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Uranium in Groundwater: Distribution and Plausible Chemo-Radiological Health Risks Owing to the Long-term Consumption of Groundwater of Panchkula, Haryana, India

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ABSTRACT

A comprehensive investigation was engaged to determine the spatial distribution of Uranium (U) and the consequential chemical and radiological health risk associated due to the consumption of groundwater containing U, in Panchkula district. A well-accepted technique of fluorescence of U estimation in an aqueous medium was employed having a detection limit of 0.50 μgL^{-1}. The chemo-radiological health risk and water quality index was computed using a standard equation of concerned agencies to determine the suitability for human health. The concentration of U was observed to vary from 1.70 – 12.28 μgL^{-1} with the mean value of 5.89 μgL^{-1} The concentration of U was far below the standard prescribed limits as per World Health Organisation, Atomic Energy Regulatory Board, and United Nation Environmental Protection Agency. Except nitrate and total alkalinity in few samples, all water quality paramters were within the recommended limit of BIS. The annual effective dose (AED), excess cancer risk (ECR), and lifetime average daily dose (LADD) indicated no potential health issue due to the consumption of groundwater of studied locations. The correlation was computed between U and various macro-anions and cations present in water samples. U was observed to have a significant weak positive correlation with total dissolved solids (TDS), electrical conductivity (EC), and salinity.

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INTRODUCTION

In recent years, there has been a growing concern over the presence of U in groundwater. Several previous investigations all over the world investigated the status of U prevalence in groundwater (Banning & Benfer, 2017; Bjørklund et al., 2017; Coyte et al., 2018; Godoy et al., 2019; Ma et al., 2020; Winde et al., 2017). Various studies from India reported the groundwater contamination due to U and their possible natural as well as anthropogenic sources (Kale et al., 2020; Machiraju et al.,2020; Prasad et al., 2019;Richards et al., 2020; Sharma et al., 2019). According to a detailed study, conducted in India, more than 90% of groundwater is used for

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irrigation purposes that causing a sharp decline in the groundwater table in many parts of the country, specifically in north-western states, viz. Haryana, Punjab, and Rajasthan (Coyte et al., 2018; Rodell et al., 2009). The first report of the high content of U in the groundwater of India was observed in the southwest regions of Punjab state in 1995 (Singh et al., 1995). Later on, the study in the southwest region of Punjab reported that the groundwater of hard rock areas along with the alluvial plains also was contaminated with a high levels of U (Panghal et al., 2017; Pant et al., 2017; Rishi et al., 2017). As per the previous investigations in the last three decades, U toxicity is the major concern to the resources (Duggal et al., 2021; Kumar et al., 2021). Conversely, the U is the least common heaviest element present in the earth's crust with a concentration of 0.0002%, but it has intense chemo-toxic and radio-toxic potential (Suma et al., 2016). It has also been reported that the U has no valid function in living beings and is considered a non-essential element. Even in 1991, USEPA considered U as a top carcinogen and suggested its non-existence as the safe limit of carcinogenic risk in drinking water. U is a naturally occurring toxic element present ubiquitously in every matrix of the environment. It enters into groundwater by a natural pathway through weathering of rocks bearing U. However, its solubility in groundwater depends on the regional lithology, geology, and geomorphology of the area (Kale et al., 2020). Some factors that make conditions conducive for U dissolution from parent rocks are partial pressure of CO_2, the concentration of different types of ions, salts (bicarbonate, phosphate, and nitrate, etc.), water-rocks interactions, and residence time (Rosen et al., 2019). The accumulation of bicarbonates enhances the dissolution of U in groundwater by forming water-soluble uranyl carbonate complexes (Coyte et al., 2018; Duggal et al., 2021b). In oxic water, U exists in the form of Uranyl ions (UO_2^{2+}) or U (VI), in which it leaches into the groundwater from the host rocks, whereas in anoxic water, it remains in the reduced form i.e., U(IV) and becomes immobile. It is revealed from the previous studies that the main exposure pathway of U in living beings is drinking water which contributes 85% while food contributes 15% (Adithya et al., 2019). Various epidemiological and health studies established the fact that the main target organs affected by U are the liver, kidney, and bones (Kim et al., 2004; Kurttio et al., 2002; Leggett, 1989; Taylor & Taylor, 1997). Although the absorption of ingested U is very low which is about less than 5% (ASTDR, 2013; Corlin et al., 2016). Ingested U is distributed in bones (66%), liver (16%), kidneys (8%), and other soft tissues of the body (10%) (ASTDR, 1999; Wagner et al., 2011). It can impair the growth and development of bones in young individuals by substituting calcium in osseous tissues (Banning & Benfer, 2017). Along with its potent chemical toxicity, it is a radioactive element that emits alpha radiation. U, on ingestion, can mutate the DNA, because the alpha particles emitted by U can be easily absorbed by the human body which can lead to genetic mutations, chromosomal aberrations, and malfunction in the apoptosis processes, resulting in abnormal cell division and proliferation of cells (Wagner et al., 2011). All these abnormalities can facilitate carcinogenesis therefore, U is considered a significant carcinogen. Wagner *et al.* (2011) in their studies found that the incidence of different types of cancer including kidney cancer may be enhanced in the areas supplied by groundwater with elevated U concentration. Previous investigations suggested a link between elevated U daughter radionuclides (radium, radon, etc.) concentration in groundwater with breast, lung, bone, and blood cancer (Bean et al., 1982; Hess et al., 1983; Petersen et al., 2015; Tanwer et al., 2021). But radon contributes significant radiation dose through the inhalation pathway but is insignificant through the drinking water pathway. Some of the reported health effects due to U exposure have been derived from experimental animal studies and human epidemiology (Au et al. 1996; Zaire et al. 1997). If a human body gets an exposure of 0.10 mg kg^{-1} of U of body weight, it can lead to serious health hazards to the lungs and kidneys (Duggal et al., 2013; Panghal et al., 2017).

Comparatively high concentration of U has been observed in the state of Punjab and Haryana over other northern states of India (Rodell et al., 2009;Chahal et al. 2019; Saini et al., 2016). Various studies in Haryana state itself have reported high U content in groundwater (Daulta et al., 2017; Duggal et al., 2017; Duggal & Sharma, 2017; Garg et al., 2014; Kansal et al., 2011; Panghal et al., 2017; Singh et al., 2014). The same concern has been observed in neighboring states like Punjab, Rajasthan, Uttar Pradesh, Himachal, Uttarakhand, Jammu, etc. (Bhardwaj et al., 2020; Mittal et al., 2017; Sharma et al., 2017; Sharma et al., 2017; Singh et al., 2016). The investigated area is devoid of such kind of study. Therefore, after going through the water quality scenario of Haryana and neighboring states, it becomes necessary to evaluate the water quality in U aspect and the radiochemical hazards attributed due to its consumption. the present study aims to 1) To evaluate the distribution of U and the chemo-radiological risks associated with its consumption, 2) Groundwater suitability was estimated using the water quality index (WQI), 3) A correlation study is also done to understand the behavior of U and possible causative ions and salts for its dissolution in groundwater.

