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Risk Assessment for Natural Uranium Present in Ground Water of Mahendragarh district of Haryana

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Abstract: In the present study, the groundwater samples from 30 locations in Mahendragarh district of Haryana state were collected and analyzed for uranium concentration using L.E.D Fluorimeter Quantalase LF-2a. This fluorimeter can measure uranium concentration in water samples from 0.5 µg/L to 1000 µg/L with an accuracy of +/- 10% or 0.05 ppb whichever is greater and repeatability of better than +/- 5%. The uranium conc. Was found to be varying in a range of 0.56 – 57.53 µg/L with the mean value of 18.87 µg/L**.** The cancer risk for mortality and morbidity was calculated and found to be in the range from 5.89 x 10^{-7} – 6.03 x 10^{-5} with the mean value of 1.98 x 10^{-5} and in the range from 9.11 x 10^{-7} – 9.32 x 10^{-5} with the mean value of 3.06 x 10^{-5} respectively. Chemical toxicity risk, which is also expressed as Lifetime Average Daily Dose (LADD) value was also calculated and found to be varying in a range of 0.01 – 1.13 µg/kg/Day with the mean value of 0.37 µg/kg/Day. Annual Effective Dose due to ingestion was found to be varying from 0.32 – 32.60 µSv/Year with the mean value of 10.69 µSv/Year.

Keywords: Uranium, Ground water, L.E.D Fluorimeter, E.C.R, Chemical Toxicity Risk, Radiological Risk, Annual Effective Dose, pH, Total Dissolved Solids (TDS), Cumulative Dose

1 Introduction

Uranium is an ubiquitous heavy element occurring naturally which becomes even more concentrated due to anthropogenic activities like due to the wastages from the nuclear industries, due to the combustion of coal and other biofuels and due to phosphate fertilizers also [1]. Natural uranium is actually the mixture of three isotopic forms, out of which two i.e. U^{238} (99.28 %) and U^{235} (0.7186 %) are primordial and one is the decay product U^{234} (0.0053%). Half Life of U^{238}, U^{235} and U^{234} is 4.51 x 10^9 years, 7.07 x 10^8 years and 2.35 x 10^5 years respectively. Pitchblende, uraninite, carnotite, autunite, and torbenite are some of the important ores of uranium. Uranium is mainly found in the oxidation states of +3, +4, +5, and +6 and the most common out of these are the tetravalent and the hexavalent oxidation states. Uranium (+4) is insoluble and can form complexes by various inorganic ligands such as fluoride, chloride, sulphate, and phosphate. However, the greater solubility of U (+6) as the uranyl (UO2++) compounds, is due to its ability to form stable complexes with various organic and inorganic ligands [2]. Natural uranium (U^{238}, U^{235} and U^{234}) and its daughter products (e.g. Ra^{226}, Rn^{222}, Pb^{210}, Po^{210} etc.) content in groundwater is often associated with the presence of granitic rocks. Dissolution of gases, rock minerals and recoil nuclides are the main causes of the presence of uranium in natural waters. Very high uranium concentration is generally observed in brine waters and spring waters [1]. Uranium is well known to be a toxic element both radiologically and chemically. In the year 1991, the United States Environmental Protection Agency (USEPA) classified uranium as a carcinogenic element (group A) and suggested that the complete absence of uranium in drinking water only should be the safe limit for the carcinogenic risk. At present, both, the United States Environmental Protection Agency (USEPA) and the World Health Organization (WHO) has proposed a realistic regulation level as maximum contaminant level (MCL) of 30 ppb [3, 4]. The major adverse health effects of uranium are due to its chemical toxicity, rather than due to its radiological hazards [5-6]. Adverse effects of this metal on kidneys are well established [7]. Ingestion through water and food are its primary sources and then it accumulates preferably in the liver, kidneys and bones [8]. Of absorbed uranium, 66% is rapidly eliminated via urine, while the rest is distributed and deposits in the kidneys (12-25 %), bone (10-15 %) and soft tissue [9]. Keeping in view its adverse effects on human health, it becomes really very important to calculate the radiological and chemical risks associated with this element.

