RJC

RASĀYAN J. Chem.
Vol. 15 | No. 1 |343-349| January - March | 2022
ISSN: 0974-1496 | e-ISSN: 0976-0083 | CODEN: RJCABP
http://www.rasayanjournal.com
http://www.rasayanjournal.co.in

SPATIAL DISTRIBUTION OF URANIUM IN GROUNDWATER AND ITS HEALTH RISK ASSESSMENT IN HARYANA, INDIA

Naresh Tanwer, Pradeep Khyalia, Meena Deswal, J.S. Laura and Babita Khosla ✉

Department of Environmental Science, MDU, Rohtak-124001, (Haryana), India
✉Corresponding Author: babitakhosla@gmail.com

ABSTRACT

Uranium contamination is reported in the groundwater of Haryana but the data of its occurrence is utterly scattered. In the present study, the data related to the distribution of uranium in Haryana was compiled and a contour map was generated. The map showed average uranium concentration in groundwater of Fatehabad (22.3 ppb), Jind (16.76 ppb), Rohtak (15.49 ppb), Mahendargarh (18.87 ppb), Gurgaon & Sohna (22.09 ppb), Rewari (14.4 ppb) below the prescribed limit of WHO while in Hisar (33.9 ppb), Sonipat (57.43 ppb), Panipat (49.42 ppb), Sirsa (49 ppb), Jhajjar (53.01 ppb) and Bhiwani (30.15 ppb) districts higher than the standard limit of 30 ppb of WHO, but below the standard limit of 60 ppb issued by AERB. The AED (Annual Effective Dose) in all districts was found to be less than 100 µSv/y; ECR (Excess Cancer Risk) was found to be less in all districts except Hisar, Panipat, Sonipat, Sirsa, Fatehabad, Sohna, Gurugram, Rewari, Jhajjar and Bhiwani districts, where ECR was higher than the prescribed limit of 1.67×10^{-4} as per AERB.

Keywords: Uranium, Groundwater, Risk Assessment, Health Impact.

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INTRODUCTION

Uranium is a lithophilic, potential toxic heavy element ubiquitously present in groundwater and exists in forms of oxide or complex salts such as uraninite, pitchblende, davidite and carnotite.[1-2] The concentration of uranium in different environmental matrix vary vastly as in seawater is about 3 ppb and in the earth crust is about 2.76 mg/kg which is far greater than gold, silver and mercury.[3]

Uranium has been considered nephrotoxic and its toxicity depends on solubility, routes of entry, exposure or contact time and routes of eliminations.[4] A large value of uranium in groundwater can cause several health problems, if the human body gets exposure to the uranium of 0.1 mg/kg of body weight, it can lead to serious health hazards to lungs and kidneys.[5-8] Mostly uranium enters human beings via drinking water (85%) and groundwater is taken as the primary source of drinking for humans because of its ease of availability and quality.[9-10] Groundwater is used throughout the world and is considered a major source of irrigation, domestic and industrial supply in semi-arid regions.[5] It was reported that 85% of drinking need and 62% of agricultural requirement is met out from groundwater.[11-12] The highest concentration of uranium in groundwater has been reported in Punjab and Haryana as compared to other states of north India.[6,13]

The content of uranium in the groundwater of Haryana varies up to a great extent and the concentration fluctuates even seasonally.[14] The possibility behind such behavior is their agriculture dominant economy and these two states have utilized a very large quantity of fertilizer for farming. Phosphate fertilizer could be one of the reasons for the enhancement of uranium in groundwater and the use of natural phosphate deposits, bearing uranium 150 mg/kg, can contaminate groundwater.[15-16]

Many people were dying every year exclusively because of cancer in northern states of India and according to India Today Report, 2018, a total 12,091 deaths occurred in Haryana state exclusively due to cancer of different types.[6,17] The data of uranium distribution in the groundwater of Haryana is quite scattered and meager.