MATERIALS AND METHODS

Panchkula has been chosen as the study location. It has an area of 898 km^2 and is in the northern part of Haryana between latitudes of 30°26' and 30°55' north and 76°46' and 77°10' east (**Figure 1**). Its borders are shared by two states -Himachal in the north and northeast and Punjab in the south as well as one district, Ambala, in the west. Siwalik Hills may be found in Panchkula's northern and northern-eastern districts, whereas alluvial plains can be found in the southern and southern-western regions. Siwalik Hills and alluvial plains are separated by gently sloping lowlands at a 500 m elevation. Ghaggar and its tributaries are the principal drainage rivers. Sirsa Nadi, a tributary of the Sutlej River, drains a small portion of the district in the northwestern corner. Surface and groundwater resources are abundant in Panchkula. Groundwater is mostly utilised for irrigation purposes. The average annual precipitation is approximately 1057 mm, and the net irrigated area as a proportion of the total cultivated area is 91.6% (CGWB, 2013) . The southwest monsoon, which occurs from June to September, contributes 86% of the total annual rainfall,

while non-monsoon rains account for 14%. Siwalik Hills, Kandi Belts, and Alluvial Plains are the three primary physiographic divisions of the district. The Siwalik Hills are distinguished by their vast tableland geography and steep slopes. A number of ephemeral streams originated from Siwaliks and flow down the outer slopes, distributing a large amount of gravel, rocks, and pebbles over the area. The soils are classed as loamy skeleton typic, lithyhic, eurtrochrepts/udorthents and range in type from loamy sands to fine sandy loams, with the exception of depressions, are well-drained, non-alkali, non-saline, non-calcareous, and usually base saturated. These soils are located in the Siwalik Range, whereas in the Yamuna Plains, water-logged soils with a loam to clay loam texture that exhibit the glazing effect are categorised as aeric/typic haplaquepts (CGWB, 2013). **Figure 1** displays the samples that were taken around the Panchkula district at various locations.

A total of 36 samples were collected from the studied region in December, 2018. Systematic sampling was implemented by gridding the district into 6×6 km^2 and most preferably collected from the center of the grid (Sharma et al., 2021, Tanwer et al., 2022a). If the center was not accessible practically, then the most populated area of the grid was the second choice for sample collection. For the sampling of groundwater, acid-treated polyethylene bottles (that had been immersed in 10% nitric acid overnight and repeatedly rinsed with distilled water to wash away impurities from the inside wall of the container) were utilized. To obtain a uniform and fresh collection of samples, the sampling source such as the hand-pump, tube well, and submersible was left to run for two to three minutes. To decrease the possibility of error, sample bottles were rinsed with source water before being collected to exclude any residues of acid or distilled water. During the collection of samples, seven parameters, including pH, Electrical Conductivity, Total Dissolved Salt, salinity, Dissolved Oxygen, Oxidation Reduction Potential, and temperature,