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Therefore, in this study we have measured the uranium conc. in groundwater from different regions of Mahendragarh district and also have calculated both the radiological and chemical risks. These results can be used to establish the regulation standards and management schemes in the area.

2 Geology of Study Area

The study area shown in Figure 1 occupies the southern extremity of the Haryana state jointly with Rewari and Gurgaon districts of Haryana. It has a total geographical area of 1899 sq. km. and falls between Latitudes 27°48'10" and 28°8'30" and Longitudes 75°54'00" and 76°51'30" at an average elevation of 262 m from sea level. Mahendragarh district is bounded by Bhiwani and Rohtak districts in its north, Rewari in its east and Alwar and Jhunjhunu (Rajasthan) districts in its south and west respectively. The district is comprised of 370 villages and 5 towns with the population of 9,21,680 souls as per 2011 census. The area forms the part of Indo - Gangetic plains and has vast alluvial and sandy tracts. It is interspersed with strike ridges which are occasionally covered by blown sands. Southwestern part of the district is occupied by blown sand and alluvium. The hill ranges are marked features of the district and are part of great Aravali chain. Light colored arid soils are found in the major part of the district. These soils are calcareous and have lime nodules in the subsurface horizons. Most of the soils in the district are medium textured.

The water supply of the district is mainly based on ground water through tube wells. The water supply of the villages is met out through installation of hand pumps and construction of dug wells by the local villagers.

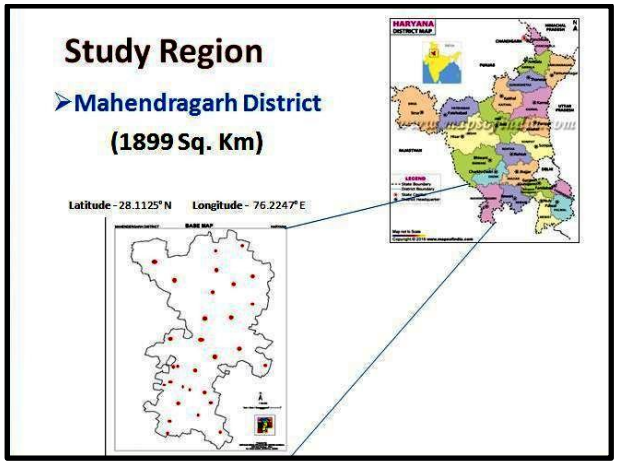

Figure 1. Map of the Study Area

Water for irrigation in the district is also based mainly on groundwater. Out of total irrigated area of 1210 sq km an area of 1190 sq km is based on ground water irrigation. Only in 20 sq km area irrigation is based on canals. Ground water is being extracted through large no of tube wells and dug wells in the district [10].

3 Experimental Techniques

3.1 Sampling

Samples were collected from 30 different locations from bore wells and tube wells. Sampling sites are shown by the red dots in the figure-2. 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.

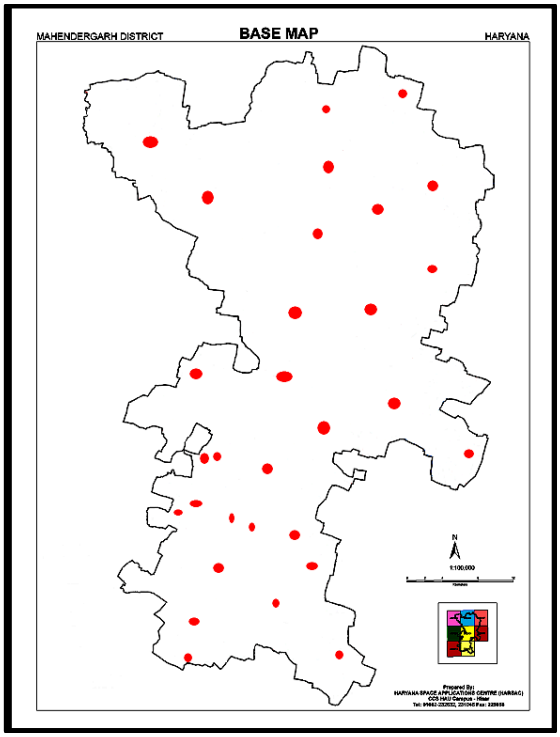

Figure 2. Sampling Sites In The Study Area.

