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Assessment of uranium contamination in the groundwater and associated health risk inNarnaul block of Mahendergarh district of SW Haryana, India

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Title Page

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Abstract

This study evaluates uranium contamination and associated chemical and radiological health risks in groundwater from Narnaul Block, Mahendragarh district, Haryana, India. Analysis of 28 groundwater samples revealed uranium concentrations exceeding the World Health Organization guideline of 30 µg/L in 28.57% of samples, with a maximum concentration of 237.84 µg/L at Jadupur. The integrated assessment of hazard quotient and excess lifetime cancer risk provides insights into the potential health implications of uranium exposure through drinking water. The findings highlight localized contamination hotspots and emphasize the need for targeted groundwater monitoring and risk mitigation in uranium-bearing geological terrains.

Keywords: Uranium Contamination, Groundwater quality, Health Risk, Excess Life Cancer Risk, Hazard Quotient, Narnaul, Mahendergarh

1. Introduction:

Water is fundamental for the basic human needs like food, clothes and shelter. For the full potential utilization of this natural resource for various needs viz. drinking, domestic, irrigation and industrial, the quality is as important as the quantity. The quantity of freshwater on Earth is limited and Groundwater contributes the major portion accounting for roughly 99% of all liquid freshwater along with other surface sources like rivers, lakes, and atmospheric moisture [1]. Throughout the world, more than 50% population is utilizing groundwater to meet their drinking water needs [2-4]. More dependency on the groundwater for drinking purpose is well evident in the arid and semi-arid area. However, groundwater is not free from contaminants as quality of groundwater is controlled by many factors including natural geogenic as well as human induced factors such as

excessive extraction; for various purposes viz. domestic, irrigation, industrial; that induce contaminations in the aquifer [5, 6]. The geogenic contaminants like uranium (U), arsenic (As), fluoride (F), and selenium (Se) are major concern as some studies shows that more than half of global aquifers are contaminated [7, 8]. The intake of drinking water having high concentration of above-mentioned elements may cause serious health issues like inappropriate functioning of essential organs of human body viz. liver, kidney, brain, heart, respiratory system, reproductive etc., which may cause behavioral abnormality., and some elements are even carcinogenic [9-13]. Therefore, the permissible limit of particular element, for specific use, is recommended by statutory bodies like World Health Organization (WHO), Bureau of Indian Standards (BIS), United States Environmental Protection Agency (USEPA). The variation from permissible limit may make it unsuitable for that particular use [14].

The uranium contamination in the groundwater above the permissible limit for drinking purpose recommended by WHO has been reported at local scale throughout the world. An extremely high concentration of U in groundwater, extracted from private well, has been detected in many countries viz. USA, Germany, Canada, Australia etc. [15-19]. South Asian countries including India, China, Bangladesh and Pakistan are also suffering with U contamination in their groundwater sources due to various geogenic as well as human induced factors [20, 21]. The various researchers from 2007 to 2021 tried to attempt the study of Uranium contamination in 23 Indian states including national capital Delhi and shows variation from 0 to 1442.9 ng/ml [22, 23]. According to report issued by Central Ground Water Board in 2020, 18 states/union territories (including Haryana) of India have uranium content more than permissible limit i.e. 30 µg/L by WHO.

uranium scattered in the crust of the Earth in various rock types such as basalt, granite, black shale coal etc. in variable amount with average concentration 2.8 µg/g [24, 25]. The main cause of uranium contamination in the groundwater is geogenic, controlled by factors like interaction with aquifer lithology, oxidizing environment, high alkalinity and total hardness [26, 27]. In agro prone area, high concentration of nitrate may cause the oxidation of uranium (IV) to uranium (VII) in subsurface water [28, 29]. Anthropogenic activities including industrial, agricultural, mining, defense & strategic (war related) practices, along with excessive extraction of groundwater enhance the concentration of uranium in groundwater at particular location [30]. Therefore, the U present in the crust of Earth is mobilized to human body through plants and dietary nutrients [31]. The presence of uranium found in the groundwater is serious concern arising from the various parts of the world as it may create many health issues in the population living there. Recent studies indicate that the presence of uranium in the drinking water may cause several serious health issues like chronic kidney disease, lung cancer along with other health problems [17].

To check the best suitability of available water for particular use and to adopt the appropriate technique of treatment, it is important to detect the presence of contamination in water. There are many techniques useful to the detect the uranium concentration in the groundwater with appropriate precession and accuracy like Raman spectroscopy, LED fluorimetry, inductively coupled plasma mass spectrometry (ICP-MS), high-resolution ICP-MS (HR-ICP-MS) and multi-collector ICP-MS (MC-ICP-MS) etc. [25]. As per World Health Organization (WHO) the recommended limit of uranium in the drinking

water is 30 µg/L [32]. Although, in Indian conditions the permissible limit has been extended upto 60 µg/L. [33]. To determine the health impacts of Uranium contaminated water, Excess Life Cancer Risk (ELCR) and Health Quotient (HQ) are useful parameters.