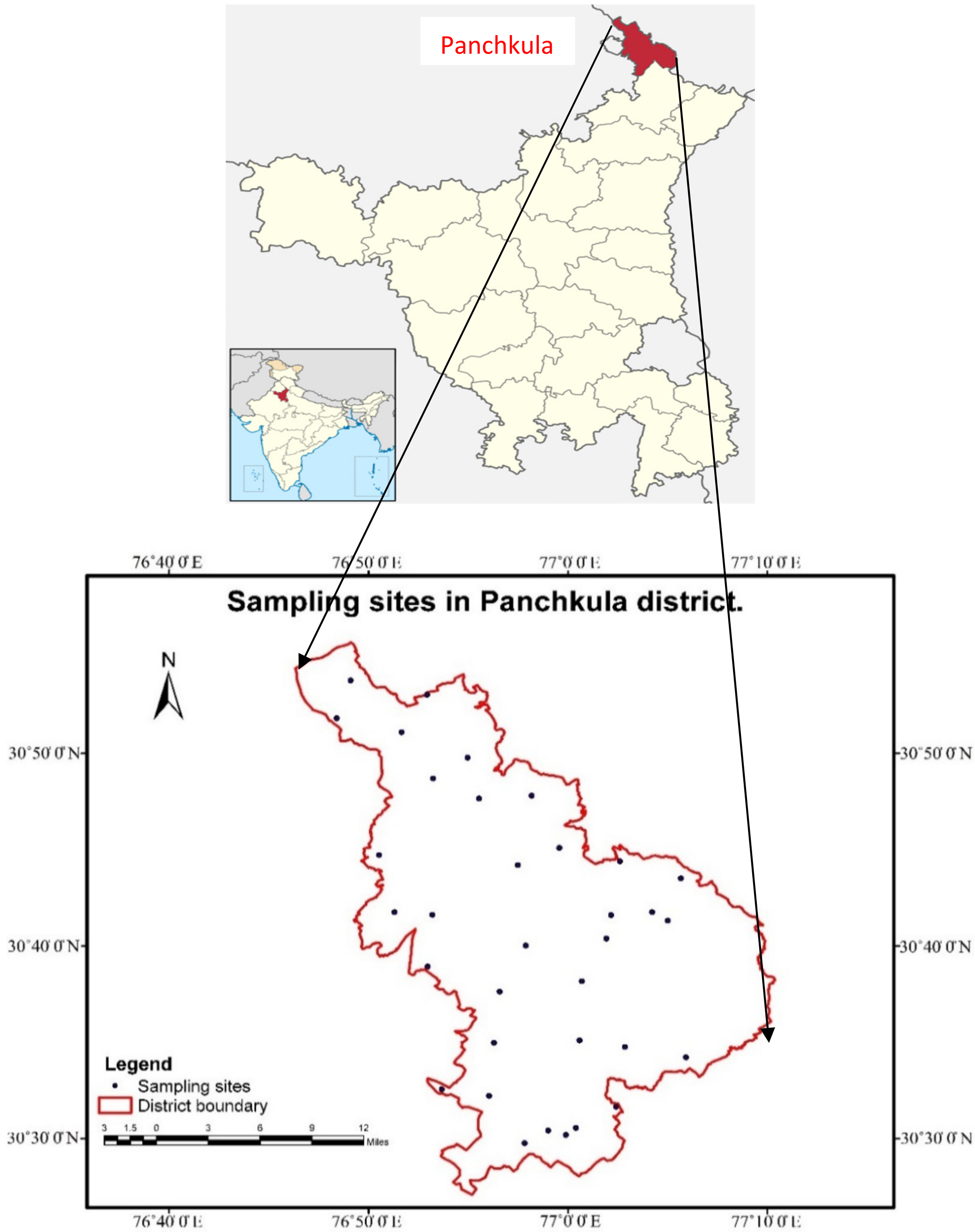

Fig. 1. Map of Panchkula showing sampling points

were measured on-site. Two bottles of one liter each were brought to the laboratory for analysis of the following parameters: hardness, alkalinity, nitrate (NO_3^-) using the first bottle and chloride (Cl^-), fluoride (F^-), sulphate (SO_4^{2-}), phosphate (PO_4^{3-}) and uranium (U) using 2nd bottle.

In order to assess the purity and potability of groundwater, 18 groundwater quality parameters were determined. Using mobile water sensors, the following seven *in-situ* water quality parameters were measured: pH, EC (Electrical Conductivity), TDS (Total Dissolved Salt), salinity, DO (Dissolved Oxygen), ORP (Oxidation Reduction Potential), and temperature. The pH, EC, TDS, salinity, and temperature were tested using multiparameter PCSTestr™35 (Eutech, Thermo Scientific, USA). The ORP and DO were measured using Waterproof ORPTestr®10

(Eutech, Thermo Scientific, USA) and DO6+ (Eutech, Thermo Scientific, USA) respectively. The portable water quality sensors were calibrated with known standards in Chemistry Research Lab, Aggarwal College Ballabgarh. According to the standard protocol from APHA (2005), the hardness and alkalinity were measured by titrating with standard solutions of EDTA (Ethylene diamine tetra-acidic acid) and sulphuric acid, respectively. By titrating with standard EDTA, Ca^{2+} hardness was determined, and Mg^{2+} hardness was obtained by deducting Ca^{2+} hardness from total hardness. Alkalinity formulas were used to compute carbonate and bicarbonate. The argentometric technique was used to analyse the chloride in water. According to the APHA, 2005 standard protocol, the SO_4^{2-}, F^-, NO_3^-, and PO_4^{3-} concentrations were measured by photo-spectrometery (UV-Visible spectrophotometer, Antech) using the turbidity method, SPADN dye method, uv spectrophotometric, and stannous chloride method respectively. Procedure blank measurements, careful standardization, spiked and duplicate samples were used to ensure the accuracy of the water quality analysis.

U estimation has been done using LED Fluorimeter Model LF-2, developed by Quantalase Enterprises Pvt. Ltd., Indore, India, with a minimum detection limit of 0.50 μg L^{-1} concentration (Saini et al., 2016). It is based on the principle of fluorescence measurement due to the presence of uranyl ions in an aqueous medium. The uranyl ions contained in the sample are excited by a UV LED in the LED fluorimeter, which emits 400 nm light. The photomultiplier tube measures the green light that the ions release when they return to the ground state. To remove fluorescence produced from the organic matter, appropriate filters are placed between the sample and the photomultiplier tube (PMT). The fluorescence from organic materials is removed using proper time gating technology because its life span is only approximately 100 ns, compared to about 200 μs for uranyl ions. PMT amplifies the fluorescence signal, which is further converted into digital form by the microcontroller. The matrix effect caused by a fluorescence quenching agent was further neglected using the standard spiking method. The fluorescence enhancement reagent, also known as fluran reagent, was made by mixing 5% sodium pyrophosphate with ortho-phosphoric acid to get the pH level to neutral. To change all Uranium species into a single form of the same fluorescence, the fluran reagent was added in a fraction equal to one-tenth of the sample volume (Sharma et al., 2019). Two subsequent standard additions were done to the sample and their corresponding fluorescence was recorded to estimate uranium concentration using the standard curve equation. U concentration in calibrated mode was calculated as given in equation 1:

$$\text{Concentration of U in unknown sample} = \text{CF} \times \left(\text{Fluorescence from sample} - \text{Fluorescence from water}\right) \tag{1}$$

Calibration factor (CF) was computed using equation 2 (Sharma et al., 2019):

$$\text{Calibration factor (CF)} = \frac{\text{Concentration of U in standard solution}}{\left(\text{Fluorescence of standard} - \text{Fluorescence of water}\right)} \tag{2}$$

The spread of Uranium in groundwater was interpolated spatially in Panchkula district using the inverse distance weight (IDW) method of ArcGIS 9.3 software. The inverse distance weight method helps to predict the value of locations close to the sampled one, which is probably more related than those which are farther apart.

Radiological risks are due to the radioactive nature of U and are calculated by the equations mentioned below-

The annual effective dose due to the consumption of U-contaminated water was calculated as per equation 3 (USEPA, 1999):

$$\text{AED } (\mu\text{Sv Y}^{-1}) = Ua \times DIW \times 365.25 \times DCF \quad (3)$$

where Ua , U activity concentration (Bq L^{-1}), 1 µg L^{-1} = 0.02528 Bq L^{-1} (Duggal et al., 2021), DIW, Daily Intake of Water, which is 4.05 liter per day (Saini et al., 2016), DCF, Dose conversion factor, which is taken as 4.5×10^{-8} Sv Bq^{-1} (ICRP, 2012).

Carcinogenic risk due to U intake in the form of excess cancer risk was calculated using equation 4 (USEPA, 1999):

$$\text{ECR} = Ua \times R \quad (4)$$

Where Ua , U activity concentration, and R, Risk Factor is calculated by equation 5 (USEPA, 1999):

$$\text{R (L Bq}^{-1}) = r \times IR \times EP \quad (5)$$

where R, risk factor (L Bq^{-1}), is the product of conversion factor (r) used here 1.9×10^{-9} Bq^{-1} (USEPA, 1999), IR, ingestion rate, taken here 4.05 liter per day (Bhardwaj et al., 2020) and EP, exposure periods, used here 23740 days including leap year days (65 years) (WHO, 2011).