Samples were collected in air-tight lab grade polypropylene bottles of 30 ml capacity. After collecting water samples they were filtered through a filter paper of pore size 0.45 micron before analyzing them for uranium conc. pH and TDS of the samples were measured within 2 days of sampling.

3.2 Measurement of Uranium in Samples

Samples were analyzed for uranium content using LED fluorimeter (Quantalase LF- 2a) shown in Fig 3. 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

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10% fluren was taken in a cuvette made from ultra-low fluorescence fused silica and then they were analyzed for uranium in the fluorimeter.

Figure 3. LED Fluorimeter (Quantalase LF-2a)

4 Methodology for Risk Assessment

In the present study, two types of risks are evaluated separately which are associated with uranium. One is the radiological risk which is due to the ionizing radiations emitted by uranium and the other is the chemical risk. Uranium is a heavy metal and it is hazardous to human health. As it was stated earlier also that the major adverse health effects of uranium are due to its chemical toxicity rather than the radiological hazards. So it becomes really very important to calculate the risks associated with it.

5 Results and Discussion

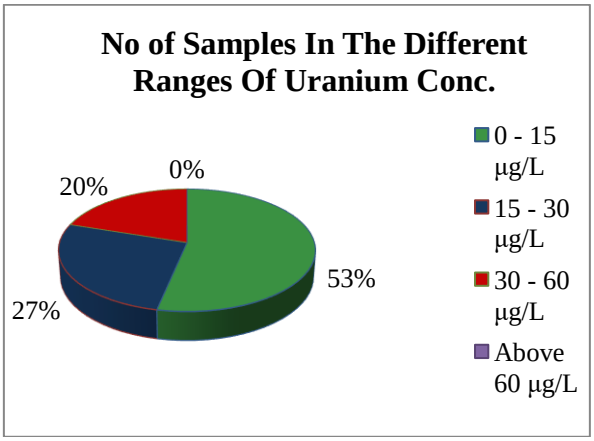

Figure 4. Pi Chart Of Different Conc. Interval Of U In Water Samples.

Uranium content of the groundwater samples of Mahendragarh district and its corresponding risks are tabulated in Table 1. The samples were analyzed by L.E.D fluorimeter Quantalase LF-2a. The uranium conc. was found to be varying in the range 0.56 – 57.53 µg/L with the mean value of 18.87 µg/L**.**

Out of 30 analyzed samples, 16 (53.33%) were found to be below 15 µg/L which is the recommended permissible limit by WHO (2004) [11]. 24 samples out of 30 (80%) were found to be below 30 µg/L which is the recommended permissible limit by USEPA [3]. 6 samples out of 30 (20%) were found to be above 30 µg/L but none of the samples was found to be above 60 µg/L which is the recommended permissible limit by AERB (AERB, DAE) [12]. No. of samples in different conc. ranges are shown in the form of a pi chart in figure 4.

5.1 Radiological Risk Assessment

Radiological risk which is also expressed as Excess cancer risk is evaluated using the following equations [13].

$$\text{Excess Cancer Risk} = U\ Conc.\ In\ Ground\ Water\ (Bq/L) \times Risk\ Factor\ (Per\ Bq/L) \quad (1)$$

$$\text{U Conc. (Bq/L)} = Measured\ Value\ Of\ U\ (\mu g/L) \times Conversion\ Factor\ (0.025\ Bq/L)$$

$$\text{Risk Factor} = Risk\ Coefficient\ (Bq^{-1}) \times Water\ Ingestion\ Rate\ (L/Day) \times Total\ Exposure\ Duration\ (Days) \quad (2)$$

Risk Coefficient for mortality and morbidity in equation (2) was taken as 1.19×10^{-9} Bq^{-1} and 1.84×10^{-9} Bq^{-1} respectively. Water ingestion rate was taken as 1.38 L/Day and total exposure duration was taken as 25509 days.

