

# Assessment of the groundwater quality by using multivariate approach and non-carcinogenic risk of uranium in the inhabitants of the Bastar district, Chhattisgarh, Central India

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## ABSTRACT

The elevated levels of uranium found in 17 states of India is alarming due to the radionuclide contamination in groundwater. Chronic ingestion can cause potential harm to humans and living things such as damage to kidneys, and cancer of the liver, lungs, and bones. The present study was undertaken to assess groundwater quality using a multivariate approach to the non-carcinogenic exposure of uranium by residents of the Bastar district, Chhattisgarh. The concentration of uranium in groundwater samples ranged from 0.50–26.4 µg/l in 70 samples, with 82% of samples being beyond the recommended limits by the International Commission on Radiological Protection. Hierarchical cluster analysis divided all sampling locations into 10 clusters explaining the similarity of geological conditions. Factor analysis extracted four principal components or factors with 70.20% cumulative variance from the entire data set. Chronic daily intake has been found above from the reference dose as 34.29, 42.86, and 51.43% for young children, children and adults. The results of hazard quotient analysis classified the degree of non-carcinogenic risk which was > 1 in 34.28, 45.71, and 41.43% for the samples from young children, children and adults, respectively. This study will generate baseline data and suggest the need for revision of water quality monitoring plans and preventive water management practices.

**Key words:** chronic daily intake, cluster analysis, factor analysis, hazard quotient, risk assessment

## HIGHLIGHTS

- The study was carried out in the Bastar division of Chhattisgarh, India.
- The concentration of uranium was found in the range from 0.50 to 26.40 µg/L.
- In total, 58 groundwater samples were found to contain higher concentrations of uranium than the ICRP recommended standard (1.9 µg/L).
- The chronic daily intake of uranium was observed to be higher in 24 samples for infants, 30 samples for children, and 36 samples for adults.

## 1. INTRODUCTION

Uranium is a techno-important radionuclide due to its radiological and chemical properties, and is found naturally in groundwater. Uranium dominates the Earth setting in all areas of the environment such as the hydrosphere, lithosphere, atmosphere and biosphere (Duggal *et al.* 2017). Water chemistry plays an important role in the dissolution of uranium in groundwater. The mobility of uranium depends on an aqueous geochemical reaction; it is highly soluble in alkaline, carbonate-containing water under oxidizing conditions, whereas it is less soluble in water under reducing and acidic conditions (Cho & Choo 2019). Uranium is a radioactive metal that decays through alpha particle emission and contributes significantly after ingestion inside the body. <sup>238</sup>U, <sup>235</sup>U and <sup>234</sup>U are isotopes commonly found in natural environments, while both <sup>233</sup>U and <sup>236</sup>U are the results of anthropogenic activities. <sup>238</sup>U contributes a maximum of 99.27% to the natural environment, while this is 0.72 and 0.0054% for <sup>235</sup>U and <sup>234</sup>U (Smedley *et al.* 2006; Kumar *et al.* 2018a, 2018b). Its half-life is very long, which leads to bioaccumulation in all environmental matrices. The bioavailability of uranium depends on complexing by inorganic and organic ligands, pH, absorption by soil and others minerals, hydroxide, interactions with organic matter, oxidation state etc. The +4

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and +6 oxidation states of uranium are most soluble in an aqueous environment and form stable complexes with various ligands (Choppin *et al.* 2002; Coyte *et al.* 2018). The distribution of U(IV) is widespread in coffinite, uraninite and pitchblende uranium ores released under a reducing environment (Sar *et al.* 2017). U(IV) dissolves in the aqueous environment and forms complexes with inorganic ligands (uranyl phosphate and carbonate) and humic substances (uranyl humate and uranyl fulvate) (Choppin *et al.* 2002).

Surplus amounts of uranium are found in groundwater in different parts of the world due to natural and anthropogenic activities. Mining of uranium ore, nuclear testing, nuclear disposal site, phosphate fertilizer and laboratory uses are the largest sources of anthropogenic contamination of uranium in the environmental matrix (Alam & Cheng 2014; Nolan & Weber 2015; Guo *et al.* 2016; Kumar *et al.* 2020a, 2020b). Natural geochemical processes such as the dissolution of minerals and the desorption of adsorbed uranium from the surface of minerals can increase the uranium contamination in groundwater, which has dual effects on the human body due to its radioactive and chemical properties. The World Health Organization (2011) and the United State Environmental Protection Agency (2011) set the guideline limit for maximum contamination levels of uranium in potable water at 30 µg/L. Ingestion, dermal adsorption and inhalation are common routes of entry for radionuclides into the human body (Arogunjo *et al.* 2009). Uranium is also transferred to plants through the root system as nutrients and minerals, as well as appearing in soil and irrigation water during growth and metabolic activity (Asaduzzaman *et al.* 2015).