Chemical risks due to its chemical toxicity are associated with the consumption of U-contaminated water. The following equations were used to quantify its considerable effects on the liver, kidney, and bones as a highly toxic heavy element:

Lifetime Average Daily Dose due to intake of U-contaminated water was calculated using equation 6 (USEPA, 1999):

$$\text{LADD } (\mu\text{g kg}^{-1}\text{ day}^{-1}) = \frac{U \times DIW \times EF \times AE}{LE \times AW} \quad (6)$$

where U is uranium concentration present in groundwater (µg L^{-1}), DIW is the daily intake of water i.e., 4.05 L day^{-1}, EF is exposure frequency i.e., 350 days $year^{-1}$, AE is average exposure duration i.e., 65 years [54], LE is life expectancy in days i.e., 25201 days taken from world bank data (World Bank, 2017), and AW is the average weight of body i.e., 53 kg (Sharma et al., 2017).

Hazard Quotient was calculated to know the chemical effects on humans due to U-contaminated water. Its value should be less than 1. If HQ value is greater than 1, it can cause a significant harmful impact on the targeted organs. It is calculated as per equation 7 (USEPA, 1999):

$$\text{HQ} = \frac{LADD}{RD} \quad (7)$$

where RD, Reference Dose, 4.53 µg kg^{-1} day^{-1} (AERB,2004).

By considering the impact of specific parameters, the water quality index is an appropriate method for assessing the overall quality of water. Water quality was determined using the computed weightage of each parameter presented in **Table 1**. Equation 8 was used to calculate it using the weighted arithmetic index approach (Brown et al. 1970) as follows:

$$\text{WQI} = \frac{\Sigma \text{Wn} \times \text{Qn}}{\Sigma \text{Wn}} \quad (8)$$

Table 1. Water quality index, their respective grade, and possible usage

Water Quality Index	Water Grade	Possible usage
0-25	Excellent	Drinking, irrigation, and industrial
26-50	Good	Drinking, irrigation, and industrial
51-75	Poor	Irrigation and Industrial
76-10	Very poor	Irrigation
<100	Not suitable for Drinking	Require treatment before use

Table 2. Water quality parameter, relative weight index and permissible limits (Brown et al., 1970).

S. No.	Parameters	W_n= K/S_n	1/S_n	Permissible limits (WHO, 2012)
1	pH	0.136	0.118	8.5
2	TDS (mg L^{-1})	0.001	0.001	1000
3	EC (μS cm^{-1})	0.001	0.001	1500
4	F^- (mg L^{-1})	0.771	0.667	1.5
5	Cl^- (mg L^{-1})	0.005	0.004	250
6	NO_3^-(mg L^{-1})	0.023	0.020	50
7	SO_4^{2-}(mg L^{-1})	0.005	0.004	250
8	U (μg L^{-1})	0.039	0.033	30
9	Total hardness (mg L^{-1})	0.002	0.002	500
10	Total Alkalinity (mg L^{-1})	0.002	0.002	500
11	Ca^{2+} (mg L^{-1})	0.004	0.003	300
12	Mg^{2+} (mg L^{-1})	0.012	0.010	100

where W_n, unit weight of n^{th} parameter used to calculate the water quality index (**Table 2**), Q_n, quality rating of n^{th} parameter considered to calculate the WQI. Unit weight (W_n) is calculated by equation 9:

$$W_n = \frac{k}{Sn} \tag{9}$$

where k, proportionality constant, S_n, permissible limit for water quality parameter as per WHO, 2012 and calculated as per equation 10:

$$k = \frac{1}{\Sigma 1 / Sn} \tag{10}$$

Quality rating (Q_n) was calculated using equation 11:

$$Q_n = \frac{(Pn - Ci)}{(Sn - Ci)} \times 100 \tag{11}$$

where P_n, is the actual value of analysed water parameter, C_i, is the ideal concentration of water parameter and is considered as Zero except for pH and DO which are considered neutral and 14.6 mgL^{-1} respectively.

RESULT AND DISCUSSION

In this study of Panchkula district, a total of 36 samples were analyzed for 18 water quality parameters including U. All the samples were preserved as per APHA (2005) and analyzed within 30 days of sampling. The statistical summary for all the estimated water quality parameters is given in **Table 3**. In the physicochemical analysis of water quality, it has been found that pH, EC, TDS, salinity, DO, ORP, and temperature were found to be in the range of 6.70 - 8.40, 302 - 1200 μS cm^{-1}, 210 - 840 mg L^{-1}, 138 - 570 mg L^{-1}, 4.10-14.05 mg L^{-1}, 135 - 392 mV and 16.10 - 24.20 ˚C respectively. TDS in all the samples were well below the permissible limit of 2000 mg L^{-1} as per BIS (2012) with a mean value of 458.67 mg L^{-1} [59]. However, in approximately 42% of samples, the TDS was observed higher than the acceptable limit of 500 mg L^{-1} as per BIS (2012). Except for nitrate, the value of major cations and anions such as fluoride, chloride, sulphate, calcium, and magnesium were below the prescribed limit of BIS (2012) (**Table 3**). Total hardness (TH) was found to be in the range of 95-350 mg L^{-1} with a mean value of 241.39 mg L^{-1}. It was observed that TH in 75% of samples was higher than the desirable limit of 200 mg L^{-1} as per BIS (2012). The study region is influenced by Siwalik formation which comprises of the rocks like limestone, sandstone, shale, and dolomite that could be the sources of the higher level of calcium and magnesium hardness, which may be a probable cause of the higher level of total hardness in groundwater of the area. The hardness was mainly contributed by the dissolution of calcium and magnesium salts and their values were observed to be varied in the range of 10-72 mg L^{-1} and 0-61.20 mg L^{-1} respectively. Nearly, 64% of samples were observed to have magnesium content higher than the acceptable limit of 30 mg L^{-1} but in all the samples calcium and magnesium content were found to have less than the permissible limit of 300 mg L^{-1} and 100 mg L^{-1} as per BIS (2012) respectively. In some samples, the magnesium content was found to be higher than the calcium. Possibly, this may be due to the possibility of calcium precipitation at supersaturation that causes the rise of magnesium content in water (Hem, 1991). This may also be dependent upon the type of rocks present in that specific region. The average value of total alkalinity was found to be 393.19 mg L^{-1}, which is less than the permissible limit of 600 mg L^{-1} as per BIS (2012). The carbonate and hydroxide ions in all the water samples collected from the study region were absent as the phenolphthalein alkalinity in all the samples was observed to be zero. This could be possible due to the fact that at pH 8.3, all carbonate ions present in groundwater convert into bicarbonates ions. Nitrate, only in two samples namely Palsara and Tagra Harisingh, and total alkalinity in one sample, Tanda Bhagwanpur, were above the threshold limit of BIS (2012). The amount of nitrate in drinking water can cause diseases like heart malfunctioning, gastric cancer, and blue baby syndrome (CPCB, 2008). The higher amounts of nitrate in groundwater at these locations indicate the pollution of groundwater due to some anthropogenic activities like application of fertilizers in the soil.