Risk Factor for mortality and morbidity was calculated to be 4.19×10^{-5} and 6.48×10^{-5} respectively. The cancer risk for mortality and morbidity was found to be varying in the range from 5.89×10^{-7} – 6.03×10^{-5} with the mean value of 1.98×10^{-5} and in the range from 9.11×10^{-7} – 9.32×10^{-5} with the mean value of 3.06×10^{-5} respectively.

5.2 Chemical Risk Assessment

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 [5].

$$\text{LADD (µg / kg / Day)} = \frac{[Ci \times IR \times EF \times LE]}{[BW \times AT]} \quad (3)$$

$$\text{HQ} = \frac{LADD}{Rfd} \quad (4)$$

Where Ci in equation (3) 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 is the body weight (kg) which is taken as 70 kg. AT is the average time (Days) which is taken as 25509 days. In Equation (4) HQ is said to be the Hazard Quotient and Rfd is said to be the Reference dose (µg / kg / Day) which is taken as 0.6 µg / kg / Day [5].

Chemical toxicity risk (LADD value) was varying in the range of 0.01 – 1.13 µg/Kg/Day with the mean value of 0.37 µg/Kg/Day. Hazard Quotient (HQ) was found to be

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varying from 0.02 – 1.89 with the mean value of 0.62.

5.3 Assessment of Annual Effective Dose

Annual Effective Dose is the measure of the whole body dose. It was estimated using the conversion factors given by W.H.O. [5].

Table 1: Conc. Of U and Calculated Radiological And Chemical Risks Associated With Each Water Sample