The absorption of ingested uranium into the blood varies in the range of 0.1% to 6% (Kumar *et al.* 2018a, 2018b) and is deposited in the liver, kidneys and bones with the kidneys showing the highest chemical toxicity. The bone is the second most cited target of chemical poisoning in the human body. The absorption contribution of uranium in drinking water (85%) is higher than in other dietary or food ingredients (15%). Prolonged exposure to uranium poses a potential risk to human health (Thorne 2020). The chemical or carcinogenic health risk of uranium is more harmful than the radiological characteristics. Nephrotoxic effects and chronic kidney disease are both associated with uranium-enriched drinking water; this was investigated in toxicological and epidemiological studies (Coyte *et al.* 2018). Chemical poisoning of uranium also causes damage to the reproductive systems and liver. Biologically dynamic toxicity, chemical toxicity and metabolism toxicity are several established health consequences of excessive ingested uranium, in addition to it being noted for potential damage to gene expression, brain, and abnormal fetal and embryonic development and reproduction (Kaur & Mehra 2019). The Atomic Energy Commission recommendation of a total daily intake 0.6 µg/kg/day of body weight with 100 as the uncertainty factor for calculating the low observational adverse effect level (LOAEL) of 60 µg/kg of body weight per day the intake or reference dose was taken. Several studies on uranium contamination in potable water have been conducted in relation to a risk assessment in different parts of India including Bihar (Kumar *et al.* 2020a, 2020b), Himachal Pradesh (Kaur & Mehra 2019), Punjab (Bajwa *et al.* 2017), Hyderabad (Balbudhe *et al.* 2012), Rajasthan (Jakhu *et al.* 2016), Haryana (Kansal *et al.* 2011), Jammu and Kashmir (Kumar *et al.* 2016), Jharkhand (Rana *et al.* 2010), as well as worldwide in Germany (Del Carmen Lamas 2005), USA (Nolan & Weber 2015), Korea (Cho & Choo 2019), Malaysia (Asaduzzaman *et al.* 2015), Nigeria (Arogunjo *et al.* 2009), the United Arab Emirates (Xiong *et al.* 2020), Canada (Chen 2018), and Australia (Priestley *et al.* 2018).

A review of the literature shows that uranium is widely available for exposure doses in different districts of the state of Chhattisgarh such as Durg (Sar *et al.* 2017), Bijapur (Singh *et al.* 2021), Balod (Sar *et al.* 2017), Bemetara (Sahu *et al.* 2019), Kanker (Sahu *et al.* 2020), but data are lacking or no such research has been conducted in Bastar district of Chhattisgarh state (India).

In recent years, many researchers have used principal component analysis (PCA) or factor analysis (FA) for understanding and grouping water quality parameters (Kumar *et al.* 2020a, 2020b). Gajbhiye *et al.* (2015) used a PCA approach on two different sources of municipal waste (Moti Nala and Urdana Nala) in Jabalpur, India. Sahu *et al.* (2018) also used this method to identify seasonal variations in fluoride-contaminated groundwater quality of Lalganj tahsil, Raebareli district Uttar Pradesh (UP), India. According to environmental analysts, the FA is more reliable than other statistical methods which has no assumptions (Sahu *et al.* 2018). The purpose of FA is to reduce the number of variables to a manageable number of indices while maintaining relationships between real data sets.

The Bastar district was selected for this study because 70% of the population belong to tribal communities and education percentage was very low (46% as per the 2011 census). Here, 80% of disease originated from ingestion of contaminated water and water quality monitoring is the first step of the water management plan. The main objective of the proposed study was to evaluate groundwater quality with a multivariate approach and to assess the risk exposure to the chemical toxicity of uranium

through ingestion of potable water by residents of Bastar district, Chhattisgarh (India). This study presented the physico-chemical parameters as pH, alkalinity, total dissolved solids (TDS), hardness,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{PO}_4^{3-}$  etc., with special reference to the multivariate approach, uranium concentration, correlation and non-carcinogenic risk of uranium and statistical approaches in the data set. The present investigation will generate baseline data for the study area, which will help the policy maker of the state for make sound decisions on managing water quality in the present and future.

