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Seasonal Variation and Spatial Distribution of Uranium in Sources of Water in Tonk District of Rajasthan, India

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ABSTRACT

This study was undertaken to evaluate the concentration of Uranium (U) in the drinking water of the Tonk district of Rajasthan (India). The main objective of the study is to determine the distribution of Uranium concentration and the geochemical behavior of Uranium in pre-monsoon (PRM) and post-monsoon (POM) drinking water samples. Uranium was measured by LED fluorimeter. Total 318 drinking water samples were collected for both seasons. It is observed that the water quality of all the samples is within the limits prescribed by WHO (30 µg/L) except a few, and can be used for domestic purposes. The Uranium concentration was found to be in the range 0.21 to 173.72 µg/L with a mean value of 8.58 µg/L in pre-monsoon and 0.21 to 162.34 µg/L with a mean value of 11.22 µg/L in post-monsoon samples. The geochemistry of the study area shows rock-water interaction. The order of average anionic concentration is found to be $HCO_3^- > Cl^- > SO_4^{2-} > NO_3^-$. Although no definite trend of seasonal variation in the concentration of U was observed, large samples have higher Uranium concentrations in post-monsoon than pre-monsoon.

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INTRODUCTION

Groundwater is an essential constituent of the ecosystem, which affects both human life and biological growth (Aravinthasamy et al., 2021; Li et al., 2017). Many countries are facing groundwater contamination, which is a matter of serious concern (Kumar et al., 2022). The high concentration of contaminants in groundwater is a result of both natural processes and human activities (Brindha et al., 2020). The presence of Uranium (U) and Arsenic (As) in India's groundwater is a national concern also gaining global attention (Balaram et al., 2022; Shaji et al., 2020).

Uranium is a natural radioactive element emits gamma radiation. It contains three isotopes, ^{234}U (0.006%), ^{235}U (0.72%), and ^{238}U (99.27%), with half-life of 2.455×10^5, 7.038×10^8, and 4.468×10^9 years, respectively (Bjørklund et al., 2020). Uranium is found in hexavalent state as uranyl (UO_2^{2+}) ion in water (D et al., 2021). Excessive use of underground water leads to the increase in salinity and heavy metal concentration with subsidence of water. Uranium may also

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be present in these aquifers which come out with water and pose radiation hazards to human and other life forms. It is omnipresent and poses a nephrotoxic effect in humans if taken above permissible limits through food or water (Raghavendra T, et.al, 2014). In recent years Uranium contamination has been vastly examined in drinking water. It is present in trace quantities in granites, metamorphic rocks, lignite, phosphate, and in many mineral deposits (Haynes W M., 2014). Uranium content in rock varies from 0.5 to 4.7 µg/g. Igneous rocks such as basalt and granites may contain up to 4.7 µg/g of Uranium. 2.0 µg/g of Uranium is found in carbonate rocks and approximately 3.7 µg/g in sedimentary rocks such as shale (NCRPM, 1984).

The concentration of Uranium in ground drinking water is affected by the geochemistry of that area and the anions and cations present. Besides that, seasonal and temporal variations are conducive to Uranium dissolution in water (Shekhar et al., 2015). Groundwater's Uranium concentrations vary widely due to pH and oxic conditions (Coyte and Vengosh, 2020). A survey is being conducted in India in collaboration with local researcher and educational institutions to map the Uranium content in drinking water sources (Sahoo et al., 2021). The impact of Uranium in groundwater has been determined, with respect to the environment and human health by using various analytical techniques (Balaram et al., 2022).

Central Ground Water Board, Ministry of India has monitored Uranium concentration in ground drinking water and analyzed 14377 samples across the country from shallow aquifers in 2019-2020 (CGWB, Govt of India, 2020). The board reported Uranium concentration in the range 0.0 to 2876 µg/L. The climate of the Rajasthan state of India is arid to semiarid. The precipitation is low and variable therefore prone to drought. In Rajasthan, salinity of water is high with high contamination of Uranium. Based on the groundwater studies in the year 2018, from different districts of Rajasthan namely Jaipur, Jodhpur, Ajmer, Bundi, Kota, Barmer, Tonk, and Dausa concluded that groundwater contamination is due to anthropogenic sources like domestic and agricultural sewage as well as geogenic sources like water-rock interactions and evapotranspiration (Coyte et al., 2018).

In the Sikar district of Rajasthan, amount of Uranium in the groundwater is reported in the range of 8.20 to 202.63 µg/L (Duggal et al., 2016). About one-fourth and one-sixth of the samples of pre- & post-monsoon periods exceeded the WHO (World Health Organization) and AERB (Atomic Energy Regulatory Board) guidelines. The Uranium level in drinking water samples of Khetri Copper Belt in Rajasthan have been found varying 0.68 to 233.99 µg/L. High TDS was also reported at some locations of Khetri (Kumar et al., 2023). In Deoghar district of Jharkhand, groundwater samples contain Uranium in the range of 0.01-11.30 µg/L and 0.15-6.50 µg/L in PRM and POM, respectively (Srivastava et al., 2022).

The variation in the amount of Uranium in groundwater depends upon the geological feature of the area, environmental variables and seasonal variations (Sahu et al., 2020a). A low concentration of Uranium is invariably present in drinking water obtained from various sources, due to percolation and leaching of radio-nuclides through rocks. The geochemical characteristics of aquifer systems control groundwater chemistry. This research work has been developed to study the Uranium concentration mapping, gamma radiation and related water quality parameters in groundwater/drinking water in the Tonk district of Rajasthan.

No previous systematic Uranium detailed mapping of the ground drinking water in the Tonk district has been done. This study would be helpful in evaluating the distribution and seasonal variation of Uranium in potable water sources in the Tonk district of Rajasthan.

MATERIAL AND METHODS

Description of study area

The study area of the present work is the Tonk district situated in the eastern part of Rajasthan. It is delimited on the South by Bundi district, West by Ajmer, East by Sawai Madhopur and North by Jaipur district. It covers an area of 7,194 km^2 stretching between 75^0 07' to 76^0 19' East meridian and 25^0 41' to 26^0 34' North Latitude (CGWB, Tonk, 2013). The district is drained principally by the river Banas and its tributaries. The supply of water in the Tonk district is largely via the Bisalpur dam through pipeline. Samples were collected from open wells, borewells, handpumps and the supply pipelines. The seven tehsils constituting the Tonk district are Tonk, Uniara, Niwai, Todaraisingh, Deoli, Malpura, and Piplu. Tonk is a semi-arid region with hot dry summer and cold winter. Major rainfall occurs between June and September. Rainfall in the district is moderate with an annual average of 536.8 mm (CGWB, Tonk, 2022).