Sr. No.	Location	Conc. Of U (µg/L)	Ac (Bq/L)	R (Mortality)	R (Morbidity)	ECR (Mortality)	ECR (Morbidity)	LADD (µg/Kg/Day)	HQ	D_E (µSv/Year)	Cumulative Dose (µSv)
1	Kheri	13.61	0.34	4.19E-05	6.48E-05	1.43E-05	2.20E-05	0.27	0.45	7.71	539.17
2	Ateli Mandi	25.62	0.64	4.19E-05	6.48E-05	2.68E-05	4.15E-05	0.51	0.84	14.52	1014.53
3	Mirzapur	16.84	0.42	4.19E-05	6.48E-05	1.76E-05	2.73E-05	0.33	0.55	9.54	666.86
4	Narnaul	36.77	0.92	4.19E-05	6.48E-05	3.85E-05	5.95E-05	0.72	1.21	20.84	1456.32
5	Tehla	38.50	0.96	4.19E-05	6.48E-05	4.03E-05	6.23E-05	0.76	1.26	21.82	1524.66
6	Mukundpur	27.61	0.69	4.19E-05	6.48E-05	2.89E-05	4.47E-05	0.54	0.91	15.65	1093.53
7	Salarpur Ki Dhani	18.48	0.46	4.19E-05	6.48E-05	1.94E-05	2.99E-05	0.36	0.61	10.47	731.73
8	Bhungarkha	27.14	0.68	4.19E-05	6.48E-05	2.84E-05	4.39E-05	0.53	0.89	15.38	1074.66
9	Nangal Chaudhary	10.94	0.27	4.19E-05	6.48E-05	1.15E-05	1.77E-05	0.22	0.36	6.20	433.21
10	Asrawas	0.86	0.02	4.19E-05	6.48E-05	8.97E-07	1.39E-06	0.02	0.03	0.49	33.91
11	Masnuta	0.95	0.02	4.19E-05	6.48E-05	9.93E-07	1.54E-06	0.02	0.03	0.54	37.56
12	Bail Ki Dhani	2.68	0.07	4.19E-05	6.48E-05	2.80E-06	4.34E-06	0.05	0.09	1.52	106.04
13	Dholera	57.53	1.44	4.19E-05	6.48E-05	6.03E-05	9.32E-05	1.13	1.89	32.60	2278.56
14	Dhanota	18.50	0.46	4.19E-05	6.48E-05	1.94E-05	3.00E-05	0.36	0.61	10.48	732.48
15	Maroli	9.31	0.23	4.19E-05	6.48E-05	9.75E-06	1.51E-05	0.18	0.31	5.28	368.77
16	Kultajpur	56.98	1.42	4.19E-05	6.48E-05	5.97E-05	9.23E-05	1.12	1.87	32.29	2256.47
17	Dhosi	0.56	0.01	4.19E-05	6.48E-05	5.89E-07	9.11E-07	0.01	0.02	0.32	22.29
18	Mohhomadpur	5.60	0.14	4.19E-05	6.48E-05	5.86E-06	9.07E-06	0.11	0.18	3.17	221.74
19	Rampura	8.95	0.22	4.19E-05	6.48E-05	9.38E-06	1.45E-05	0.18	0.29	5.07	354.59
20	Nangal Siroi	24.49	0.61	4.19E-05	6.48E-05	2.57E-05	3.97E-05	0.48	0.80	13.88	969.98
21	Bewal	10.54	0.26	4.19E-05	6.48E-05	1.10E-05	1.71E-05	0.21	0.35	5.97	417.43
22	Bhojawas	8.60	0.22	4.19E-05	6.48E-05	9.01E-06	1.39E-05	0.17	0.28	4.87	340.71
23	Buchawas	7.91	0.20	4.19E-05	6.48E-05	8.29E-06	1.28E-05	0.16	0.26	4.48	313.35
24	Malra	35.35	0.88	4.19E-05	6.48E-05	3.70E-05	5.72E-05	0.70	1.16	20.03	1399.96
25	Mahendergarh	17.02	0.43	4.19E-05	6.48E-05	1.78E-05	2.76E-05	0.34	0.56	9.65	674.18
26	Dalanwas	3.21	0.08	4.19E-05	6.48E-05	3.36E-06	5.19E-06	0.06	0.11	1.82	126.96
27	Satnali	7.87	0.20	4.19E-05	6.48E-05	8.24E-06	1.27E-05	0.16	0.26	4.46	311.55
28	Bassai	52.95	1.32	4.19E-05	6.48E-05	5.55E-05	8.57E-05	1.04	1.74	30.01	2097.05
29	Baghot	9.18	0.23	4.19E-05	6.48E-05	9.61E-06	1.49E-05	0.18	0.30	5.20	363.37
30	Kanina	11.51	0.29	4.19E-05	6.48E-05	1.21E-05	1.86E-05	0.23	0.38	6.52	455.90

Table 2: Statistical Parameters of Obtained Data

Statistical Parameter	Conc. Of U (µg/L)	Ac (Bq/L)	ECR (Mortality)	ECR (Morbidity)	LADD (µg/Kg/Day)	HQ	D_E (µSv/Year)	pH	TDS (mg/L)	Cumulative Dose (µSv)
Range	0.56 - 57.53	0.01 - 1.44	5.89E-07 - 6.03E-05	9.11E-07 - 9.32E-05	0.01 - 1.13	0.02 - 1.89	0.32 – 32.60	7.16 - 8.15	90 - 3480	22.29 – 2278.56
Mean	18.87	0.47	1.98E-05	3.06E-05	0.37	0.62	10.69	7.69	1072	747.25
Median	12.56	0.31	1.32E-05	2.03E-05	0.25	0.41	7.12	7.75	888	497.53

$$D_E = Ac \times F \times I_{annual} \quad (5)$$

Where D_E in equation (5) is the annual effective dose (µSv/Year), Ac is the activity conc. (Bq/L), F is the effective dose per unit intake (µSv/Year/Bq/L) which is

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taken to be 4.5 x 10^{-8} and I_{annual} is the annual ingestion which was taken to be 503.7 L (1.38 x 365).