## 2. MATERIALS AND METHODS

### 2.1. Study area

Bastar is a district located in the central Indian state of Chhattisgarh as presented in [Figure 1](#). It is situated between 19.5676 degrees north and 81.6912 degrees east. Jagdalpur is the district headquarters of Bastar and is surrounded by Nabarangpur and Koraput districts of Odisha state to the east, Narayanpur district to the northwest, Kondagaon district to the north and Dantewada and Sukma to the south. According to the census report, the total population of the district is 8,34,873 people, living in a total area of 6,597 square kilometers. Geologically, the district is covered with gneiss, granite, meta-sedimentary, basalt and gabbroic rocks and the landscape is dominated by dense forest, with half the district being mountainous and rocky. The soil of the district has widely varied such as loam, alfisol, red gravel and red sandy, these are the soils that cover most of the area. The Indravati river system, along with the Markandi and Narangi rivers, serves as the main drainage system of the district. The Indravati River and the Sabari River, both tributaries of the Godavari River, cover about 97% of the area in this region. The average annual rainfall of the district is 1,386.77 mm. The annual temperature ranges from 10.6 °C in the winter to 46 °C in summer. The relative humidity ranges from 90% in the wet season to 30% to 40% in the winter season.

### 2.2. Sample collection

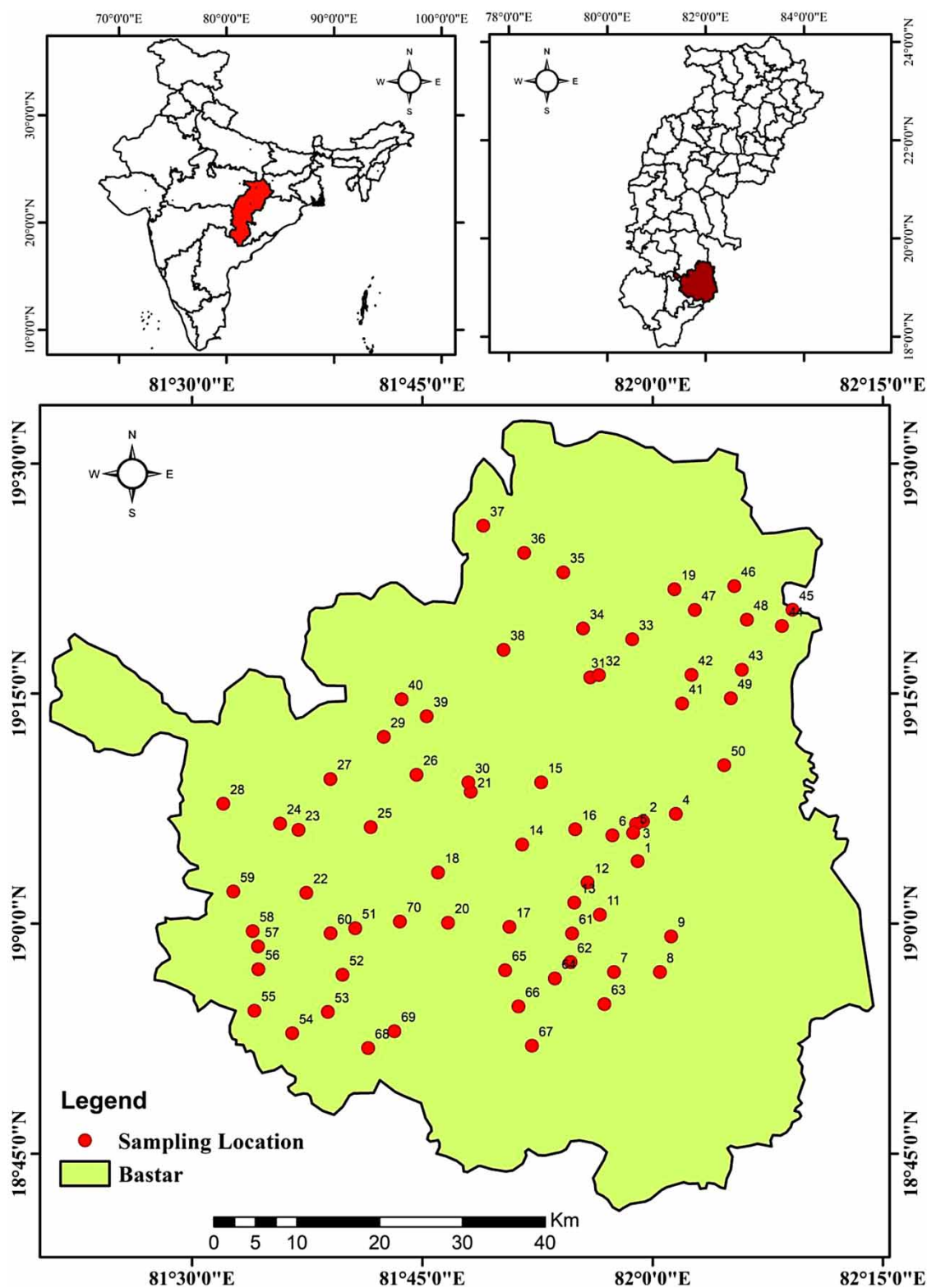
Seventy groundwater samples were taken from hand pumps at different places in Bastar district and sampling was carried out during the period of April 2019, water samples were drawn for drinking purposes by the local population. All samples were collected in pre-washed polypropylene bottles after 5 min of hand pumping and washing, three times from the respective samples. The polypropylene bottles are air-tight laboratory grade in nature, with a 2 liter capacity, that have been infused with 10% nitric acid overnight to eliminate grime on the inner surface of the container. A 0.45- $\mu\text{m}$  Whatman filter paper was used to filter all water samples prior to analysis. APHA (American Public Health Association) standard protocols were followed for the special storage, handling and transportation of water samples. ([APHA 2005](#))