Geology and Hydrogeology

Tonk district is underlain by the Bhilwara Super Group of rocks, primarily mica schist, gneisses, phyllites, and quartzites with some intrusive granite. Alluvium of recent to sub-recent age, primarily composed of clay, sand, and silt, is found on top of these hard rocks (CGWB, Tonk, 2013). In the Tonk district's Malpura and Todaraisingh tehsils, granite gneisses are revealed. Groundwater is under phreatic conditions. The Major types of aquifers are alluvium and hard rock aquifers found in the study area. Mica-schist comprises 65% of the entire area, whereas around 20% of the area is covered by alluvium and 15% northwest area contains a gneissic aquifer (CGWB, Tonk, 2013).

The hydrogeology of the study area is mainly composed of an alluvium aquifer, mixed consolidated sedimentary (limestone, phyllite/schist, sandstone, slate), and crystalline basement/hard rock (mica-schist, granite-gneiss).

Sampling and Analytical Method

In pre-monsoon (PRM) 159 water samples were taken from March to May 2019 and in post-monsoon (POM) 159 water samples were taken from October to November 2019. In total 318 drinking water samples were collected from various villages in the Tonk district of Rajasthan, and were analyzed for Uranium and other physicochemical parameters. To investigate the effects of seasonal variation on the concentration of Uranium and other ions in the research area, the samples were collected twice. Water samples were taken from a variety of drinking water sources, including handpumps (35%), borewells (15–20%), Bisalpur supply (37–42%), and open wells (5%). The depth of the water table varied from 5 to 350 feet. The sampling strategy for uranium content and other water quality parameters were followed as per guidelines of geochemical mapping developed by the International Union of Geological Sciences (IUGS, 2005) Commission. To ensure unbiased sampling, the sampling area was divided into a grid of 6 x 6 km^2, with latitude and longitude serving as reference coordinates. All samples were taken close to the centroid of each grid to cover whole geographical area and a single sample was taken from each grid (Sahoo et al., 2021). The coordinates of the places were recorded using a portable GPS device (Garmin etrex-30), and the gamma radiation was analyzed by Polimaster (PM1403). Figure 1 display the GPS coordinates of sampling sites in red colour dots.

Standard sampling methods were adopted for the collection of water samples in the pre-treated PET bottles from the site of investigation. Samples were collected in duplicate to analyze physicochemical parameters in one and the Uranium concentration in another. To prevent Uranium from adhering to the bottle wall, 2 mL of concentrated HNO_3 (1 N) was added to the water sample. Using a portable multiparameter probe sensor (Eutech PCS-35, ORP, and DO meter), the pH, temperature, salinity, dissolved oxygen (DO) total dissolved solids (TDS), electrical conductivity (EC), and oxidation-reduction potential (ORP) of the water samples were measured at the sampling site. Major anions such as fluoride (F^-), nitrate (NO_3^-), phosphate (PO_4^{3-}), and sulphate (SO_4^{2-}) were examined in a laboratory by UV-visible spectrophotometer (Thermo-Fischer model Aqua Orion 8000). Argentometric method was used to determine chloride (Cl^-) in the sample and the titrimetric method was used to analyze carbonate (CO_3^{2-})

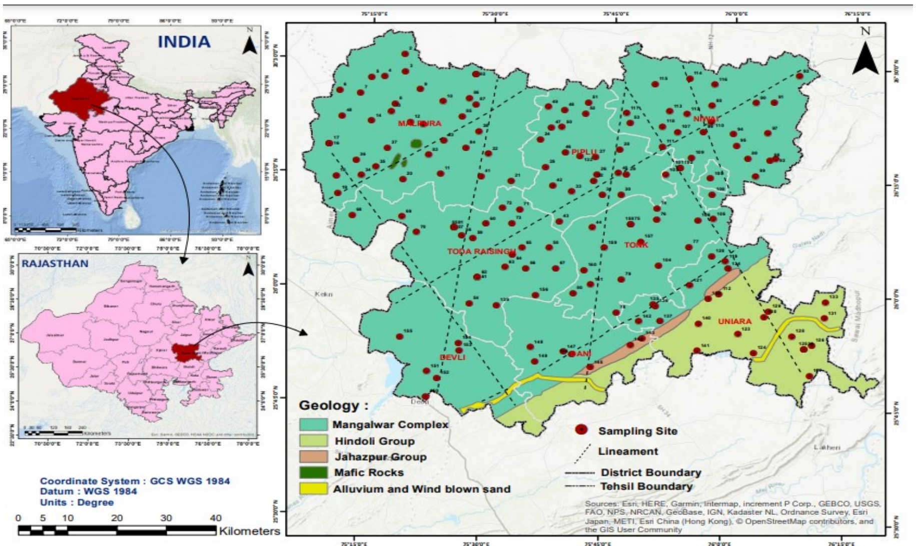

Fig. 1. Geographical Map and Sampling site of Tonk district

and bicarbonate (HCO_3^-). EDTA was used to evaluate hardness in the samples. The following equation was used to compute Mg^{2+} and Ca^{2+} from Mg-H and Ca-H, respectively(Patni et al., 2021):

$[Mg^{2+}]$ = Magnesium hardness/4.11

$[Ca^{2+}]$ = Calcium hardness/2.5 (2)

Uranium was analyzed with the help of LED fluorimeter model L2. To control the quality of data all instruments were calibrated with a certified standard solution before use and the minimum detection limit was determined. The APHA protocol (APHA, 2005) is followed for the analytical technique of each parameter. All measurements were taken in triplicate and the average value is considered for the result. The accuracy of result is found in the range of ±2-8%. Samples above 10% error were discarded.