In the present study, the authors tried to present an overview of uranium distribution in the groundwater of Haryana and its consequential health impacts.

http://dx.doi.org/10.31788/RJC.2022.1516608

EXPERIMENTAL

Material and Methods

This study was performed to understand the scenario of uranium present in the groundwater of Haryana. The data was collected from Google Scholar, Research Gate, Springer, Elsevier, Semantic Scholar, Scopus etc. and analyzed thoroughly to present uranium occurrence in the groundwater of Haryana. The results reported in investigations were tabulated and presented through maps. The value of uranium concentration was also compared with the recommended limit of uranium given by WHO and AERB (Atomic Energy Regulatory Board) of India, 2004 to understand its consequent health issues. The health risk (radiological as well as chemical) due to consumption of uranium-contaminated water were calculated in form of Annual Effective Dose (AED), Excess Cancer Risk (ECR), Lifetime Average Daily Dose (LADD) and Hazard Quotient (HQ).

Radiological Risk

Risk due to the radioactive nature of uranium was calculated in form of AED through the eqn.-1:

$$\text{AED } (\mu Sv/y) = Ua \times DIW \times 365.25 \times DCF \quad (1)$$

Where Ua, uranium activity concentration (Bq/L), 1 ppb = 0.02528 Bq/L, Daily Intake of Water (DIW) which is 4.05 litre per day.[13] Dose conversion factor (DCF) which is taken as 4.5×10^{-8} Sv/Bq.[18]

Carcinogenic Risk

Carcinogenic risk due to uranium intake was calculated in form of ECR through eqn.-2:

$$\text{ECR} = Ua \times R \quad (2)$$

$$\text{Where R (Risk Factor)} = r \times IR \times EP \quad (3)$$

Where Ua is uranium activity concentration and R is a risk factor (Bq/L). In eqn.-3, r is the conversion factor which has a constant value of 1.9×10^{-9}/ Bq, IR (ingestion rate), taken as 4.05 liter per day and EP (Exposure Frequency) is 23740 days including leap years days (65 years).[19]

Chemical Risk

As a potent chemical toxic heavy element, it has a significant impact on the liver, kidney and bones which was calculated in form of LADD using the eqn.-4:[20]

$$\text{LADD } (\mu g/kg/day) = \frac{U \times DIW \times EF \times AE}{LE \times AW} \quad (4)$$

Where U is uranium concentration in water (ppb), DIW is a daily intake of water (4.05 L per day), EF is Exposure Frequency (350 days per year), AE is average exposure duration (65 years), LE is Life Expectancy in days (25201 days, 69 years including leap years) as per latest record of world bank data.[21-22] AW is the average weight of the body (53 kg).[23]

Hazard Quotient (HQ)

HQ was calculated to know the chemical effects of uranium-contaminated water on humans. HQ value is greater than one indicated significant harmful impact of uranium on targeted organs. It was calculated by using eqn.-5:

$$\text{HQ} = \frac{LADD}{RD} \quad (5)$$

Where RD= Reference Dose (4.53 µg/kg/day)[24]

For the generation of the contour map, geo-referencing was done using ArcGIS 9.3 software. The projection system for geo-referencing and digitization was UTM (Universal Transverse Mercator) and the standard system model was WGS84. A contour map was generated from the minimum and maximum value of uranium (ppb) given in the literature.

RESULTS AND DISCUSSION

The data of uranium concentration in the groundwater of Bhiwani revealed that 15% of groundwater samples of Bhiwani district was above the safe limit of 30 ppb as given by WHO[21] and 33% of samples exceeding the safe limit of 60 ppb as per AERB.[24-25] In Hisar city, only 16% of groundwater samples were safe for drinking as per the limit of WHO.[26] In another study of Haryana, it was reported that 16.67%