### 2.3. Analysis of physico-chemical parameters

For the purpose of analysis, all water samples were brought to the Department of Chemistry, National Institute of Technology, Raipur, India. Seventeen water quality parameters were examined, most in accordance with a series of World Health Organization ([WHO 2008](#)) and Bureau of Indian Standards (IS 3025) drinking water analysis manuals. *In situ* parameters such as TDS, EC (electrical conductivity), pH, temperature, oxidation–reduction potential (ORP), salinity, and DO (dissolved oxygen) were tested using the Hanna instrument (HI 98194) at the time of sampling. The total hardness in water samples with calcium hardness as  $\text{CaCO}_3$  was calculated via the ethylenediaminetetraacetic acid (EDTA) complexometric method accomplished using Eriochrome Black T and Murexide indicators, respectively. Further magnesium hardness was calculated by subtracting the calcium hardness from the total hardness. A standardized solution of 0.02 N of  $\text{H}_2\text{SO}_4$  and a methyl orange indicator were used to estimate the total alkalinity in the water sample, and Mohr's titration method was used to calculate the chloride ions in the groundwater sample. The technique used for nitrate analysis was a ultraviolet-visible (UV-vis) light spectrophotometer (UV-1800, Shimadzu, Kyoto, Japan) using 1 N HCl at 220 and 275 nm. Conditioning reagent ( $\text{NaCl} + \text{HCl} + \text{alcohol} + \text{glycerol}$ ) and  $\text{BaCl}_2$  were used to determine the amount of sulphate in the sample. The absorbance of the turbid solution was measured after 2–3 minutes at 420 nm wavelength. For phosphate measurement, color was developed by addition of 10% ascorbic acid, and mixed reagent i.e. 125 ml 10% ammonium molybdate + 9 N  $\text{H}_2\text{SO}_4$  + 2.5% potassium antimony tartarate. Absorbance was taken after thorough mixing for 10 minutes after the development of a blue color at 690 nm, ([APHA 2005](#)).

### 2.4. Uranium measurement in groundwater

Uranium in drinking water was tested using an LF-2a LED fluorimeter manufactured by Quantalase Enterprises Pvt. Ltd, Indore, India. The minimum and highest detection limits of the device are 0.2  $\mu\text{g/L}$  and 1,000  $\mu\text{g/L}$ , respectively, with a  $\pm 10\%$  precision. The buffer solution sodium pyrophosphate (5%) was prepared in Milli-Q water and orthophosphoric acid



**Figure 1** | Chhattisgarh map showing Bastar district (study area).

was used to adjust drop by drop to pH 7. This solution acts as a fluorescence enhancer. The fluorescence yields for the different uranium complexes varied greatly. Therefore, a reagent, i.e. fluorescence-enhancing solution, was added to the sample to ensure a uniform conversion of fluorescence yield for all the complexes in the same manner. A 5.0 ml water sample was

placed in a cuvette and 0.5 ml of 5% sodium pyrophosphate was added, and the uranium concentration was determined using an LED fluorimeter. (Singh *et al.* 2021).