In the Haryana state, the uranium contamination in groundwater has been studied in Bhiwani, Hisar, Fatehabad, Sirsa, Gurugram districts and some parts of western Haryana and varies from 0.10 to 223 ng/ml [34-37]. The current study area i.e. Narnaul block of Mahendergarh district in SW Haryana, falls in the semi-arid region of the country where groundwater is the main source of drinking water.

Due to semi-arid conditions, along with hard rock aquifers and granitic terrain, in this region, makes it favorable for uranium contamination [38]. In this area, the previous researchers (39-41) attempted to cover the entire district with wide sample spacing. Therefore, block level assessment with relatively dense sampling and geospatial driven analysis is lacking. This kind of approach will help in decentralizing the planning & implementation strategies, for better treatment and management of drinking water, at block level to avoid the health issues arises from uranium contaminated groundwater in the study area.

The current study meets the above-mentioned research gap with special emphasis on following objectives: (a) Evaluation of uranium contamination in the groundwater of Narnaul block. (b) Spatial distribution of uranium in the groundwater of the study area. (c) Assessment of human health risk associated with uranium contamination.

2. Materials and methods

2.1. Area of study:

In India, blocks are important administrative units with sizable area and population. The current study area i.e. Narnaul block of Mahendergarh district in SW Haryana have total geographical area of 281.03 Km^2 [44] with 106540 souls. (Census, 2011). The Narnaul block extends between latitudes $27^0$47'N to $28^0$26'N and longitudes $75^0$56'E to $76^0$51'E with an average hight of 262 meters (859 feet) amsl having beautiful landscape controlled by the Aravalli hillocks and valley of Dohan and Krishnawati Rivers. Narnaul city, as the district headquarter of Mahendergarh district, is situated about 150 km SSW of Delhi. It can be approached from National capital Delhi via Gurgaon, Dharuhera and Rewari through NH-8. Narnaul is also directly connected with state capital, Chandigarh, through road network. The study area is also accessible through railway transportation as Narnaul railway station is falling on Delhi-Udaipur railway line passing via Ajmer junction in Rajasthan.

The research area falls in tropical steppe climate characterized by hot summer and cold winter, with average rainfall 500 mm. The Narnaul block is dominated by agriculture class of landuse/Landcover having Loamy sand, Sandy soil types, and main khrif crops are Bajra, Gawar, Cotton whereas rabi crops are Wheet, Oilseeds and Mustard [42-43]. Being semi-arid region and lacking the surface water bodies, the main source of irrigation and drinking water supply is groundwater. According to Atal Bhujal project

report-2023-24 of IRRIGATION & WATER RESOURCE DEPARTMENT [44] Haryana, Narnaul block is under over exploited category in year-2022, due to more extraction of groundwater than supply by recharge.