Annual Effective Dose was found to be varying in the range from 0.32 – 32.60 µSv/Year with the mean value of 10.69 µSv/Year. which is well below the recommended limit of 0.1 mSv [4].

Annual effective dose, when calculated for the whole of the lifetime is said to be the cumulative dose. It was also calculated and found to be varying in the range from 22.29 – 2278.56 µSv with the mean value of 747.25 µSv.

pH of the samples was found to be varying from 7.16 – 8.15 with the mean value of 7.69 which is well within the acceptable limit of 6.5 – 8.5 [14]. Total Dissolved Solids (TDS) of the samples was found to be varying in the range 90 – 3480 mg/L with the mean value of 1072.73 mg/L respectively. Out of 30 analyzed samples, TDS of 23 (76%) samples was found to be higher than the recommended limit of 500 mg/L by BIS [14]. TDS of 13 (43%) samples was found to be higher than 1000 mg/L which is the recommended TDS limit by WHO [4]. Statistical parameters of the obtained data are tabulated in table 2.

As it was stated earlier also that uranium is a ubiquitous element. A good amount of literature is present for its occurrence in groundwater worldwide. Variation of Uranium conc. in drinking water samples from different cities of India and from some other countries are tabulated in table 3 and table 4 respectively.

Table 3: U Conc. In Drinking Water Samples From Different Cities Of India

Sr. No.	Cities	Basic Source	U Conc. (µg/l)	References
1	Himachal Pradesh	Groundwater	0.56 – 10.11	[15]
2	Shri Ganganagar (Rajasthan)	Groundwater	2.5 - 171	[16]
3	Churu (Rajasthan)	Groundwater	13 - 95	[16]
4	Sikar (Rajasthan)	GroundWater	3 – 136	[16]
5	Khalilabad, Gorakhpur, Maharajganj, Kushinagar (Uttar Pradesh)	Bore well, River water Tap water, open well	0.02 – 64.00	[17]
6	Fatehabad (Haryana)	Groundwater	0.3 – 48	[18]
7	Western Haryana	Groundwater	6.37 – 43.31	[19]
8	Mansa (Punjab)	Groundwater	5.90 – 645.22	[20]
9	Bathinda (Punjab)	Groundwater	7.0 – 323.94	[20]
10	Amritsar (Punjab)	Groundwater	0.87 – 42.51	[20]
11	Hoshiarpur (punjab)	Groundwater	0.48 – 25.19	[20]
12	**Present Study (Mahendragarh District)**	Groundwater	0.56 – 57.53	

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Table 4: U Conc. In Drinking Water Samples From Different Countries

Sr. No.	Country	Basic Source	U Conc. (µg/L)	References
1	Amazonas (Brazil)	Groundwater	0.01 – 1.36	[21]
2	Southwestern Sinai (Egypt)	Groundwater	328 - 560	[22]
3	Northern Greece	Groundwater	0.01 – 10	[23]
4	Russia	Groundwater	>477	[24]
5	Ulaanbaatar (Mongolia)	Groundwater	<0.01 – 57	[25]
6	Switzerland	Groundwater	0.05 – 92.02	[26]

4 Conclusions

The present study reveals that the mean radiological, as well as the chemical toxicity risks in the area is negligible and well below the permissible limit. Out of 30 water samples, 24 samples i.e. 80% of the samples were below 30 µg/L. Only 6 samples out of them i.e. 20% of the samples were having the uranium conc. greater than 30 µg/L. Hazard quotient (HQ) was calculated for the samples analyzed and it was found to be varying from 0.02 – 1.89 with the mean value of 0.62. For these 6 samples it was found to be greater than 1 which shows that the water from these sources was unfit for drinking purposes.

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