## 2.5. Quality control and quality assurance

Analytical research-grade (AR) chemicals (Merck and Sigma-Aldrich, Germany) were used throughout the research work. There was no further purification or disturbance to them. The glasswares were first cleaned with dilute nitric acid ( $\text{HNO}_3$ ) (1.15 N) and then; it was washed with distilled water. Milli-Q water was used to make all reagent and calibration standards for analysis. All analyzes were performed in triplicate. In addition, all reference and stock solutions were kept at 4 °C until they were used in the analysis. As a part of quality assurance, rigorous washing/cleaning procedures, as well as monitoring of blank levels of equipment, solvents and other items were ensured.

## 2.6. Statistical analysis

Multivariate statistical analysis such as FA, cluster analysis and spearman correlation etc. were selected to analyzed the huge datasets generated from 70 sampling locations. All the statistical analyses were run in SPSS software (version 22) to inspect the associations among multiple variables in the physico-chemical data set of the groundwater. Varimax with Kaiser Normalization method was used to interpret the major factors contributing to the decline in water quality. Cluster analysis such as the Rascal distance method was applied to determine how close two samples were and physico-chemical parameters, while the Rascal distance was defined as the difference between the transformed values of the samples. Spearman correlation analysis was performed in two ways at 0.05 and 0.01 level of  $\alpha$  to identify the correlation between physico-chemical parameters.

## 2.7. Non-carcinogenic risk assessment

Hazard quotients (HQ) indicate the level of harm caused by consuming uranium-contaminated water; HQ was used to measure chemical risk, which was calculated using Equation (1), and chronic daily intake (CDI), which was calculated using Equation (2) (Singh *et al.* 2021):

$$HQ = \frac{CDI}{RfD} \quad (1)$$

$$CDI = \frac{CDw * IR * ED * EF}{AT * BW} \quad (2)$$

where CDI refers to chronic daily intake ( $\mu\text{g}/\text{kg}/\text{day}$ ), RfD denotes the reference dose of uranium ( $0.6 \mu\text{g}/\text{kg}/\text{day}$ ), CDw = concentration of uranium in drinking water ( $\mu\text{L}$ ), IR = drinking water ingestion rate in L/day, ED = exposure duration in years for different classified age groups (4, 12, and 30 years for young children, children, and adults, respectively), EF = Exposure frequency 365 days a year, AT = average time (days) and BW = body weight in kg.

The ingestion rate and body weight were assessed through a random survey of residents in the research area. IR was 6, 3, and 1 L for adults, children, and young children respectively. BW was 60, 40, and 14 kg for the same age groups (Singh *et al.* 2021). In total, 300 inhabitants were involved individually through the random survey for the ingestion study, they did not filter/treated the water system.

## 3. RESULT AND DISCUSSION

### 3.1. Evaluation for drinking water quality

Seventeen physico-chemical parameters of groundwater collected from 70 locations of Bastar district were analyzed (Table 1). The groundwater of the study area was found to be slightly acidic in nature. The pH values of the water samples ranged from 5.2 to 8.2 with an average of  $6.74 \pm 0.08$ . This was under the prescribed drinking water standard (6.5–8.5) of the Bureau of Indian Standards (BIS 10500) and the World Health Organization (WHO 2011). According to the WHO, pH 8.5 water has no apparent effect on users (WHO 2011). Consuming drinking water containing high levels of TDS may result in gastrointestinal pain that consumers' experience (Sahu *et al.* 2018). The TDS in groundwater samples ranged from 96 to 389 mg/L, which is the TDS limit for groundwater samples and found to be below the WHO recommended value. Industrial wastewater, natural sources, sewage, urban and agricultural runoff, all contributed to the TDS in groundwater (Duggal *et al.* 2021). The groundwater temperature ranged from 27.80 to 32.80 °C and the groundwater was extracted from 80 to 330 feet below the surface. The electrical conductivity of the water samples analyzed