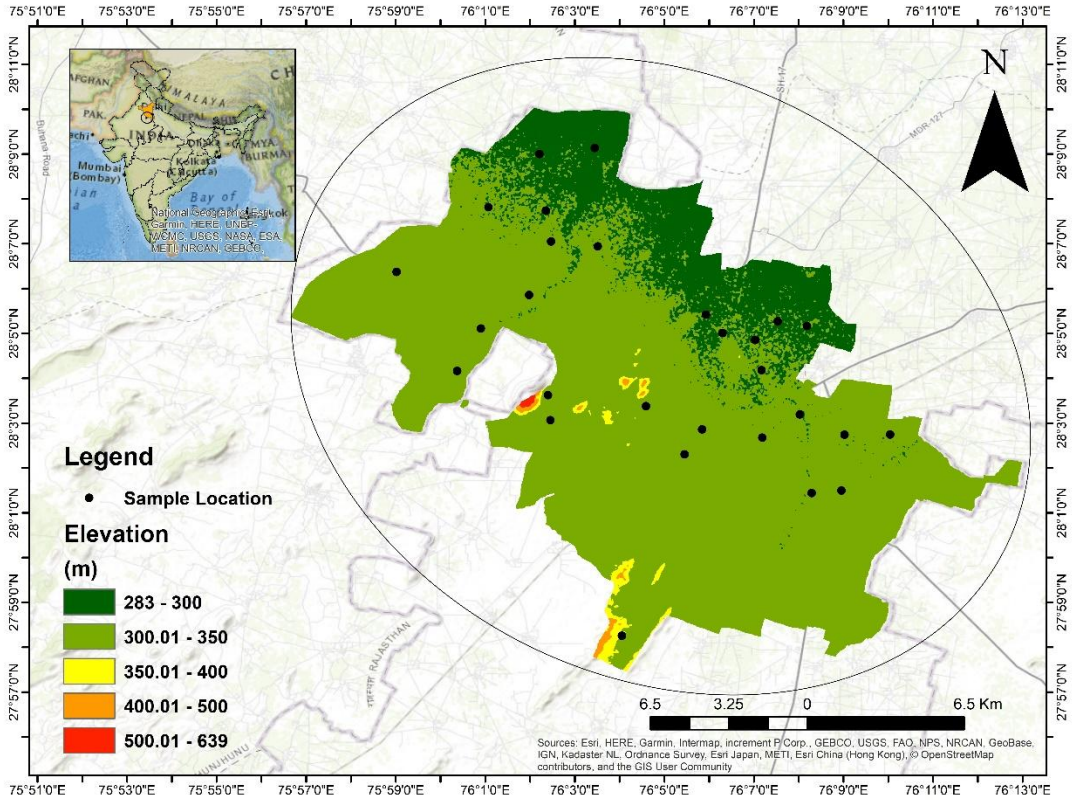


Fig. 1 Location map showing the geographical setting of the study area in the Narnaul blocks of Mahendragarh District, Haryana, India

2.2. Geological Setting & Hydrogeology:

Geological setting of the study area is characterized by mature topography predominantly alluvial plains dotted with isolated hillocks and crossed by parallel ridges of resistant rocks composed of quartzite, crystalline limestone, Calc-Silicate Rocks (CSR) and intrusive with strike parallel to regional trend i.e. NE-SW [45]. Stratigraphicaly, rock formations belong to Ajabgarh group of Delhi supergroup with quartzite, phyllite, mica-schist, limestone, granite, gneiss etc. and post delhi Igneous intrusions exposed in the form of inselbergs. The alluvial and soils are characterized by presence of sand, silt with kankar and weathered products of exposed rock types in the region. The aquifer system consists of course grained sand and fractured hard rock formations. The presence of granitic lithology and semi-arid conditions makes it vulnerable location for uranium contamination in the groundwater [46-48]. Dominant geological units in the study area are represented in the fig: 2.

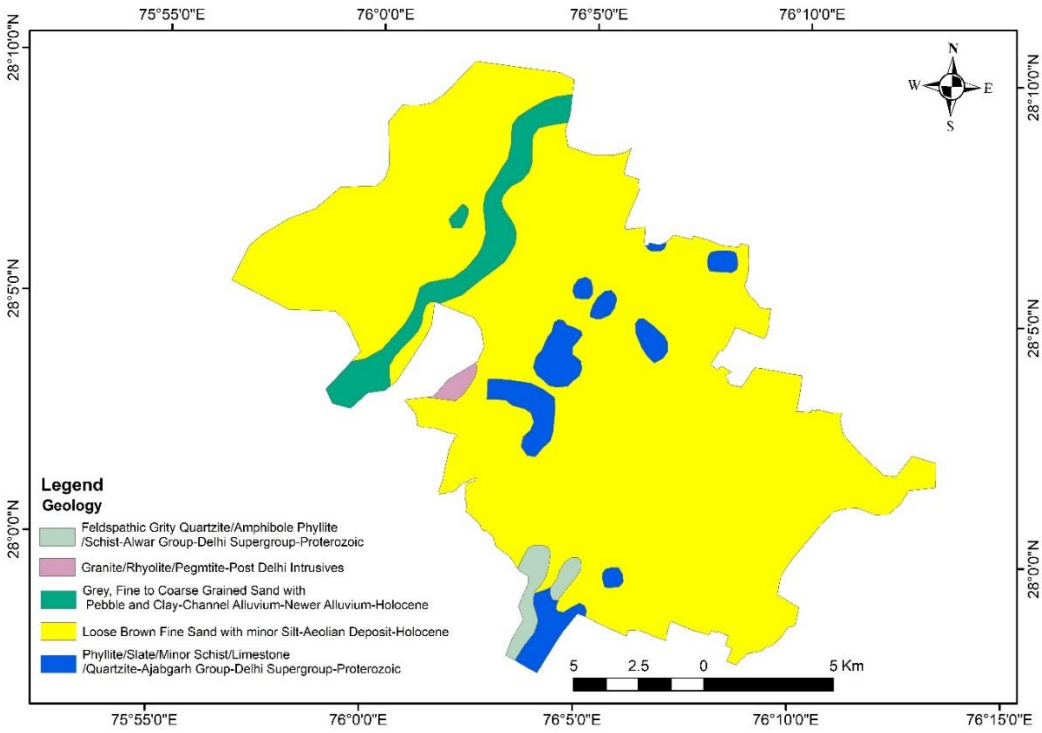


Fig. 2 Geological map of Narnaul block of Mahendergarh district (Modified after IRRIGATION & WATER RESOURCE DEPARTMENT, Haryana Atal Bhujal report 2023-24) [44]

2.3. Sample Collection and Analytical Procedures:

2.3.1 Collection of Samples:

Total 28 groundwater samples were collected from bore wells, tube wells and hand-pumps available in the study area and as confirmed by locals, the water of these sources is using for drinking and irrigation purpose. For sample collection, high density polyethylene (HDPE) bottles were selected and cleaned well with dilute liquid detergent and after that rinsed with distilled water before allowed to dry overnight. To confirm the availability of fresh sample of groundwater, the pump was allowed to flow for 5-10 minutes before collection of samples at each location. At some locations (village/town) more than one sample are collected to check the influence of multiple factors like mining activities, granitic exposure in the vicinity or size set of living population in village/town.