RESULT AND DISCUSSION

Physicochemical parameters and Uranium concentration

The geochemistry and physicochemical parameters, namely pH, TDS, ORP, DO, temperature, and the ability of inorganic anions to combine with water (SO_4^{2-}, F^-, PO_4^{3-}, NO_3^- and silicate) are the factors which affect the availability of Uranium in groundwater. To explore the link between the physicochemical parameters and Uranium concentration, 16 water quality indicators, including pH, TDS, EC, DO, ORP, temperature, salinity, hardness, sulphate, fluoride, phosphate, nitrate, alkalinity, and chloride, have been analyzed by standard method. Depending on the geochemistry of the terrain, these hydro-chemical characteristics may have a favorable or negative impact on the Uranium content in water. Statistical analysis via hydrochemical analysis and correlation coefficient evaluation has been done to determine the relationship between Uranium and other hydrochemical parameters. To study the area, a total of 318 drinking water samples in PRM and POM periods were taken from the Tonk district of Rajasthan (India). In samples of groundwater the concentration of Uranium was found in the range of 0.21 to 173.72 µg/L with an average of 8.35 in PRM, and in POM it varied from 0.21 to 162.34 µg/L with an average of 11.22 µg/L.

Gamma radiation in six districts was found to be 70 to 325 nSv/hr in pre-monsoon and 89 to 215 nSv/hr in post-monsoon season.

Values of the gathered reports, comparisons between them

Several studies have been performed worldwide and in India. Uranium concentration in drinking water has been determined by a Laser Fluorimeter (Table 1a and 1b). It depicts the comparison of Uranium content in water present globally and in Indian states.

In the 2019–2020 report (CGWB, 2020), ICPMS was used to analyze approximately 671 water samples from various districts of Rajasthan for the determination of Uranium concentration. In

Table 1. Distribution of Uranium in Groundwater (a) Worldwide (b) Indian states

(a) Country	Uranium conc. (µg/L)	References
France	0.18-37.2	(UNSCEAR, 2000)
Ontario, Canada	0.05-4.21	(OMEE, 1996)
Egypt	1-519.4	(Dabous et al., 2002)
Brazil	0.2-667	(Almeida et al., 2004)
Korea	0.1-402.3	(Kim et al., 2004)
USA	1.8-7780	(Orloff et al., 2004)
Australia	0.05-160	(Landstetter and Katzlberger, 2009)
USA	2.9-4546	(Warner et al., 2011)

(b)Study area	Uranium conc. (µg/L)	References
Nalgonda, Andra Pradesh	0.2-68	(Brindha et al., 2011)
Bhnduhurang, Jharkhand	<0.5-27.5	(Giri et al., 2011)
Hisar, Haryana	5.3-113.5	(Garg et al., 2014)
Southwest, Punjab	0.5-579	(Bajwa et al., 2015)
Bangalore	0.24-770.1	(Nagaiah et al., 2013)
Eastern, Haryana	9.1-155.1	(Daulta et al., 2017)
Sonipat, Haryana	1.07-40.25	(Panghal et al., 2017)
Chhattisgarh	(PRM)1.15-83.5 (POM) 0.68-96.08	(Sahu et al., 2020b)
Jaipur & Dausa, Rajasthan	5-145	(Pant et al., 2019)
Beed, Maharashtra	0.03-32.85	(Kale et al., 2020)
Gurudaspur,Punjab	0.01-38.1	(Sharma et al., 2019)
Tamilnadu	0.2-14.9	(Raja et al., 2021)
Pithoragarh, Uttarakhand	(PRM) 0.1-9.90 (POM) 0.1-8.32	(Patni et al., 2020)
Assam	(PRM) 0.3-7.1 (POM) 0.6-10.3	(Saikia et al., 2021)
Kerela	(PRM) 0.5-12.54 (POM) 0.5-5.93	(Shalumon et al., 2021)
Tonk, Rajasthan	(PRM) 0.21 to 173.72 (POM) 0.21- 162.34	Present study

some of the districts tehsils of Ajmer, Bhilwara, Jaipur, Rajasamand, and Nagaur, Uranium levels were greater than 60 µg/L were found. In May 2019, 17 samples were taken from various villages in Tonk, with concentrations ranging from 0.517 to 59.484 µg/L. We conducted our study in Tonk in the same year and covered the entire district.

In the 2020-2021 Groundwater Report (CGWB, 2021) Uranium was analysed for 643 samples by LED fluorimeter. As per the report, out of 33 districts, 22 districts had Uranium levels found to be higher than 30 µg/L. In May 2020, 15 samples taken from Tonk were examined. Out of them samples four samples have Uranium concentrations higher than 30µg/L.

Water Quality parameters, condition and calculation

The maximum, minimum, and average values of Uranium and measured physicochemical parameters of the drinking water samples, standard deviation, and skewness of Tonk district were taken into account (Table 2). It compares Uranium concentration and other hydrochemical parameters of present work with BIS(BIS, 2012) and WHO standards (WHO, 2011). A number of samples above the permissible limits are also mentioned. 37% of the collected drinking water samples were from the area where the water is supplied by Bilaspur dam. For each of these samples Uranium and other water quality parameters obtained are similar.

Most samples had pH values between the WHO's acceptable range of 6.5 and 8.5. PRM pH ranges from 6.06 to 8.85 with an average of 7.62, while POM pH ranges from 5.80 to 8.42 with an average of 7.24. TDS (Total Dissolved Solid in water which includes, salts, minerals and metals) is the qualitative concentration of anions and cations and has a significant relationship with conductivity. TDS varied from 315 to 5990 mg/L, having an average of 957.54 mg/L in the PRM period, and from 269 to 6330 mg/L having an average of 910.28 mg/L in the POM period. The electrical conductivity (EC) for PRM ranges from 364 to 8535 µS cm^{-1} and 378 to 8998 µS cm^{-1} for POM with average values of 1404.13 µS cm^{-1} and 1295.57 µS cm^{-1} respectively. TDS and EC are correlated by a factor of 0.70 in both seasons.

ORP (Oxidation Reduction Potential) of samples ranges from (-283) to 194 mV in PRM and (-103) to 196 mV in POM season and no significant correlation is seen with Uranium. DO (Dissolved Oxygen) has an essential role in biological processes, directly and indirectly. A certain amount of space is occupied by dissolved oxygen in water. If the levels of dissolved oxygen are high, then water is not capable of holding other dissolved substances. On the other hand, if the level of dissolved oxygen is low, the minerals from runoff will start dissolving into the water. Average range of DO is found to be 4.34 mg/L and 4.98 mg/L in PRM and POM, respectively.