**Table 1** | Description of physico-chemical parameters in the groundwater of Bastar district

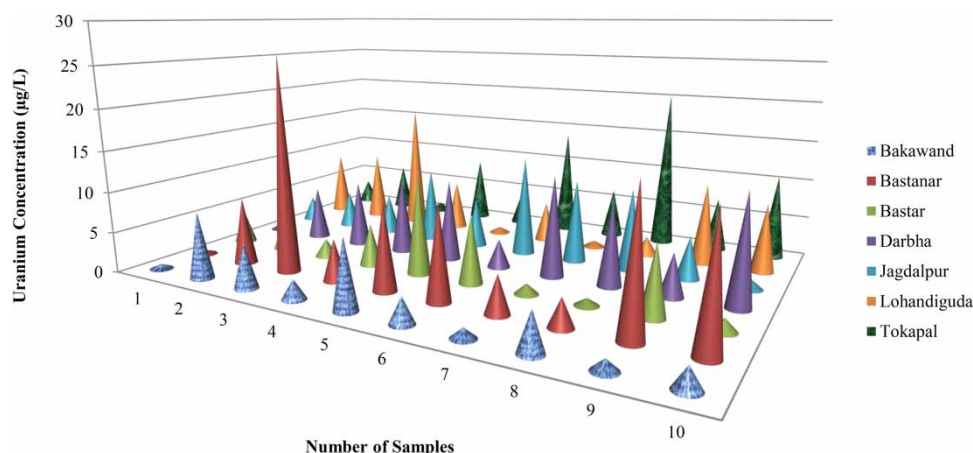
Parameters	Unit	Minimum	Maximum	Mean	SEM	BIS/WHO limits
Depth level	(Feet)	80.0	330.0	201.43	8.07	–
Gamma radiation	(nSv/h)	98.0	256.0	155.16	3.47	–
pH	–	5.20	8.20	6.74	0.08	6.5–8.5
TDS	(mg/L)	96.0	389.0	229.57	9.03	500–2,000
EC	( $\mu$ S/cm)	192.0	778.0	459.14	18.06	–
ORP	(+/-mV)	–94.6	96.1	20.61	5.37	–200 +200
Temperature	( °C)	27.8	32.8	29.85	0.13	–
Salinity	(mg/L)	80.0	380.0	216	8.99	–
DO	(mg/L)	2.1	5.4	3.8	0.1	–
Chloride	(mg/L)	16.0	80.0	40.59	1.57	250
Nitrate	(mg/L)	2.30	28.0	11.72	0.67	45
Sulfate	(mg/L)	2.60	34.2	9.44	0.65	200–400
Phosphate	(mg/L)	0.1	0.14	0.1	0	–
Uranium	( $\mu$ g/L)	0.50	26.4	6.97	0.6	1.9*
Total hardness	(mg/L)	40.0	288.0	149.59	7.31	200–600
Ca hardness	(mg/L)	26.0	236.0	104.83	5.87	75–200
Mg hardness	(mg/L)	12.0	96.0	44.76	2.2	30–100
Total alkalinity	(mg/L)	24.0	214.0	115.43	5.79	200–600

SEM standard error mean, total hardness, Ca hardness and Mg hardness and Mg Hardness was calculated as  $\text{CaCO}_3$ . \*Standard guideline for uranium in drinking water as per the International Commission on Radiological Protection.

ranged from 192 to 778  $\mu\text{S}/\text{cm}$  and oxidation redox potential was  $-94.60$  to  $96.10$  mV. The presence of dissolved minerals and electrolytes might contribute to the higher electrical conductivity. The high chloride content in drinking water gives it a salty taste and can also cause constipation (Rao 2006). The chloride, salinity and phosphate concentration in the groundwater samples of the study area ranged from 16 to 80, 80 to 380 and 0.10 to 0.14 mg/l. Total hardness is measured in  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  concentrations and is usually expressed as  $\text{CaCO}_3$ . The key parameters for total hardness are calcium ( $\text{Ca}^{2+}$ ) and magnesium ( $\text{Mg}^{2+}$ ) from 26 to 236 mg/L and 12 to 96 mg/L, respectively. Sedimentary rocks are the most common natural source of hardness in groundwater. Groundwater may dissolve  $\text{CaCO}_3$  during infiltration or along with the flow,  $\text{CaCO}_3$  and  $\text{Mg}(\text{CO}_3)_2$  present in the rocks will enhance the concentration of calcium ions in groundwater. Hard water is usually found in areas with dense topsoil and limestone formation. The total hardness concentration in the research area was found to be between 40 and 288 mg/L. The total hardness value of water samples is well below the recommended limit (WHO 2011). The sum of carbonate and bicarbonate indicates total alkalinity, and carbonate was not found in groundwater belonging to this research area. According to the WHO, the appropriate range of total alkalinity in drinking water is 200–600 mg/L. The alkalinity in the groundwater samples of Bastar district ranged from 24 to 214 mg/L. The concentration of nitrate was determined to be between 2.3 to 28 mg/L, which was within the drinking water standard.  $\text{NO}_3^-$  in groundwater comes from organic industrial effluents, fertilizers, or nitro-fixing bacteria, as well as animal dung, sewage, and septic tanks seeping into the soil and the water matrix. Daily consumption of drinking water containing nitrate  $>45$  mg/l can causes blue baby syndrome or methemoglobinemia, gastric cancer, goitre, birth malformations, and hypertension (Sahu *et al.* 2018). The dissolved oxygen in groundwater of district Bastar ranged between 2.10 to 5.40 mg/l.