2.3.2 Treatment & Analysis:

During sample collection due care has been taken to avoid the contamination and treated immediately with nitric acid (1% v/v) to prevent the microbial growth during preservation. Before analysis the samples are filtered through 0.45 µm filter paper to remove the solid particles, if any. As the recommended value of Uranium in the drinking water is 30 µg/l so highly sophisticated technique like ICP-MS is appropriate for sample analysis [49].

ICP-MS analysis are carried out at Department of Geology, Panjab University, Chandigarh to determine the uranium concentration in the groundwater samples by Agilent 7850. The obtained values of uranium concentration is compared with permissible limit for drinking water set by i.e. 30 μgL−1 and 60 μgL−1 by the WHO & the Atomic Energy Regulatory Board (AERB), India respectively.

2.4. Health Risk Assessment

The presence of U in drinking water may affect the human health in two ways, as U is a radioactive element so generate radiological effects (radiological risks) as well as chemical nature of element may cause harmful effects (Chemical risk) radiological effects may cause cancer hence called carcinogenic effects whereas just chemical effects are known as non-carcinogenic effects so both kind of risks are considered and calculated.

2.4.1. Radiological Risk Assessment:

Radiological Risk (RR): It is the product of average daily dose (ADD), average life span (63.7 years) and slope factor (SF) so that

$$RR = ADD * 23250.5 * SF \quad (1)$$

According to HDR, 2009, the average daily intake of water= 4.05 L/day. The activities has been converted to pCi with the help of conversion factor (1 Bq= 27 pCi) given by USEPA in 1997 for slope factors of cancer by radionuclides and for Uranium, it is $6.2*10^{-11}$ Risk per pCi.

2.4.2. *Chemical Risk Assessment:*

The most commonly used parameter to calculate the non-carcinogenic impact on human body is the Hazard Quotient (HQ), which is the indicator of magnitude of damage arises from the uranium contaminated water consumption. The Hazard Quotient is the ratio of average daily dose (ADD) to reference dosage (RfD), calculated by USEPA (1989a, 1997) as 0.0003 mg/kg of human body/day.

$$\frac{HQ=ADD}{RfD} \quad (2)$$

2.4.3. *Spatial Variation Analysis:*

To understand the spatial distribution of uranium in the groundwater and consequently Radiological Risk factors as well as Chemical Risk factor i.e. HQ, the most common methods are kriging, spline and inverse distance weighted (IDW) (Gong et al., 2014). All these interpolation techniques have their own pros and cons. For more accuracy, simple to use and efficiency reasons, here we adopted IDW technique to determine the data of in-between sampling area. In this technique of interpolation, the effect of value at particular station decrease as the distance increase from that point. The spatial variation of Uranium content is shown in the fig-4.

3. Results and Discussion:

3.1. Uranium contamination in the Groundwater

The amount of uranium content in the groundwater samples collected from the study area is given in the table- 01. The spatial variation is represented in figure-4. The available uranium content is compared with the recommended value by World Health Organisation (WHO) for drinking water and expressed by figure-03. The value of uranium has been well exceeding the recommended value in 8 samples i.e. 28.57% with highest value 237.84 ppm at Jadupur village. The uranium content in the collected groundwater sample is found above the recommended value by Atomic Energy Regulatory Board (AERB), India i.e. 60 ppb at 3 locations. The sample collection location and Uranium concentration in each sample are presented in the table-1 and graphical presentation is given in Fig. 3.