In the present study, most of the samples has fluoride are within the limits but in some places, the POM groundwater has high fluoride concentration. The fluoride content varies from 0.017 to 1.31 mg/L and 0.04 to 1.33 mg/L with a mean of 0.57 mg/L and 0.68 mg/L in pre- and post-monsoon respectively. Chloride is found to be 72.47 mg/L in the (Bhojpuri hamlet) in Uniara Tehsil to 1419.29 mg/L (at Motooka) in Tonk tehsil in PRM. In POM, it ranged from 57.48 mg/L at Lamba Harisingh in the Malpura Tehsil to 1419.43 mg/L (at Motooka hamlet) in Tonk. The amount of nitrate in PRM is found to be 0.92 to 69.4 mg/L (at

Mootoka village) and in POM it is 2.77 mg/L in the Malpura Tehsil (Amli village) to 47.98 mg/L in the Uniara Tehsil (Payga hamlet) of the Tonk district. Approximately 3 % of samples exceed the permitted limit of 45 mg/L in both the seasons. The sulphate ions range from 2.21 to 289.4 mg/L and 3.51 to 162.6 mg/L with an average of 42.38 mg/L and 38.26 mg/L in PRM and POM, respectively. Most of the samples in the study region have low phosphate concentrations, but some areas may have high phosphate concentrations as a result of anthropogenic activity such as the use of phosphate containing fertilizers in agriculture. The amount of phosphate varied from 0.001 to 0.22 and 0.003 to 0.29 mg/L in pre- and post-monsoon season respectively.

Water hardness results due to the presence of minerals such as Magnesium and Calcium in

Table 2. Descriptive statistical analysis of Uranium and associated water quality parameters of groundwater of Tonk district during PRM &POM

Water quality parameters		PRM(N=159)				Range		POM(N=159)			Range	
	BIS (2012) (Acceptable limit)	Min	Max	Average	Std. dev	Skewness	No of samples above limits	Min	Max	Average	Std. dev	Skewness
pH	**6.5-8.5**	6.06	8.85	7.62	0.64	-0.6	1	5.8	8.42	7.24	0.69	-0.26
TDS	**500**	315	5990	957.54	895.9	2.55	94	269	6330	910.28	907.48	2.59
EC	**300**	364	8535	1404.13	1348.2	2.34	159	378	8998	1295.57	1279.33	2.64
ORP		-283	194	105.3	64.4	-2.06		-103	196	96.35	53.98	-1.84
Temp.		25.8	35.3	29.5	1.71	0.53		23.4	31.5	25.68	2.21	0.33
Salinity		226	4870	683.4	700	2.79		183	5423	653.03	713.10	3.08
DO		1.48	5.81	4.34	1.31	-0.16		2.45	6.78	4.98	1.00	-0.74
F^-	**1**	$<0.017^{\$}$	1.31	0.57	0.30	0.61	18	0.04	1.33	0.68	0.27	0.53
Cl^-	**250**	72.47	1419.3	191.66	197.83	3.56	28	57.48	1419.4	187.52	169.24	3.88
NO_3^-	**45**	$<0.92^{\$}$	69.4	10.95	12.8	1.98	6	2.77	47.98	18.88	12.99	0.81
SO_4^{-2}	**200**	$<2.21^{\$}$	289.4	42.38	54.47	2.33	6	3.51	162.6	38.26	33.74	1.45
PO_4^{-3}		$<0.001^{\$}$	0.22	0.05	0.03	0.77	-	0.003	0.29	0.11	0.06	-0.32
U	**$30^{a,b}$**	0.21	173.72	8.35	18.42	6.04	**6**	0.21	162.34	11.22	21.68	4.29
TH	**200**	115	3300	323.76	319.34	5.7	85	105	3420	319.15	340.20	5.33
Ca	**75**	24	762.0	95.60	86.3	3.79	67	24	950	85.38	89.03	6.19
Mg	**30**	2.43	339.4	20.38	31.9	7.09	27	4.87	254.26	25.65	30.70	3.58
TA	**200**	130	1735	339.28	252.6	2.57	93	90	1685	273.58	204.44	3.06
CO_3^{-2}		0	60	1.38	7.42	6.18		0	40	0.44	3.95	9.10
HCO_3^-		130	1735	337.89	252.8	2.58		90	1685	273.14	204.08	3.07

Electrical conductivity (EC), µS/cm; Total dissolved solids (TDS), Salinity, Total hardness (TH), Total alkalinity (TA), Nitrate, Sulphate, Phosphate, Ca, Mg, Cl, F, mg/L; Oxidation reduction potential (ORP), mV; Uranium, µg/L ; [$]BDL (Below detection Limit) [a](WHO, 2011) [b](BIS, 2021)

