg/Day with the mean value of 0.1367 µg/Kg/Day. The obtained values of Chemical toxicity risk (LADD value) in the studied samples are well below the recommended limit of 1.0 µg/Kg/Day (WHO, 2011). Fig 4.shows variation of Life time Average Daily dose with different locations.

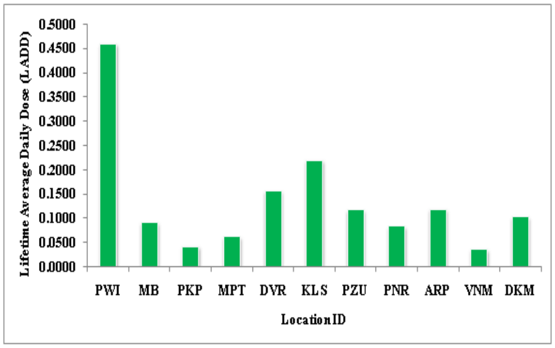

Fig.4: LocationsVs Life time Average Daily dose (LADD).

3.3.2 Hazard Quotient (HQ)

A hazard quotient is the ratio of the potential exposure to a substance and the level at which no adverse effects are expected. If the Hazard Quotient is calculated to be less than 1, then no adverse health effects are expected as a result of exposure. If the Hazard Quotient is greater than 1, then adverse health effects are possible. The Hazard Quotient cannot be translated to a probability that adverse health effects will occur, and is unlikely to be proportional to risk. The hazard quotientcan be estimated using the following equation [19].

$$HQ = LADD/Rfd \text{ ------ (6)}$$

Where Lifetime Average Daily Dose (LADD) and Rfd is said to be the Reference dose (µg / kg / Day) which is taken as 0.6 µg / kg / Day[14]. The calculated values of the hazard quotient for the studied samples are presented in Table 2. Hazard quotient (HQ) was found to be varying from 0.0624 – 0.7689 with the mean value of 0.2279. The hazard quotient values of all the samples is well below the recommended value of hazard < 1.0 which is proposed by AERB [11] and WHO [8]. Fig 5 shows variation of Hazard Quotient with different locations.

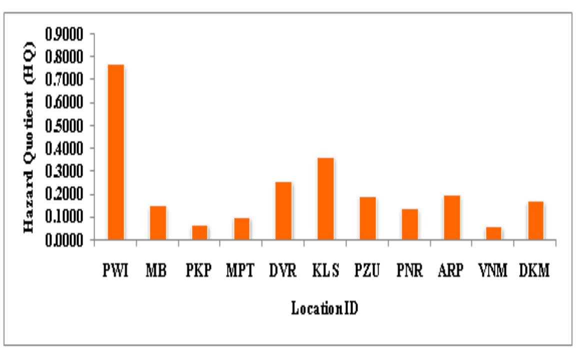

Fig. 5: Location ID versus Hazard Quotient (HQ).

3.3.3Assessment of Annual effective Dose

Effective dose is a dose quantity in the International Commission on Radiological Protection (ICRP) system of radiological protection. It is the tissue-weighted sum of the equivalent doses in all specified tissues and organs of the human body and represents the stochastic health risk to the whole body, which is the probability of cancer induction and genetic effects, of low levels of ionising radiation. Annual effective dose (µSv y^{-1}) for human (for different age groups) due to uranium consumption was quantified using Equation below [20].

$$D\ (\mu Sv y^{-1}) = AC \times F \times I_{annual} \text{------- (7)}$$

Where AC is the activity concentration of uranium (Bq/L), F is effective dose per unit intake (µSv y^{-1}/ Bq/L) which is taken to be 4.5×10^{-8} and I_{annual} is the annual ingestion which was taking to be 503.7L (1.38 × 365). The results for annual effective radiation dose due to intake of uranium through the drinking water for different age groups are presented in Table 2. The estimated annual effective dose due to the intake of uranium through drinking water for various groups ranged from 0.1325 to 6.3466 µSv y^{-1} with an average value of 2.7371×10^{-6}µSv y^{-1}. The recommended level of annual effective dose to human from water consumption is 100 µSv y^{-1}[8]. The annual effective dose was found to be lower than the WHO recommended level of 100 µSv y^{-1}. Fig 6.shows variation of annual effective dose with different locations.

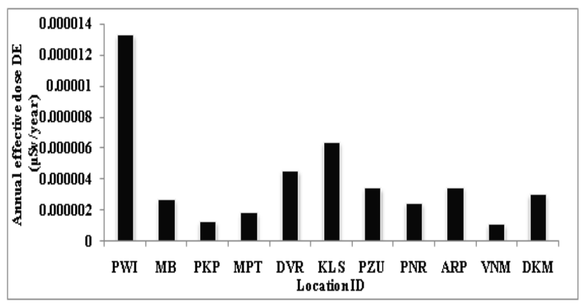

Fig. 6: Location ID versus Annual effective dose DE (µSv/year**).**

3.3.4 Cumulative Dose

Cumulative dose is the total dose resulting from repeated exposures of ionizing radiation to an occupationally exposed worker to the same portion of the body or to the whole body, over a period of time. Annual effective dose, when calculated for the whole of the lifetime is said to be the cumulative dose. The cumulative dose can be estimated by the equation

$$CD = \text{Annual effective dose} \times \text{Average life time} \text{ ------- (8)}$$

Where Annual effective dose is taken from the eqn (7) and average life time of human beings is taken as 70 yrs.

The calculated values of the cumulative dose for the studied samples are presented in Table 2. It is found to be varying in the range from 19.831 – 676.893 µSv with the mean value of 324.4 µSv. Fig 7. shows variation of cumulative dose with different locations.

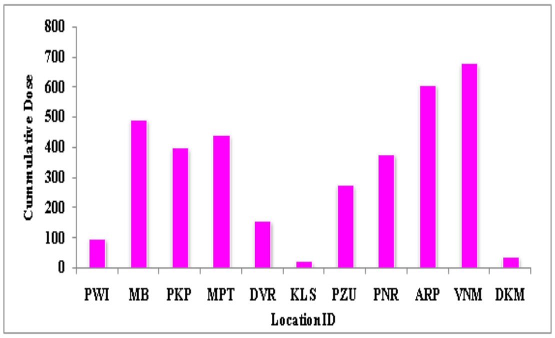

Figure 7. Location ID versus Cumulative Dose

4 Conclusions

The uranium concentration in none of the ground water samples in this study were exceeding the permissible limits of WHO, USEPA, ICRP and UNSCAR. This study has provided an insight into uranium levels in drinking water in rural areas of some villages in Tiruvannamalaidist, Tamilnadu. The laser fluorimetric technique was observed to be very efficient for the analysis of trace level uranium concentrations in water. The results of the measurements could be of vital in radio-epidemiological assessment, diagnosis and prognosis of uranium induced diseases in the local population of the area under investigation. It is suggested that constant monitoring and detailed investigations are warranted to get baseline values of uranium concentration in the ground water of this region. A more systematic study covering wider area in drinking water is highly warranted.

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