To analyse U, each sample was filtered through the 0.45μ Whatman filter paper to remove any suspended solids in the water. The estimated U concentration was found to be in the range of 1.70 μg L^{-1} – 12.28 μg L^{-1} with a mean value 5.89 μg L^{-1}. The U in the groundwater of all studied locations of Panchkula district was found to be far below the permissible limit of WHO (2011), AERB (2004), and USEPA (2011). The statistical analysis of the U data displayed that the mean value of U was slightly higher than the median which indicates that some samples are on the higher side as compared to the lower side of U. Out of 36 samples, in 16 samples, the U content was higher than the average value of U found in the region. The 75th percentile of U data was 7.32 μg L^{-1} which is even far below the most acceptable concentration of 20 μg L^{-1} as per Canadian Health guidelines for U in drinking water (Health Canada, 2019). The skewness value of U data was greater than zero indicating the slight positive skewness in U data distribution. This is also well cleared from the distribution curve (**Figure 2**). The continuous distribution of U is well presented through the interpolation map of Panchkula district (**Figure 3**). The inverse

Table 3. Descriptive statistical analysis of water quality data

	pH	TDS (mgL^{-1})	EC (μScm^{-1})	ORP (mV)	Temp (°C)	Salinity (mgL^{-1})	DO (mgL^{-1})	F^- (mgL^{-1})	Cl^- (mgL^{-1})	NO_3^- (mgL^{-1})	SO_4^{2-} (mgL^{-1})	PO_4^{3-} (mgL^{-1})	U (μgL^{-1})	TH (mgL^{-1})	Ca^{2+} (mgL^{-1})	Mg^{2+} (mgL^{-1})	TA (mgL^{-1})	HCO_3^- (mgL^{-1})
Mean	7.38	458.67	645.33	285.72	20.79	309.03	7.83	0.16	33.69	18.51	30.55	0.02	5.89	241.39	42.44	32.47	393.19	392.64
Standard Error	0.06	18.94	27.01	6.12	0.38	13.16	0.29	0.03	1.84	7.70	1.61	0.00	0.41	9.56	2.99	2.11	13.12	13.23
Median	7.35	472.50	655.00	288.00	21.10	322.00	8.19	0.06	32.49	8.58	26.87	0.02	5.55	247.50	40.00	32.40	382.50	382.50
Mode	7.10	440.00	650.00	292.00	20.20	340.00	8.80	0.00	27.49	NA	20.05	0.02	NA	240.00	50.00	31.20	375.00	375.00
Standard Deviation	0.36	113.61	162.08	36.72	2.28	78.99	1.76	0.21	11.03	46.19	9.65	0.01	2.49	57.35	17.95	12.68	78.71	79.38
Kurtosis	1.93	2.97	3.26	8.97	-0.58	2.70	4.01	1.26	16.14	30.37	-0.65	1.90	0.00	0.24	-0.93	0.58	2.14	2.04
Skewness	1.13	0.39	0.49	-1.38	-0.45	0.40	0.53	1.46	3.46	5.36	0.74	1.13	0.54	-0.59	-0.02	-0.13	0.99	0.96
Range	1.70	630.00	898.00	257.00	8.10	432.00	9.95	0.76	64.98	276.88	33.62	0.03	10.58	255.00	62.00	61.20	390.00	390.00
Minimum	6.70	210.00	302.00	135.00	16.10	138.00	4.10	0.00	22.49	0.73	17.50	0.01	1.70	95.00	10.00	0.00	260.00	260.00
Maximum	8.40	840.00	1200.00	392.00	24.20	570.00	14.05	0.76	87.47	277.61	51.12	0.04	12.28	350.00	72.00	61.20	650.00	650.00
1st Quartile	7.10	422.00	598.75	275.00	19.83	280.25	7.24	0.00	27.49	3.13	23.31	0.02	3.99	207.50	29.50	24.90	345.00	345.00
3rd Quartile	7.50	514.00	722.50	303.75	22.20	345.50	8.80	0.28	37.48	13.03	36.66	0.02	7.32	281.25	58.00	41.10	425.00	425.00
Acceptable limits (BIS,2012)	6.5-8.5	500	NA*	NA*	NA*	NA	NA*	1.0	250	45	200	NA*	30**	200	75	30	200	NA*
Permissible limits (2012)	NR***	2000	NA*	NA*	NA*	NA*	NA*	1.5	1000	NR***	400	NA*	NA*	600	300	100	600	NA*