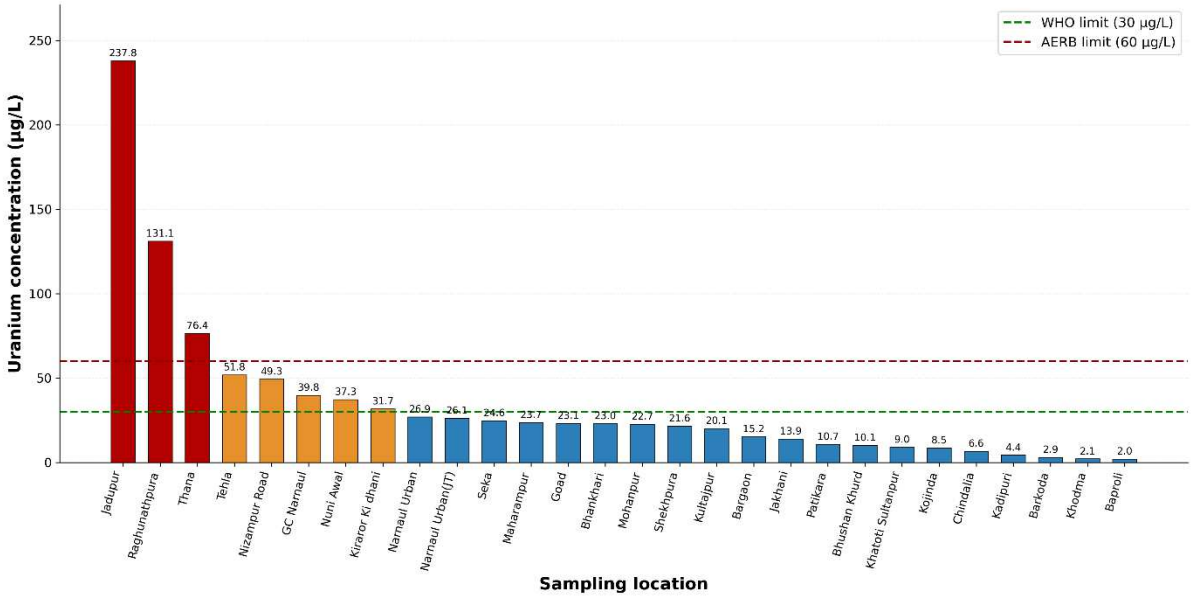


Fig. 3 Uranium concentrations in groundwater samples collected from different locations in the Narnaul block, compared with the WHO guideline value (30 µg/L) and the AERB drinking-water limit (60 µg/L)

Table 1 Uranium concentration in the groundwater samples collected from study area.

Sr. No.	Location	Sample ID	Coordinates	U (Ppb)
1	Bhankhari	BKH-W1	N28.085146, E76.015062	23.02
2	Jadupur	JDP-W1	N28.06942, E76.006265	237.84
3	Goad	GD-W1	N28.106221, E75.983713	23.10
4	Khatoti Sultanpur	KHS-W1	N28.097625, E76.033027	8.98
5	Narnaul Urban	NNL(U)-	N28.047672,	26.95

		W1	E76.097382	
6	Raghunathpura	RP-W1	N28.056301, E76.076511	131.08
7	Kultajpur	KJP-W1	N28.051074, E76.040935	20.10
8	GC Narnaul	NNL(U)-W2	N28.044606, E76.119831	39.82
9	Shekhpura	SKP-W1	N28.069673, E76.119508	21.55
10	Nuni Awal	NA-W1	N28.080946, E76.117175	37.26
11	Bargaon	BG-W1	N28.087813, E76.125548	15.17
12	Barkoda	BK-W1	N28.086092, E76.13643	2.91
13	Nizampur Road	NNL(U)-W3	N28.038397, E76.090934	49.32
14	Narnaul Urban(JT)	NNL(U)-W4	N28.083502, E76.105039	26.12
15	Kiraror Ki dhani	DKR-W1	N28.09032, E76.098823	31.73
16	Maharampur	MRP-W1	N28.115711, E76.058591	23.66
17	Khodma	KH-W1	N28.128995, E76.039267	2.13
18	Baproli	BPR-W1	N28.152292, E76.05746	2.05
19	Chindalia	CD-W1	N28.150003, E76.036834	6.56
20	Jakhani	JKH-W1	N28.117496, E76.041185	13.88
21	Mohanpur	MP-W1	N28.130261, E76.017884	22.71
22	Thana	TH-W1	N28.060413, E76.040003	76.44
23	Tehla	THL-W1	N27.945789, E76.047742	51.79
24	Kojinda	KJ-W1	N28.024021; E76.138192	8.51
25	Seka	SE-W1	N28.024968; E76.149249	24.65
26	Kadipuri	KDP-W1	N28.045681, E76.150453	4.36
27	Bhushan Khurd	BHK-W1	N28.045728, E76.167383	10.08
28	Patikara	PTK-W1	N28.053225, E76.133781	10.69

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256 *3.2. Drinking Water Quality Assessment:*

On the basis the uranium content in the analysed groundwater samples and the comparison with recommended values by WHO & AERB having standard values 30 ppb and 60 ppb respectively, 3 classes are identified as mentioned by National Institutes of Health and shown in the table: 2

Table 2 Classification of water quality based on Uranium content.

Sr. No.	Uranium content (ppb)	Class	Grade
1.	< 30	I	Safe / Desirable
2.	30-60	II	Elevated / Action Required
3.	>60	III	Unsafe / High-Risk Class

Out of total (28) collected samples, 20 (71.42 %) are falling under class-I for negligible chemical toxicity effects and radiological health risks for lifetime consumption whereas 5 (17.85%) are found under class-II representing chances of long-term chemical toxicity risks (such as potential kidney/nephritic damage). Remaining 3 (10.71%) samples collected from Thana, Raghunathpura and Jadupur village comes under the class-III shows requirement of immediate termination of use for drinking and cooking and adopt alternative water provisioning such as bottled water or verified municipal supplies or shift to water treatment techniques like reverse osmosis or ion exchange.