groundwater samples of Jind and 18.18% groundwater samples of Rohtak were found to be crossing the prescribed limit of 30 ppb of uranium given by WHO but all the samples were found to lie below the permissible limit of 60 ppb prescribed by AERB.[4] In Fatehabad district, the results reported that 11% of groundwater samples were higher in uranium content than the prescribed limit of AERB and 22% of samples were crossing WHO limit of uranium content in groundwater.[27-29] In Sonipat and Panipat district 66.67% and 82.14% groundwater samples respectively were observed to be crossing the threshold limit of WHO.[30] Further, 33.33% groundwater samples of Sonipat and 46.43% groundwater samples of Panipat were found to be surpassing even permissible limit of AERB (60 ppb). A study conducted in Sirsa which is the largest district of Haryana, revealed that uranium content in 44.44% of groundwater samples was exceeding the permissible limit of WHO (30 ppb) and even uranium in 22.22% of groundwater samples was found to be crossing the threshold limit of AERB (60ppb).[31]

The uranium in 20% groundwater samples of Mahendargarh district was higher than the prescribed limit of WHO (30 ppb), however, the samples were found to be within the AERB limit (60ppb).[32] According to the latest study of Sohna fault regions of Haryana, 11.6% and 18% groundwater samples were exceeding the threshold limit of uranium as per AERB (60 ppb) and WHO (30 ppb) respectively.[6] The concentration of uranium in Haryana ranges from 0.1 - 290 ppb as reported by different researchers. The ranges of uranium concentration in groundwater and technique used in previous studies conducted in Haryana are presented below in Table-1.

Table-1: Presence of Uranium in Groundwater of Haryana

Reference	Location	Types of Samples	Techniques Used	Uranium Conc. (ppb)
26	Hisar	Groundwater	Laser Fluorimetry	5.3-113.5
4	Jind	Groundwater	LED Fluorimeter	7.31-34.5
4	Rohtak	Groundwater	LED Fluorimeter	6.97-37.84
29	Fatehabad	Groundwater	LED Fluorimeter	1.1-113.0
30	Sonipat	Groundwater	LED Fluorimeter	9.1-155.1
30	Panipat	Groundwater	LED Fluorimeter	14.9-123.3
31	Sirsa	Groundwater	LED Fluorimeter	0.19-290
32	Mahendargarh	Groundwater	LED Fluorimeter	0.56-57.53
6	Gurugram and Sohna	Groundwater	LED Fluorimeter	0.1-223.16
Unpublished data	Rewari	Groundwater	LED Fluorimeter	0.1-61.5
Unpublished data	Jhajjar	Groundwater	LED Fluorimeter	1.73-179.64
Unpublished data	Bhiwani	Groundwater	LED Fluorimeter	1.44-226.71

The average uranium concentration in Fatehabad (22.3 ppb), Jind (16.76 ppb), Rohtak (15.49 ppb), Mahendargarh (18.87 ppb), Gurgaon & Sohna (22.09 ppb) and Rewari (14.4 ppb) were found to be below the prescribed limit of WHO as highlighted in brown color and groundwater of Hisar (33.9 ppb), Sonipat (49.42 ppb), Panipat (57.43 ppb), Sirsa (49 ppb), Jhajjar (53.01 ppb) and Bhiwani (30.15 ppb) districts were found to be higher than the standard limit of 30 ppb of WHO, highlighted through blue color in the contour map (Figure 1). The concentration of uranium varies widely and depends on various factors such as rock types, oxidation state and chemistry of groundwater that leads the formation of soluble $UO_2(CO_3)$ complexes.[33] Along with natural factors, the anthropogenic activities further enhance the process of uranium contamination in groundwater such as application of phosphate fertilizers and thermal power generation are some of them. The possibility of high uranium in these districts may be due to the geological composition, which is also influenced by Aravali hills, present in the proximity of the southwestern region of Haryana. The major parts of these districts such as Panipat, Sonipat, Sirsa, Fatehabad, Hisar and Jhajjar have older alluvium of quaternary periods. These alluviums contain a polycyclic sequence of sand, silt and brown color layered clay with Kakar, deposited by the sediments, which were transported from adjoining areas such as Siwaliks, granites and other metamorphic rocks.[34] In many studies, researchers have reported the high uranium concentration in granitic terrains. Therefore, the high uranium content in these areas may be due to the leaching of uranium in groundwater from the deposited granitic and Siwaliks sediments.[35]