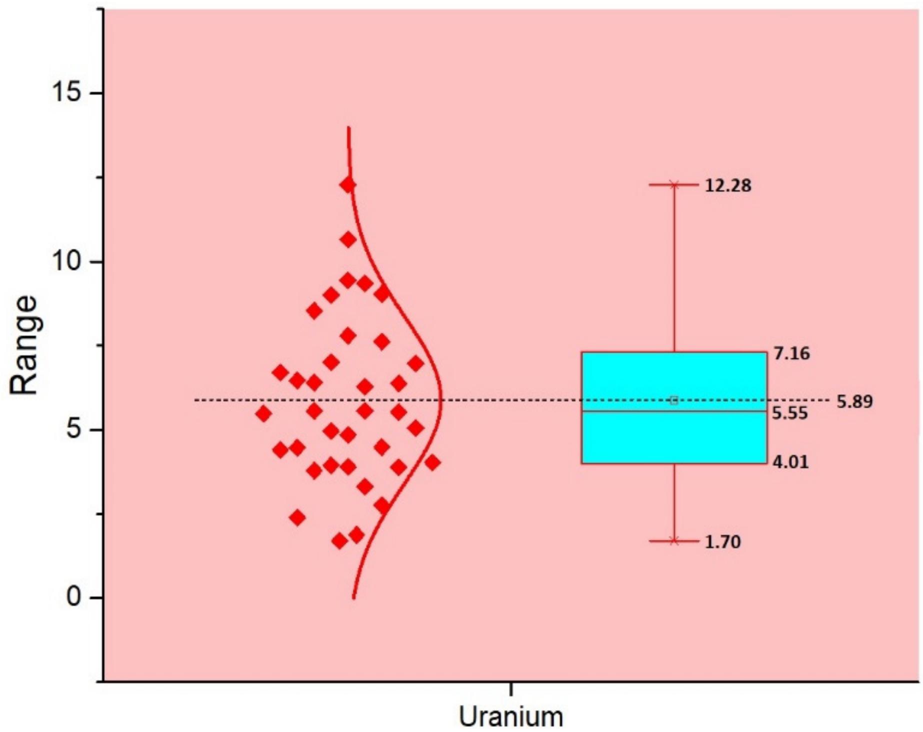

Fig. 2. Whisker box plot and distribution curve of U

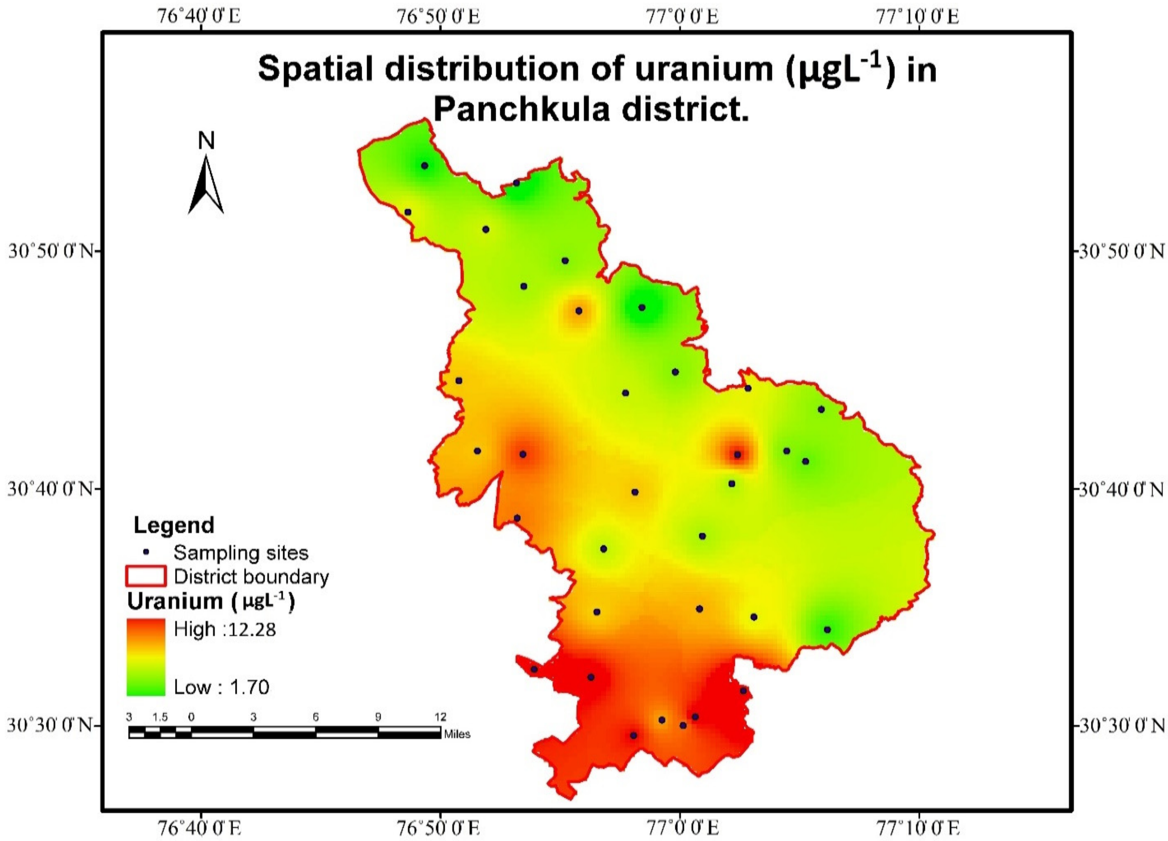

Fig. 3. Interpolation map of U distribution in Panchkula district of Haryana

distance weighted method was employed to predict the spatial distribution of U from sampled locations to unknown ones. It has been observed that the southern region of Panchkula was found to bear comparatively more concentration of U than the northern and north-eastern regions shown in red colour in the interpolation map (**Figure 3**). The few samples in western and central regions of Panchkula are also in the higher range than the mean value of U. The natural geology and underlying rocks of the area may be responsible for such behavior. High Uranium contamination was found in the alluvial plains of a nearby district as reported in earlier investigations of groundwater (Rishi et al., 2017). The sediments of these districts' plains were transported from neighbouring Siwalik, granitic, and metamorphic rock formations (Pant et al., 2017). Due to Uranium mineralization, Atomic Minerals Directorate for Exploration and Research (AMDER) has investigated the geology of the Siwalik areas. Therefore, such behaviour of relatively higher concentration of U in the southern region of district, may be accounted to the geology and geochemistry of underlying rocks of this region. This region is also in close contact with Punjab district that has been found to bear exceptionally high U in groundwater (Bajwa et al., 2017; Saini & Bajwa, 2016; Saini et al., 2016). The recent studies of Haryana district reported that groundwater of Hisar, Sonipat, Panipat, Sirsa, Jhajjar, Palwal and Bhiwani districts were found to be higher than the permissible limit of 30 ppb of WHO and BIS (Singh et al., 2021; Tanwer et al., 2022a, b).

Using the U content and their corresponding activity in groundwater, the chemical and radiological toxicity was computed. It was observed that the annual effective dose (AED) due to the U-contaminated water was found to vary in the range of 2.86 – 20.66 μSv Y^{-1} with the mean value of 9.91 μSv Y^{-1}, which is far below than the standard permissible value of 100 μSv Y^{-1} as per WHO (2011) (**Table 4**). The value of AED was observed to be 17.80 μSv Y^{-1}, which is due to the inconsistent distribution of U in the groundwater of the area. The AED value in 44.44% of samples was higher than the mean value of the region. The ECR was estimated and found to vary from 4.92E-06 to 3.55E-05 with an average of 1.70E-05. The value of ECR due to all the groundwater samples of Panchkula district was found to be less than the permissible limit of 1.67E-04 as per AERB (2004). The LADD was found to vary in the range of 0.12-0.85 μg kg^{-1} day^{-1} with an average value of 0.44 μg kg^{-1} day^{-1}. 44.44% of samples were found to have LADD values more than the average value of the district. However, in all the samples, the LADD value was observed to be less than the permissible limit of 4.53 μg kg^{-1} day^{-1} as per AERB (2004), this implies the suitability of groundwater for drinking purposes from the U point of view. HQ was computed based on LADD and observed that 44.44% of samples bear HQ values higher than the mean value of 0.09. Although, HQ value due to the consumption of U-contaminated groundwater in all the samples was less than 1, thereby indicating no significant chemo-toxic effect. Therefore, observed results of chemical and radiological risk assessment, display no potential chemo-radiological impacts due to U contamination in the groundwater of Panchkula district.