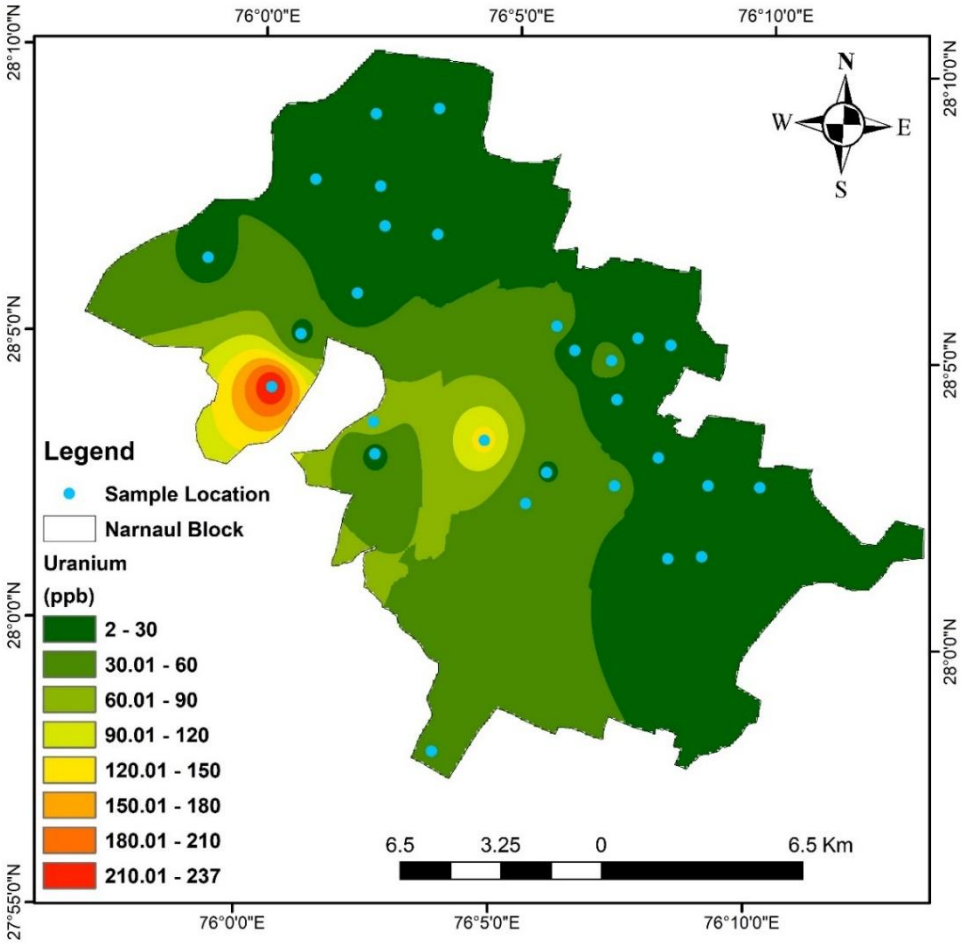


Fig. 4 Spatial variation of Uranium value in the Groundwater collected from the study area.

3.3. Health Risk implications:

Prolong intake of uranium contaminated water may cause many health-related issues including failure of renal, harmful effects on the growth of bones and even damage the DNA (Brugge et al., 2005). Adverse effects of uranium contamination are caused by its chemical toxicity as well as radiological nature. Although, chemical toxicity of uranium is main threat for human health (Zamora et al., 2009) but radiological effects also need due consideration.

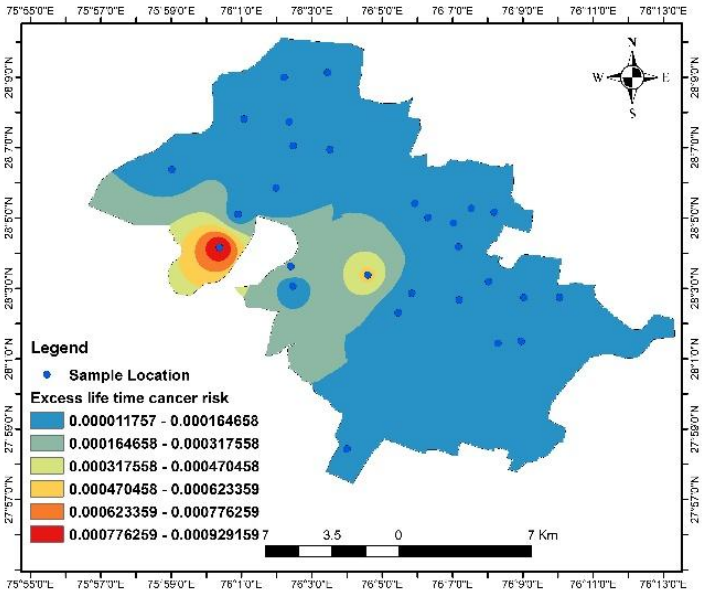
In the present investigations, the radiological risk is calculated in terms of Excess Life Time Cancer Risk (ELCR) whereas risk of chemical toxicity is expressed in terms of the Hazard Quotient (HQ), as per USEPA guidelines, for all 28 samples collected from the study area and presented in table-3. The values of ELCR vary from 8.02×10^{-6} at Baproli village to 9.30×10^{-4} at Jadupur village. When compared with USEPA's safe limits (1×10^{-6} to 1×10^{-4}), it was found that 10 sites (36%) have ELCR values beyond the acceptable limit. The HQ values range from 0.052 to 6.058. The HQ value above the 1.0 shows the risk of chemical toxicity due to Uranium contamination and exceeded the safe limit at 6 locations. Spatial variation of ELCR & HQ is displayed in the Figure-5 and Figure-6 respectively. There is linear relationship