### 3.2. The concentration of uranium in groundwater

The concentration of uranium in the water samples was measured in the range of 0.50 to 26.4  $\mu\text{g}/\text{L}$  with an average of 6.96  $\mu\text{g}/\text{L}$  (Figure 2). For human safety, environmental protection organizations have set a safe limit value for uranium concentration in drinking water. According to the International Commission on Radiological Protection (ICRP), the safe limit

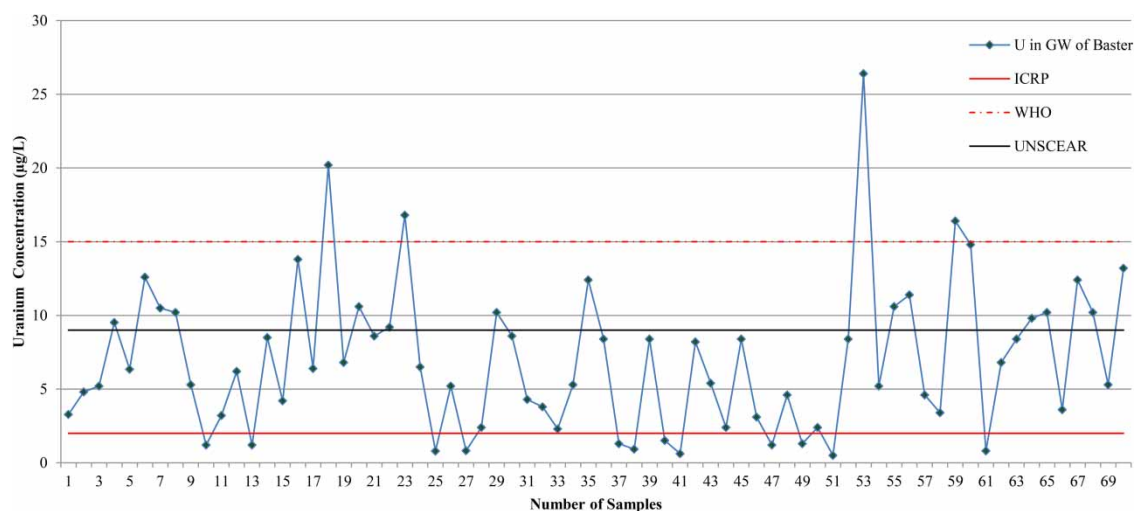


**Figure 2** | Tahsil wise comparatively demonstration of uranium in groundwater of Bastar district.

for uranium in drinking water is 1.9 micrograms per liter (ICRP 1979). Here, 58 groundwater samples were found to exceed the permissible level set by the ICRP (Figure 3). The uranium in groundwater released from natural origin or uranium-containing minerals such as coffinite and pitchblende has been found in the geology of the Indrāvati basin (Kumar *et al.* 2018a, 2018b). Consumption of uranium-contaminated water for drinking purposes can be harmful to human health. The major targets of action of uranium are the kidneys, liver and bones (Sar *et al.* 2017).