WQI is a crucial tool for determining the quality and long-term sustainability of water for drinking. These water quality indicators are regarded as reporting tools for assessing water quality. The water quality index was calculated based on U and some other water quality parameters that have significant impacts on human health too have been checked and compared with the permissible limits recommended by concerned agencies for them. The value of WQI was estimated to be in the range of 0.36-47.43 with an average of 14.06. The WQI value calculated for each sample is given in **Table 4**. The WQI of all the samples was found to be less than 50. The WQI value of 86% of samples was in the range of 0-25 which indicates the quality of groundwater samples is excellent and can be used for drinking, irrigation, and industrial purposes while 14% of samples were found to range from 26-50, stating that the quality of water is good and can also be used for all purposes. No sample was found in the range of 51-75, 76-100, and above 100 which implies the unsuitability of water for drinking purposes. Thus, all the

Table 4. Chemo-radiological effects and water quality index

S. No.	Location name	Sample No.	U (μgL^{-1})	Uranium Activity (BqL^{-1})	AED (μSvY^{-1})	Excess Cancer Risk	LADD ($\mu gkg^{-1}day^{-1}$)	HQ	WQI
1	Bhaoli	1	12.28±0.10	0.31	20.66	3.55188E-05	0.85	0.19	30.61
2	Thanura	2	9.44±0.09	0.24	15.89	2.73043E-05	0.65	0.14	22.09
3	Bhagwali	3	7.80±0.08	0.20	13.13	2.25608E-05	0.54	0.12	7.47
4	Golpura	4	7.01±0.08	0.18	11.80	2.02758E-05	0.48	0.11	47.43
5	Kakrali	5	9.01±0.07	0.23	15.16	2.60606E-05	0.62	0.14	19.19
6	Bhagwanpur	6	9.35±0.10	0.24	15.73	2.7044E-05	0.64	0.14	15.69
7	Sangwana	7	10.66±0.09	0.27	17.94	3.08331E-05	0.74	0.16	8.86
8	Jalauli	8	6.28±0.09	0.16	10.57	1.81643E-05	0.43	0.10	21.94
9	Kot	9	4.85±0.07	0.12	8.16	1.40282E-05	0.33	0.07	3.25
10	Bunga	10	6.40±0.08	0.16	10.77	1.85114E-05	0.44	0.10	5.25
11	Ramgarh	11	7.62±0.07	0.19	12.82	2.20402E-05	0.53	0.12	0.36
12	Nada Sahib	12	8.54±0.09	0.22	14.37	2.47012E-05	0.59	0.13	43.93
13	Chandi Mandir Sec-4	13	6.47±0.06	0.16	10.89	1.87139E-05	0.45	0.10	8.75
14	Salutri	14	6.38±0.06	0.16	10.74	1.84536E-05	0.44	0.10	7.48
15	KharkuaJala	15	4.96±0.06	0.13	8.35	1.43464E-05	0.34	0.08	0.45
16	Kidarpur	16	3.90±0.06	0.10	6.56	1.12804E-05	0.27	0.06	6.57
17	Tipra	17	5.57±0.05	0.14	9.37	1.61107E-05	0.38	0.08	5.77
18	Koti	18A	5.56±0.06	0.14	9.36	1.60818E-05	0.38	0.08	21.83
19	Koti	18B	3.95±0.05	0.10	6.65	1.1425E-05	0.27	0.06	13.85
20	Morni	19	3.32±0.03	0.08	5.59	9.6028E-06	0.23	0.05	13.42
21	Balag	20	3.79±0.03	0.10	6.38	1.09622E-05	0.26	0.06	11.46
22	Palsara	21	9.04±0.11	0.23	15.21	2.61474E-05	0.62	0.14	9.16
23	Paonti	22	4.49±0.07	0.11	7.56	1.29869E-05	0.31	0.07	10.02
24	Parwala	23	4.47±0.05	0.11	7.52	1.29291E-05	0.31	0.07	34.18
25	Raipurani	24	6.71±0.08	0.17	11.29	1.94081E-05	0.46	0.10	5.90
26	Sultanpur	25	5.53±0.08	0.14	9.31	1.5995E-05	0.38	0.08	13.79
27	Bhud	26	2.76±0.05	0.07	4.64	7.98305E-06	0.19	0.04	4.69
28	Damdame	27	6.97±0.06	0.18	11.73	2.01601E-05	0.48	0.11	29.57
29	Damala	28	4.41±0.03	0.11	7.42	1.27555E-05	0.30	0.07	2.78
30	Chirnian	29	5.06±0.03	0.13	8.52	1.46356E-05	0.35	0.08	7.99
31	Tanda Bhagwanpur	30	5.48±0.08	0.14	9.22	1.58504E-05	0.38	0.08	11.60
32	Lahroundi	31	2.39±0.03	0.06	4.02	6.91286E-06	0.16	0.04	6.08
33	Tapriyan	32	4.03±0.03	0.10	6.78	1.16564E-05	0.28	0.06	20.01
34	BanoiKhuda Box	33	1.88±0.02	0.05	3.16	5.43773E-06	0.13	0.03	9.77
35	TagraHarisingh	34	3.89±0.06	0.10	6.55	1.12515E-05	0.27	0.06	16.24
36	Janauli	35	1.70±0.06	0.04	2.86	4.9171E-06	0.12	0.03	8.77

samples of Panchkula district are well-suitable for drinking purpose but a few parameters like total alkalinity, nitrate, and U needs to be monitored regularly.

Correlation was estimated between the water quality parameters to understand the relation with U and their interdependency with each other. The matrix generated shown in **Figure 4** indicates the correlation between various parameters estimated in groundwater samples of Panchkula. The t-test was also used to validate the significance of the association between the two variables. The correlation coefficients with p-values less than 0.05 are seen to be significant because the significance level (p-value) less than 0.05 indicates the statistical significance of the correlation with a 95% level of confidence. Cross marked cells in **Figure 4** represent an