between uranium content and derived excess life cancer risk (ELCR) and Hazard Quotient (HQ) as shown in figure-7.

Table 3 Values of excess life cancer risk (ELCR) and Hazard Quotient (HQ) calculated from U concentration as per USEPA guidelines.

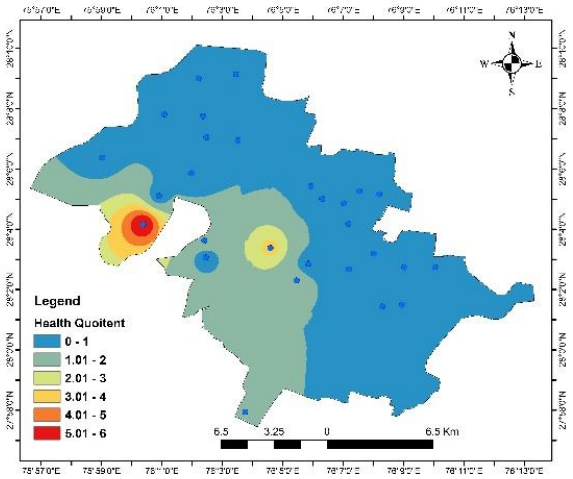
Sr. No.	Location	U Conc. (Ppb)	Total Lifetime Activity Intake (TLAI) in pCi	Excess life time cancer risk (ELCR)	ADD (Chemical) µg/kg/day	HQ (EPA)
1	Bhankhari	23.02	1.45×10^{6}	9.00×10^{-5}	1.759	0.586
2	Jadupur	237.84	1.50×10^{7}	9.30×10^{-4}	18.175	6.058
3	Goad	23.10	1.46×10^{6}	9.04×10^{-5}	1.765	0.588
4	Khatoti Sultanpur	8.98	5.67×10^{5}	3.51×10^{-5}	0.686	0.229
5	Narnaul Urban	26.95	1.70×10^{6}	1.05×10^{-4}	2.059	0.686
6	Raghunathpura	131.08	8.27×10^{6}	5.13×10^{-4}	10.016	3.339
7	Kultajpur	20.10	1.27×10^{6}	7.86×10^{-5}	1.536	0.512
8	GC Narnaul	39.82	2.51×10^{6}	1.56×10^{-4}	3.043	1.014
9	Shekhpura	21.55	1.36×10^{6}	8.43×10^{-5}	1.647	0.549
10	Nuni Awal	37.26	2.35×10^{6}	1.46×10^{-4}	2.847	0.949
11	Bargaon	15.17	9.57×10^{5}	5.93×10^{-5}	1.159	0.386
12	Barkoda	2.91	1.84×10^{5}	1.14×10^{-5}	0.222	0.074
13	Nizampur Road	49.32	3.11×10^{6}	1.93×10^{-4}	3.769	1.256
14	Narnaul Urban(JT)	26.12	1.65×10^{6}	1.02×10^{-4}	1.996	0.665
15	Kiraror Ki dhani	31.73	2.00×10^{6}	1.24×10^{-4}	2.425	0.808
16	Maharampur	23.66	1.49×10^{6}	9.25×10^{-5}	1.808	0.603
17	Khodma	2.13	1.34×10^{5}	8.33×10^{-6}	0.163	0.054
18	Baproli	2.05	1.29×10^{5}	8.02×10^{-6}	0.157	0.052
19	Chindalia	6.56	4.14×10^{5}	2.57×10^{-5}	0.501	0.167
20	Jakhani	13.88	8.76×10^{5}	5.43×10^{-5}	1.061	0.354
21	Mohanpur	22.71	1.43×10^{6}	8.88×10^{-5}	1.735	0.578
22	Thana	76.44	4.82×10^{6}	2.99×10^{-4}	5.841	1.947
23	Tehla	51.79	3.27×10^{6}	2.03×10^{-4}	3.958	1.319
24	Kojinda	8.51	5.37×10^{5}	3.33×10^{-5}	0.650	0.217

25	Seka	24.65	1.56×10^{6}	9.64×10^{-5}	1.884	0.628
26	Kadipuri	4.36	2.75×10^{5}	1.71×10^{-5}	0.333	0.111
27	Bhushan Khurd	10.08	6.36×10^{5}	3.94×10^{-5}	0.770	0.257
28	Patikara	10.69	6.74×10^{5}	4.18×10^{-5}	0.817	0.272



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297 **Fig. 5** Spatial variation of Excess Lifetime Cancer Risk (ELCR) in the Groundwater samples



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299 **Fig. 6** Spatial variation of Health Quotient (HQ) in the Groundwater samples.