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As per the studies, exceptionally very high uranium concentrations of 80,000 ppb and 14,870 ppb were observed in sedimentary and granitic formations.[36] The districts such as Bhiwani, Rewari and Gurugram majorly have Aeolian deposits of Holocene period comprising loose brown fine sand and minor rocks of Delhi Supergroup of Proterozoic Eon comprising Phylite, Amphibole Phylite, Slates, Limestone, Quartzite, Feldspathic Grity Quartzite and schist of Ajabgarh Group and Alwar Group. The Proterozoic deposits bear the 75% of the uranium resources of India.[37]

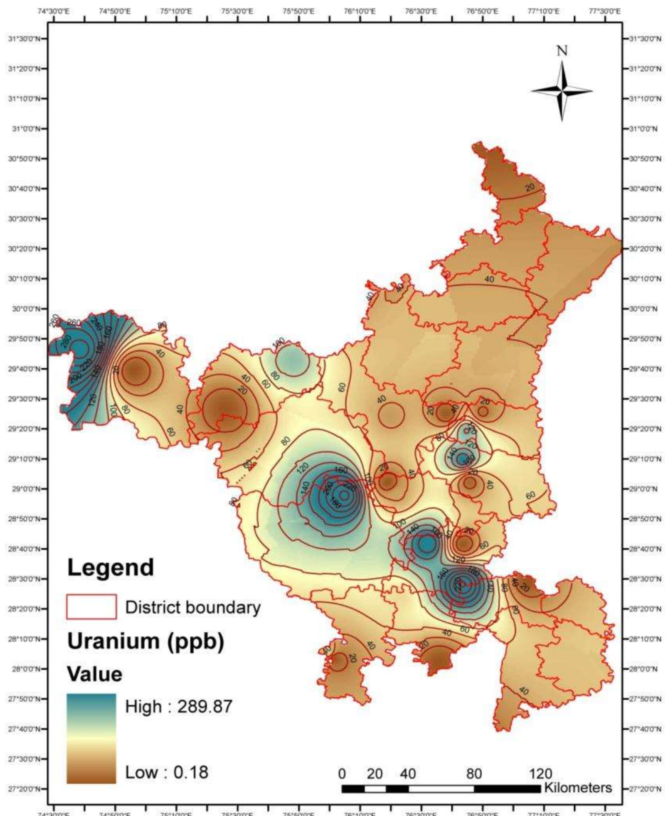

Fig.-1: Contour Map of Uranium Distribution in Groundwater of Haryana

Atomic Minerals Directorate (AMD) had conducted an intensive exploration of minerals that helped in uranium identification at various places of Delhi Supergroup of Proterozoic Eon and reported that Ajabgarh and Alwar Groups are intruded with Granite, Pegmatite and Quartz veins.[38-41] The intrusion of rocks in these regions could be the source of high uranium in groundwater.[42] Further, the groundwater of these regions is having a high concentration of carbonate and bicarbonate ion species resulting in the formation of UO_2CO_3. $UO_2\ (CO_3)_2^{2-}$ and $UO_2\ (CO_3)_3^{4-}$ which are soluble in groundwater.[43-44] Out of all carbonate complexes, $UO_2\ (CO_3)_2^{2-}$ is the most abundant one in the pH range of 6.5 to 8.

The evaluation of radiological risk (AED, ECR) as well as chemical risk (LADD, HQ) of minimum, maximum and mean value of uranium available from literature was calculated in groundwater of Haryana district and shown in Table-2.

Table-2: Table showing Uranium Activity, Excess Cancer Risk, Lifetime Average Daily Dose and Hazard Quotient in different districts of Haryana.