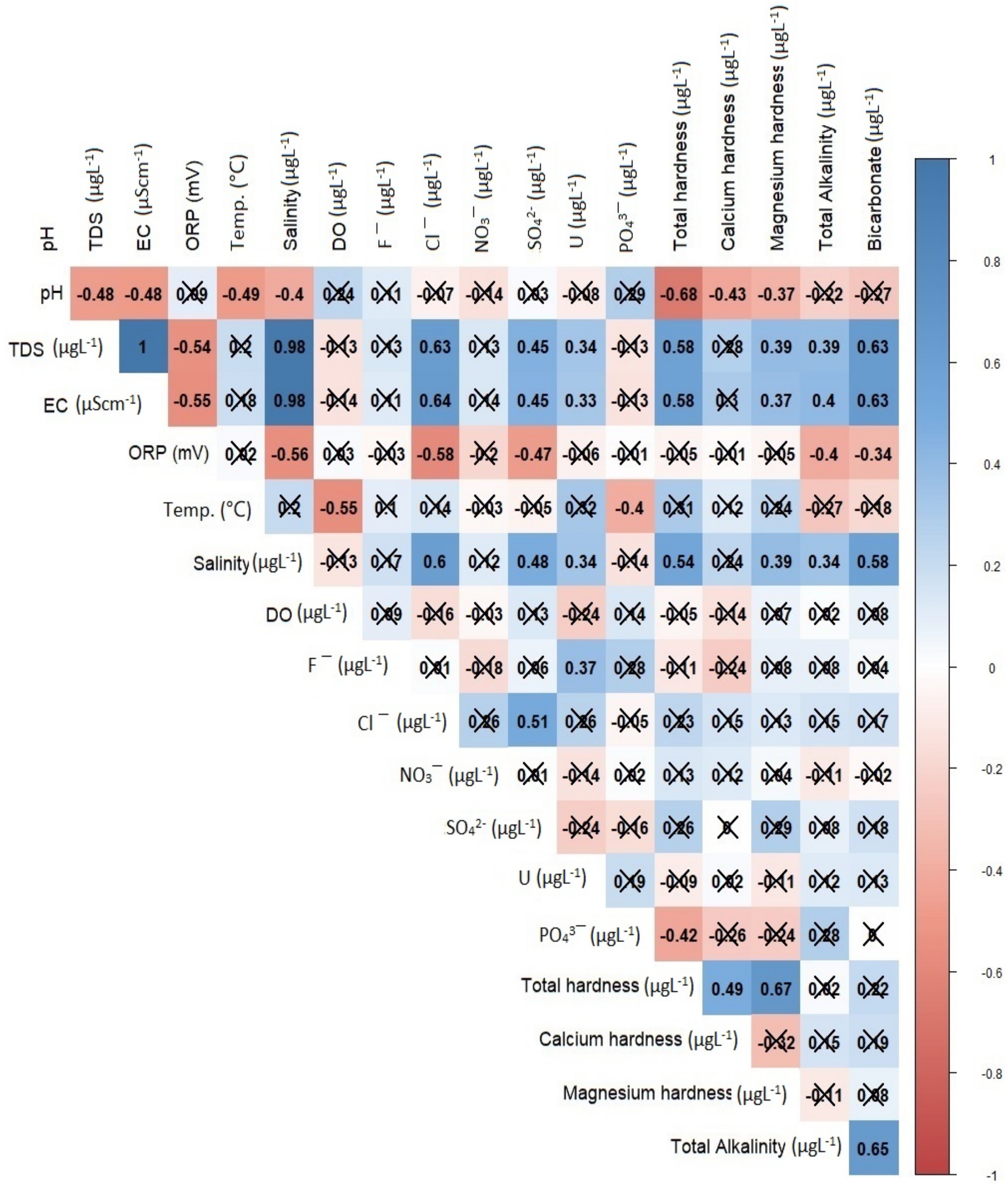

Fig. 4. Matrix of correlations between water quality parameters, as observed. Correlation strength is color-coded in the index bar. The statistical significance of the calculated correlation is indicated by non-crossed cells (p-value less than 0.05).

insignificant correlation coefficient at the 0.05 level of significance, whereas non-crossed cells indicate a significant relation. Correlation study also helps to understand the behaviour of U in groundwater and role of different anions and cations, to make conditions favourable to let it dissolve in groundwater from surrounding rocks and soil. In this study, no strong correlation of U was observed with the water quality parameters. A weak positive correlation was observed between U and TDS, EC, salinity and fluoride. It can be assumed that the high concentration of ions in groundwater interacts with U present in host rocks and brings it in water in the forms of different water-soluble complexes (Bhardwaj et al., 2020; Ortega et al., 1996; Sharma et al., 2019). According to previous studies, a strong positive correlation of U has been observed between U and TDS (Saini et al., 2016; Saikia et al., 2021), and a very weak positive correlation was seen between U and anions such as chloride and phosphate as observed by Sharma et al. (2019). U is also found to be positively correlated with calcium and magnesium which indicates the possibility of the formation of Ca (UO_2) $(CO_3)_3$ and Mg (UO_2) $(CO_3)_3$ (Fox et al., 2006; Sahu et al., 2020). Between U and water quality parameters like nitrate and sulfate, a weak negative correlation was found. TDS was found to be in positive correlation with chloride and total hardness indicating the probability of leaching of anions from rocks into groundwater (Bhardwaj et al., 2020). Total hardness was strongly found to be correlated with pH, EC, TDS, and salinity. Total hardness was found to have a strong negative correlation with pH, while a strong positive correlation was seen with EC and TDS.

CONCLUSION

In present study, a detailed investigation of U distribution has been done to estimate chemo-radiological impacts and possible significant factors that contribute to its dissolution in the groundwater of Panchkula district. The overall quality of groundwater using the water quality index was determined to evaluate its suitability for drinking purposes. U content in groundwater was observed in the range of 1.70-12.28 μg L^{-1} with the majority of samples of high U content in the southern region of Panchkula but no sample was found to cross the permissible limit of WHO (2011), AERB (2004) and USEPA (2011). Nearly 56% of samples were found with U concentration less than the average value of U in the groundwater of Panchkula district. The health risk analysis has found that the values of AED, ECR, and LADD are less than the standard permissible limits, recommended by AERB (2004). The HQ values in all the groundwater samples were less than 1 indicating that there is no probability of significant health risk. Except nitrate in two samples i.e., Palsara and Tagra Harisingh, and total alkalinity in one samplei.e., Tanda Bhagwanpur, all parameters studied were below the prescribed limits of BIS (2012). The Water Quality Index was found to be below 50, which implies the purity of water is good enough to be used for drinking purposes. The major portion of samples are observed in the category of excellent water and the rest are in the good category. The overall quality of water indicates that water is well-suitable for drinking purposes. In correlation, U was found to have a weak positive correlation with TDS, EC, salinity, and fluoride. A weak negative correlation of U was observed with nitrate and sulfate.

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CONFLICT OF INTEREST

The author declares that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission and redundancy have been completely observed by the authors.

LIFE SCIENCE REPORTING

No life science threat was practiced in this research.

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