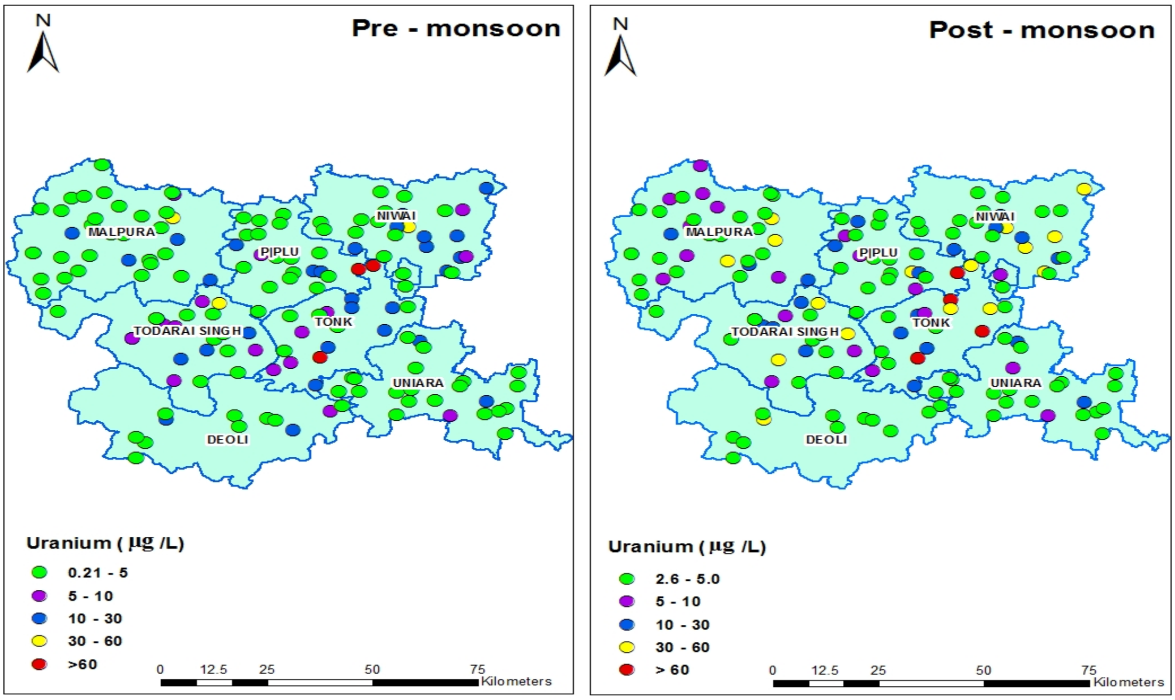

Fig. 2. Uranium distribution in pre- and post-monsoon seasons in Tonk district

the form of chlorides, bicarbonates, carbonates, and sulphates. The Calcium was found in the range 24 to 762.0 mg/L with an average of 95.60 mg/L in PRM and from 24 to 950.0 mg/L with an average of 85.38 mg/L in POM. Examining the average values in POM we can conclude that the dilution brought by the rainfall may be the causal factor in the drop of Ca^{2+} content in the post-monsoon season. Magnesium concentration is found in the range of 2.43 to 339.4 mg/L with an average of 20.38 mg/L in PRM and in POM, Magnesium ranges from 4.87 to 254.26 mg/L with an average of 25.65 mg/L. As per the data analysis, the Ca^{2+}/Mg^{2+} molar ratio is more than 1, with an average of 4.24 mmol/L in PRM and 2.72 mmol/L in POM season. This indicates that a high concentration of the calcite mineral was present in the area of study. It is observed that in Mootoka village of Tonk tehsil, maximum hardness with the range of 3300 and 3420 mg/L is found in pre- and post-monsoon samples respectively. Approximately 50% of samples are under hard category in both seasons.

The total alkalinity of water reflects the buffer capacity and it is mainly due to the amount of bicarbonate, carbonate and hydroxide present in it. The bicarbonate ion was found in the range 130 to 1735 mg/L and 90 to 1685 mg/L with mean values of 337.89 mg/L and 273.14 mg/L, in pre-and post-monsoon respectively. The alkalinity is over the 200 mg/L threshold. Most of the samples had significant levels of bicarbonate ion but very low level of carbonate, probably because of carbonate and silicate weathering.

The skewness coefficient of Uranium distribution is 6.04 in pre-monsoon and 4.29 in post-monsoon indicating that its distribution is asymmetric and positively skewed. The tehsil-wise summary of Uranium is given in Table S1. The highest Uranium concentration is reported in Tonk tehsil and the lowest in Uniara tehsil.

Uranium distribution

The spatial distribution of Uranium in PRM and POM seasons is presented in the schematic map (Figure 2). The highest Uranium concentration 173.72 µg/L has been found in Motooka village of Tonk tehsil in PRM and in POM at Nayagaon of Tonk with 162.34 µg/L. In POM period source of water is different from PRM, at Nayagaon village. From the distribution of data, it was found that 66.66%, 11.32%, and 18.23% of Uranium samples lie in the range of 0.1 to 5 µg/L, 5 to 10 µg/L, 10 to30 µg/L in PRM respectively, whereas in POM, 61.63%, 25.78% samples lied in the range of 0.1 to 5 µg/L and 5 to 30 µg/L respectively. Overall, 3.77% (6 samples) in pre-monsoon and 12.57 % (20 samples) in the post-monsoon season had Uranium concentration above 30 µg/L, the permissible limit. From the data, we found that the mean and median of Uranium concentration is substantially low than the recommended level of BIS, in both seasons (BIS, 2021).

Uranium concentration and physico-chemical parameters correlation

High alkalinity and hardness were observed in most of the analyzed samples. The bicarbonate ions are responsible for the alkalinity, as well as the formation of highly soluble uranyl-carbonate (Katsoyiannis IA, 2007). This increases Uranium content in the water samples. The migration of Uranium is affected by surface and groundwater chemistry.

The order of average anionic concentration in the study area in both seasons was found to be HCO_3^- > Cl^- > SO_4^{2-} > NO_3^-, which suggests water-rock interaction and mild acidic conditions. The correlation matrices for 15 variables i.e., Uranium, pH, EC, Salinity, ORP, DO, TDS, TA, TH, F^-, Cl^-, NO_3^-, SO_4^{2-}, PO_4^{3-}, Ca^{2+} and Mg^{2+}, are summarized in Table S2 and S3 for PRM and POM which exhibit moderate to strong correlation.

From the Pearson correlation matrix, a moderate to strong correlation between Uranium and hydro-chemical parameters is found in the study area. It is evident that there is a considerable correlation of Uranium with TDS (0.57), TH (0.70), HCO_3^- (0.43), Cl^- (0.55), SO_4^{2-} (0.62), and NO_3^- (0.56) in pre-monsoon season (Figure 3). The moderate positive correlation with bicarbonate indicates the uranyl-carbonate complex formation in aquifers (Cho and Choo, 2019). In the POM correlation matrix, a strong positive correlation can be seen between Uranium and other geochemical parameters such as TDS (0.63), TH (0.58), HCO_3^- (0.60), Cl^- (0.59) whereas moderate correlation is found with SO_4^{2-} (0.29) and NO_3^- (0.48). A negative correlation is associated with pH and ORP in both seasons.