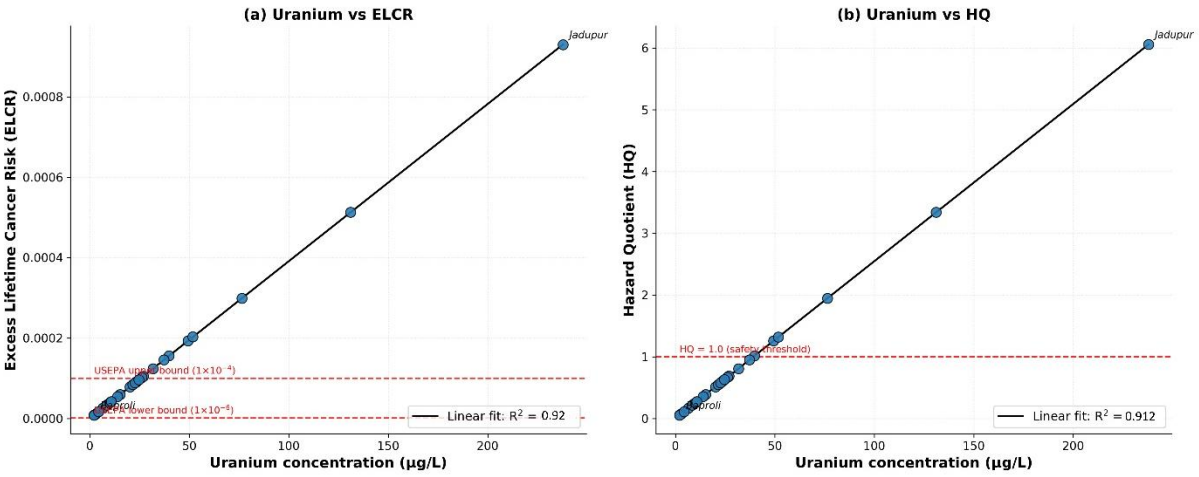


Fig. 7 Linear Relationship between groundwater uranium concentration and derived radiological (ECLR) and chemical (HQ) risk indices

4. Conclusion:

In the current study, a systematic evaluation of uranium contamination in the groundwater of Narnaul block of Mahendergarh district in SW Haryana has been carried out with special emphasis on the human health in terms of Excess life cancer risk and Hazard quotient for drinking purpose. Total 28 groundwater samples were collected from various sources such as borewells, tubewells, handpumps from the study area and analyzed with ICPMS technique at Department of Geology, Punjab University, Chandigarh to determine the uranium content. The uranium content varies from 2.05 ppb at Baproli village to 237.84 ppb at Jadupur village. The obtained values are compared with the reference permissible values (30 ppb and 60 ppb) recommended by WHO and AERB respectively for drinking water. The spatial variation modelling is done by geoinformatics technology.

As uranium contamination have chemical toxicity as well as radiological affects so the associated health risk are calculated in terms of Excess Life Cancer Risk (ELCR) and Hazard Quotient (HQ) as per guidelines of USEPA-2011 and presented in table-3. The values of ELCR varies from 8.02×10^{-6} at Baproli village to 9.30×10^{-4} at Jadupur village. When compared with USEPA's safe limits (1 × 10-6 to 1 × 10-4), it was found that 10 sites (36%) have ELCR values beyond the acceptable limit. The HQ values range from 0.052 to 6.058. The HQ value above the 1.0 shows the risk of chemical toxicity due to uranium contamination and exceeded the safe limit at 6 locations. Spatial variation of ELCR & HQ is displayed in the Figure-4 and Figure-5 respectively.

The results of this research will provide a baseline for further work on groundwater quality assessment particularly for uranium contamination in the semiarid regions and identification of sources for such contamination. This study will also help the district administration for block level action plan to ensure the availability of pure drinking water to everyone. The research will also function as awareness document regarding alarming chronical disease due to uranium like contamination and need of an appropriate treatment before indigestion of water particularly from groundwater sources.

5. Authorship contribution statement

Jaipal: Conceptualization, Writing – original draft, Methodology, Formal analysis; **Naresh Kumar:** Writing – review & editing, Supervision. **Baru Ram:** Software, Formal analysis, **Ankush Sagwal:** Sample collection, Investigation, Data curation.

6. Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. There is no conflict of interest in authors.

7. Data availability

All data generated or analysed during this study are included in this published article.

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