District		Uranium (ppb)	Uranium Activity (Bq/L)	AED (µSv/Y)	ECR	LADD (µg/Kg/Day)	HQ
Hisar	Min	5.3	0.134	8.92	1.53E-05	0.37	0.08
	Max	113.5	2.869	191.00	3.28E-04	7.83	1.73
	Mean	33.9	0.857	57.05	9.81E-05	2.34	0.52
Jind	Min	2.65	0.067	4.46	7.66E-06	0.18	0.04

	Max	34.05	0.861	57.30	9.85E-05	2.35	0.52
	Mean	16.76	0.424	28.20	4.85E-05	1.16	0.26
Rohtak	Min	1.07	0.027	1.80	3.09E-06	0.07	0.02
	Max	37.84	0.957	63.68	1.09E-04	2.61	0.58
	Mean	15.49	0.392	26.07	4.48E-05	1.07	0.24
Fatehabad	Min	1.1	0.028	1.85	3.18E-06	0.08	0.02
	Max	113	2.857	190.16	3.27E-04	7.80	1.72
	Mean	22.3	0.564	37.53	6.45E-05	1.54	0.34
Panipat	Min	9.1	0.230	15.31	2.63E-05	0.63	0.14
	Max	155.1	3.921	261.00	4.49E-04	10.70	2.36
	Mean	49.42	1.249	83.16	1.43E-04	3.41	0.75
Sonipat	Min	14.9	0.377	25.07	4.31E-05	1.03	0.23
	Max	123.3	3.117	207.49	3.57E-04	8.51	1.88
	Mean	57.43	1.452	96.64	1.66E-04	3.96	0.87
Sirsa	Min	0.19	0.005	0.32	5.50E-07	0.01	0.00
	Max	290	7.331	488.01	8.39E-04	20.01	4.42
	Mean	49	1.239	82.46	1.42E-04	3.38	0.75
Mahendar garh	Min	0.56	0.014	0.94	1.62E-06	0.04	0.01
	Max	57.53	1.454	96.81	1.66E-04	3.97	0.88
	Mean	18.87	0.477	31.75	5.46E-05	1.30	0.29
Fatehabad	Min	1.1	0.028	1.85	3.18E-06	0.08	0.02
	Max	113	2.857	190.16	3.27E-04	7.80	1.72
	Mean	22.3	0.564	37.53	6.45E-05	1.54	0.34
Gurugram and Sohna	Min	0.1	0.003	0.17	2.89E-07	0.01	0.00
	Max	223.16	5.641	375.54	6.45E-04	15.39	3.40
	Mean	22.09	0.558	37.17	6.39E-05	1.52	0.34
Rewari	Min	0.1	0.003	0.17	2.89E-07	0.01	0.00
	Max	61.5	1.555	103.49	1.78E-04	4.24	0.94
	Mean	14.4	0.364	24.23	4.17E-05	0.99	0.22
Jhajjar	Min	1.73	0.044	2.91	5.00E-06	0.12	0.03
	Max	179.64	4.541	302.30	5.20E-04	12.39	2.74
	Mean	53.01	1.340	89.21	1.53E-04	3.66	0.81
Bhiwani	Min	1.44	0.036	2.42	4.17E-06	0.10	0.02
	Max	226.17	5.718	380.60	6.54E-04	15.60	3.44
	Mean	30.15	0.762	50.74	8.72E-05	2.08	0.46

CONCLUSION

The mean values of uranium in groundwater of Fatehabad, Jind, Rohtak, Mahendargarh, Gurgaon & Sohna and Rewari were observed to be less than the prescribed limit of WHO while in Hisar, Sonipat, Panipat, Sirsa, Jhajjar and Bhiwani districts were found to have higher than the standard limit as per WHO but not greater than the permissible limit of uranium as per AERB. The values of AED and ECR due to the average value of uranium in each district were below the prescribed limit of 100 µSv/y as per WHO and 1.67×10^{-4} as per AERB respectively. LADD due to the mean value of uranium in each district, except Rewari, was higher than the threshold limit of 1.0 µg/kg/day as per WHO, 2011; but less than 4.53 µg/kg/day as per AERB, 2004. The study concluded that the carcinogenic effect due to consumption of groundwater may be negligible but the non-carcinogenic effect due to chemical toxicity may be significant.

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[RJC-6608/2021]