Hydrogeochemical mechanism

To understand the hydrogeochemical mechanism, Gibbs (Gibbs, 1970) classified

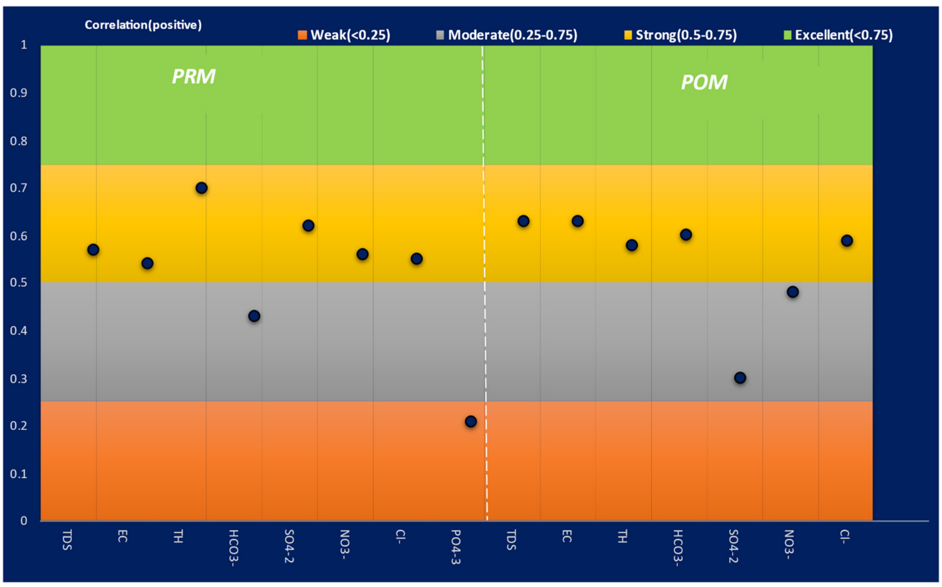

Fig. 3. Correlation of Uranium with other water parameters in pre-& post-monsoon seasons

groundwater into three dominant zones viz atmospheric-precipitation, evaporation-dominance, and water-rock interaction, based on TDS and ion ratio of groundwater. Gibbs ratio 1 is plotted (for samples where Uranium concentration >5µg/L) between TDS and anions ($Cl^-/Cl^-+HCO_3^-$), indicating the geochemistry of the aquifer controlled mainly by rock-water interaction and evaporation in both seasons of the study area (Figure 4). During its flow, groundwater interacts with the aquifer matrix, and this exchange of ionic components is responsible for the dissolved ions in groundwater. It seems that precipitation did not play a substantial role in governing hydrochemistry (Figure 4). Few samples fall in the evaporation dominance zone. As the sampling was done in the summer, the high temperature (28 to 43°C) may have triggered the evaporation process, leading to the observed rise in the concentration of various ionic species in the groundwater (Sharma et al., 2021). Samples falling in evaporation zone could be due to the presence of chloride ion, as it is a dominant anion next to the bicarbonate, in the study area indicates presence of high concentration of Na-Cl in water. Human activities like irrigation return flow and industrial pollution could be the major reasons for enhancing evaporation processes in the underground water (Kumar and Ekanthalu, 2020).

The bivariate plot of $Ca^{2+} + Mg^{2+}$ versus $HCO_3^- + SO_4^{2-}$ (Figure 5a) shows the majority of samples lie above the median line (Masoud et al., 2018). This explains the excess of $Ca^{2+} + Mg^{2+}$ over $HCO_3^- + SO_4^{2-}$ in water may be due to the dissolution of calcite minerals and carbonate weathering in both pre-& post-monsoon. The molar ratio plot (Figure 5b) of Calcium and magnesium also confirms the above fact. In the pre-monsoon period, silicate weathering is dominant over calcite and dolomite dissolution, whereas in post-monsoon most of the samples lie in the region of calcite and silicate weathering over dolomite dissolution (Brindha, K., Paul, R., Walter, 2020; Ghesquière et al., 2015).

Seasonal change in Uranium concentration in study area

The study of seasonal variation assists in identifying the factors contributing to high or low concentrations of specific ions and Uranium in groundwater. Seasonal variations in Uranium concentration showed irregular trends. According to CGWB 2019-2020 (CGWB, 2020), Tonk received 30.68% more rainfall than the normal. The depth of water level in district is reported in the range of 2.50 - 36.45 mbgl in PRM and 0.65 to 34.15 mbgl in POM seasons (Ground Water Department, 2020). Adequate rainfall recharges the groundwater level and Uranium is

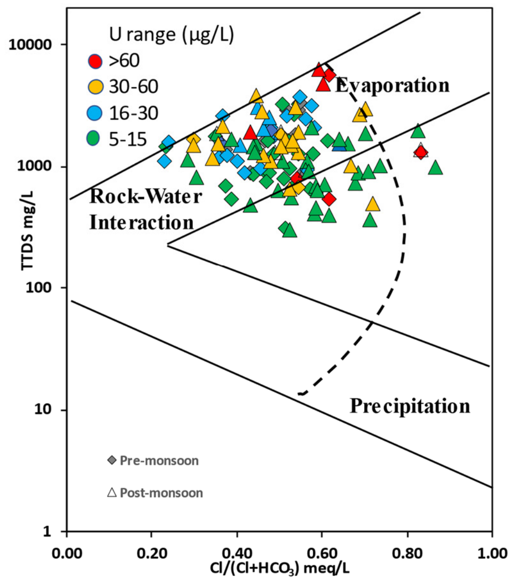

Fig. 4. Gibbs plot showing rock water interaction

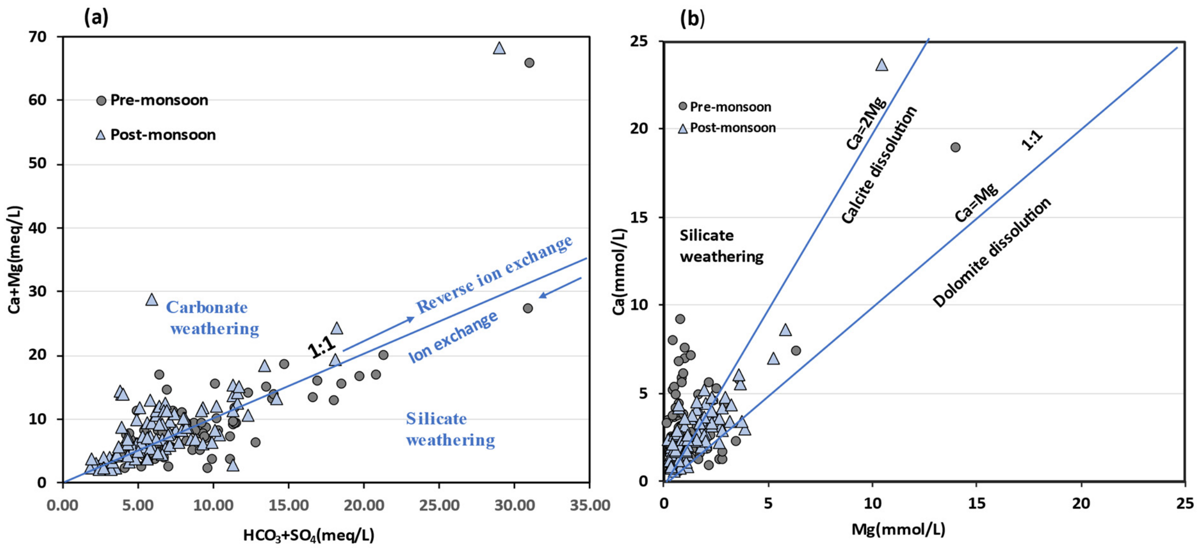

Fig. 5a-5b. Bivariate plot explaining weathering and dissolution process

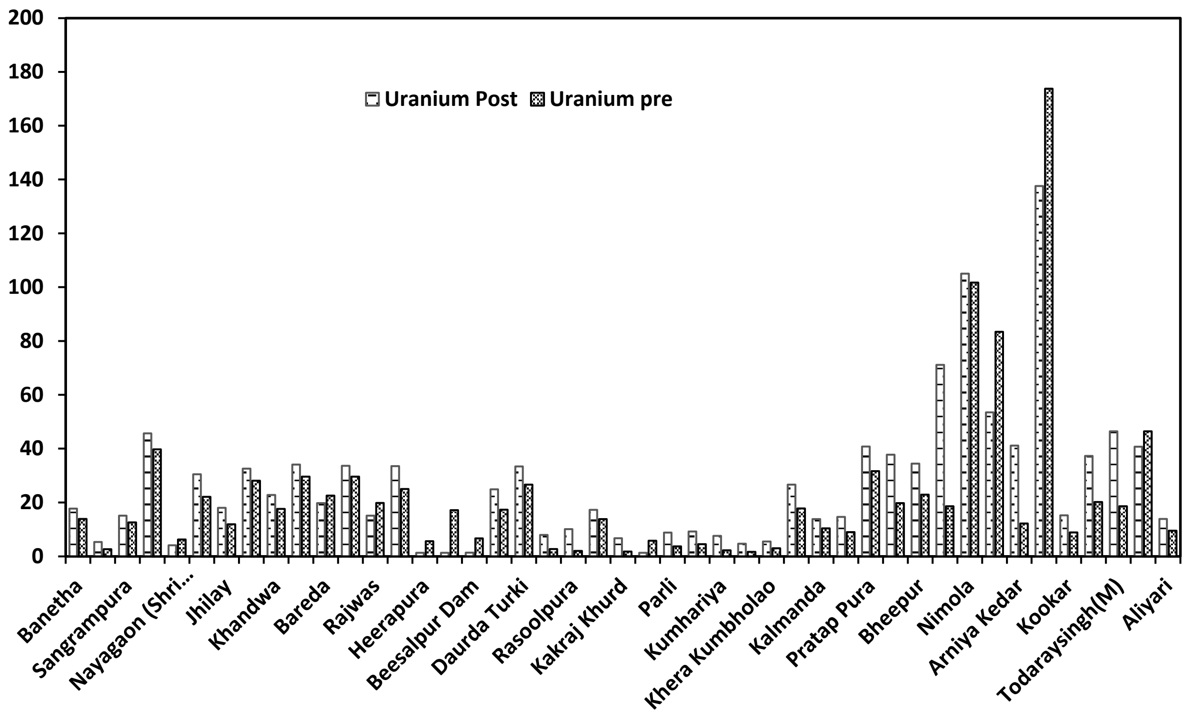

Fig. 6. Seasonal variation in Uranium distribution of Tonk in POM and PRM

prone to leach out**.** However, if recharge continues the concentration of Uranium in ground water may decrease owing to the dilution (Sahu et al., 2020a).

In most of our samples, high Uranium concentration is observed in POM as compared to PRM (Figure 6). From the correlation matrix, it has been found that bicarbonate is more positively correlated in post-monsoon as compared to pre-monsoon. Bicarbonate plays a significant role in the leaching of Uranium and thus dissolution of it in groundwater. Uranium was found to be positively correlated with Total Hardness (TH), Ca^{2+} and Mg^{2+} ions in both seasons. The presence of Ca^{2+} and Mg^{2+} ions affect Uranium speciation and its mobilization through the formation of $Ca(UO_2)(CO_3)_3$ and $Mg(UO_2)(CO_3)_3$ complexes (Fox et al., 2006).

The positive correlation of Uranium with Cl^-, NO_3^- and SO_4^{2-} is observed in both seasons which indicate that these anions form soluble complexes with uranyl cation in groundwater and prevent its adsorption on the sedimentary matrices. A significant correlation was observed for Uranium and TDS which results in mineral dissolution. In POM samples, Uranium concentration > 30µg/L shows significant positive correlation with TDS, HCO_3^-, TH and chloride (Table S4). This supports the fact that bicarbonate and chloride may take part in mineral dissolution resulting in enhanced mobility of Uranium concentration in the post-monsoon period. Stronger correlation matrix of Uranium with bicarbonate in POM suggests that its presence plays a significant role in facilitating the dissolution of minerals and subsequent migration of Uranium. Yadav (2019) highlights the importance of understanding the correlation between Uranium and bicarbonate.

The bedrock is mainly covered by mica schist, which is composed of quartz and mica. Mica contains hydrous potassium, aluminium silicate minerals, and phyllosilicates. The weathering of quartz may cause mineral dissolution. The groundwater in the area of study is rich in Cl^-, SO_4^{2-}, and NO_3^- and has high Calcium content as compared to Magnesium, evident from the geochemical mechanism. When CO_2 dissolves in underground water it forms carbonic acid. This carbonic acid reacts with calcareous soil and produces bicarbonates which act as a good leaching agent for Uranium (Bajwa et al., 2015).

The Uranium distribution in PRM and POM seasons (of the study area) is shown in the Box-whisker plot for seven tehsils (Figure 7a-7b). The first to third quartiles are shown in the central rectangle (the interquartile range). The median is depicted by a segment inside the rectangle, and the minimum and maximum points are indicated by whiskers above and below the box. The points above whisker are called outliers. The highest Uranium concentration was observed in Tonk tehsil and the minimum in Uniara tehsil in both seasons. A longer whisker in the positive direction and a greater mean than the median indicate that the Uranium distribution is positively skewed in Tonk and Niwai in both seasons. Mean value is larger than the median in every tehsil in both seasons. Tonk, Piplu and Deoli have far outlier values as compared to others. From box plot it is clear that average Uranium in PRM and POM is 29 µg/L and 41 µg/L respectively in Tonk tehsil. In Deoli, median and Q1 are equal in post-monsoon. In Piplu, Malpura, and Uniara, mean, Q1 and Q3 values are quite close to each other in PRM season.

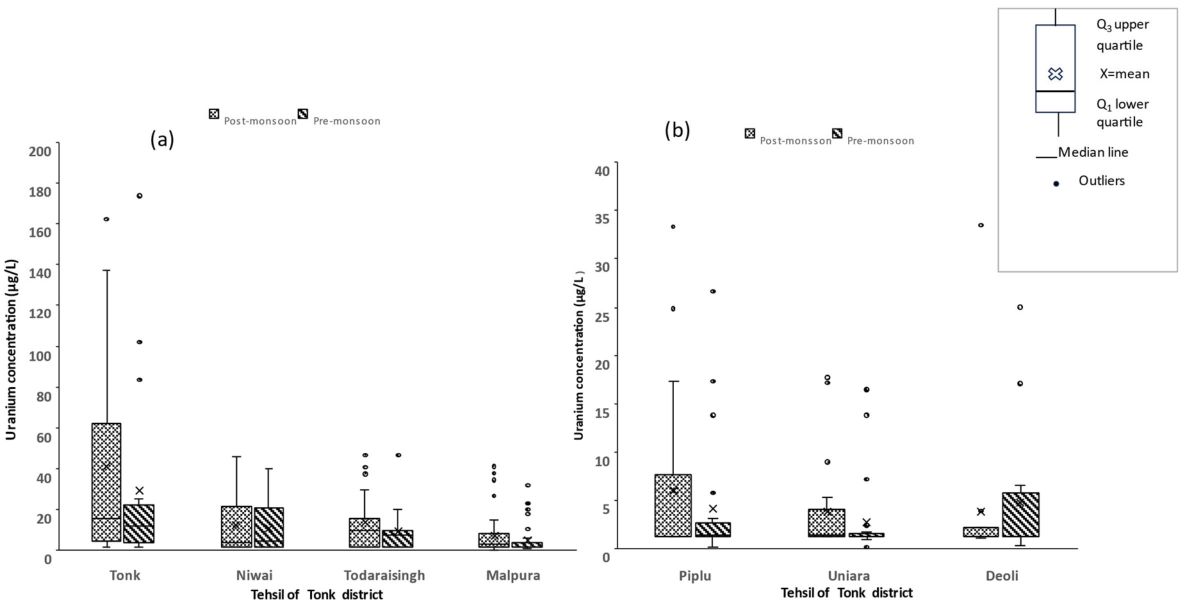

Fig. 7a -7b. Box plot of Uranium concentration in seven Tehsil of Tonk district in pre-& post-monsoon

The smallest whisker with the lowest median in Uniara and Deoli tehsil exhibits the least scattered distribution of Uranium in both seasons. In Uniara and Deoli lower quartile is equal to the minimum and the upper quartile is quite close to the maximum and the whisker is small reveals that Uranium concentrations are nearly in the same range. This is because approximate 50 % of collected samples from these two tehsils have the same supply of drinking water i.e from Bisalpur dam.

CONCLUSION

In total 318 drinking water samples were collected and analyzed in Tonk district during PRM and POM seasons. The heterogenous distribution of Uranium is found in drinking water sources in study region. The average Uranium concentration is found below the BIS/WHO limit in Tonk. This suggests that geochemical parameters influence and control the mobility of Uranium in groundwater. According to Uranium limit (BIS, 2021; WHO, 2011) 3.77 % of pre- and 12.57 % of samples in the POM of Tonk district exceeds the limit of 30 µg/L. Out of seven tehsils of Tonk district, major seasonal impact can be seen in Tonk tehsil. From the analysis of the results, it was observed that the concentration of Uranium is higher during the POM as compared to the PRM. This could be attributed to the fact that Uranium gets dissolved from rocks due to the input from rock-water dominance, which enhances the mobility of Uranium in groundwater. Furthermore, the decrease in Uranium concentration at certain locations during the post-monsoon season could be due to the dilution caused by rainwater. The hydrochemistry of the study area can be explained by the chemical interaction of groundwater with aquifer rock and the evaporation-sedimentation process. This is supported by high TDS and conductivity. The average concentration of anions is found to be in the following order $HCO_3^- > Cl^- > SO_4^{2-} > NO_3^-$ (water rock interaction). Phenolphthalein alkalinity is almost zero, carbonate alkalinity is quite low and bicarbonate ions are abundant. Calcium rich salts (Calcite) are found for most of the samples i.e., $Ca^{2+} > Mg^{2+}$, and certain areas $Mg^{2+} > Ca^{2+}$ due to Mg-rich dolomite. For the molar ratio of ions Ca/Mg, it is found that Calcium has a greater contribution to calcite minerals and proves the dominance of carbonate and silicate weathering. From the study finally we conclude that Tonk, Niwai, and Todaraisingh tehsils have a greater number of samples with high Uranium concentration in potable water as compared to Malpura, Piplu, Uniara and Deoli tehsils. Therefore, continuous monitoring of Uranium concentration of drinking water source in hot spot area must be done seasonally. This study will provide valuable insights into the potential impact of Uranium on the scientific front, allowing us to reassure society about its safety. Furthermore, preventative or corrective measures can be taken to remove excess uranium from drinking water, if required.

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CONSENT FOR PUBLICATION

All of the authors consented to publish this research article.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

LIFE SCIENCE SUPPORTING

No life science threat was practiced in this research work.

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