### 3.3. Factor analysis

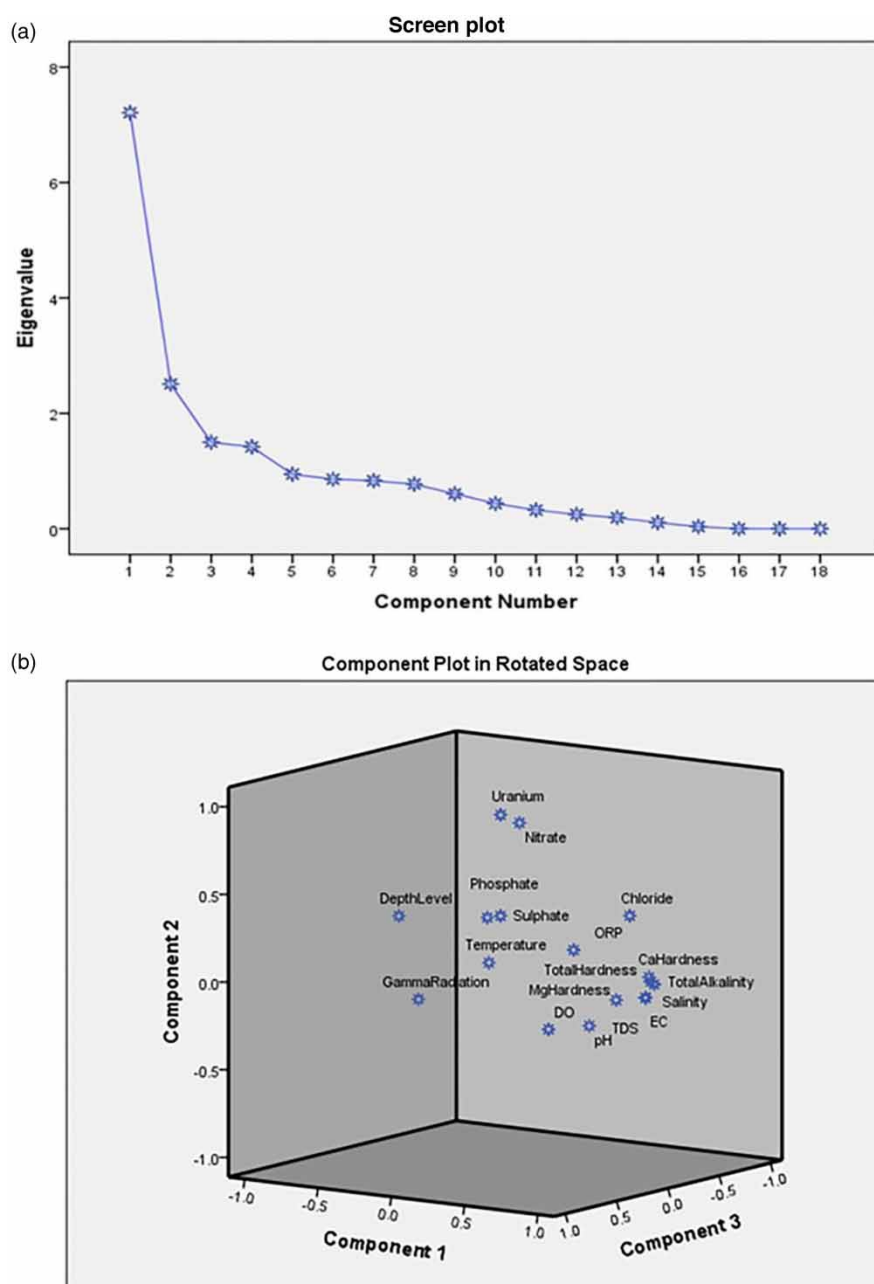
Factor analysis is usually normalized to physico-chemical data and eliminates misclassification due to the different range of variance and the order of magnitude of the analytical parameters. Factor analysis collects the data and describes variables with drawing clear conclusions about the correlation between variable (physico-chemical parameters) and factors. The rotation of the factors was performed by Varimax with Kaiser Normalization. This analysis is used to extrapolate the computational pattern between 17 physico-chemical parameters and explain the sources of affecting water quality. Four factors were yield with  $>1$  eigenvalue from 16 physico-chemical parameters given in Table 2 and Figure 4(a) and 4(b). The rotation sums of the square loadings measuring the degree of closeness between the variables and the values of the factor or PCs are presented in the Table 3. Both positive and negative loadings are used in factor loading. A weak correlation is indicated by a loading between 0 and  $\pm 0.49$ , while a loading between  $\pm 0.5$  and  $\pm 0.74$  indicates a moderate correlation, a loading greater than  $\pm 0.75$  indicates a strong correlation and a loading closer to  $\pm 1$  point indicates the highest correlation (Sahu *et al.* 2018). Factor analysis 1 accounts for 39.864% variance of the total cumulative variance; it has the highest loading



**Figure 3** | Comparative demonstration of concentration of uranium with standard of various International regulatory organizations.

**Table 2** | Total variance explained in factor analysis for physico-chemical variables of groundwater of Bastar district**Total variance explained for physico-chemical parameters**

Component	Initial eigenvalues			Extraction sums of squared loadings			Rotation sums of squared loadings		
	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %	Total	% of Variance	Cumulative %
1	7.21	40.054	40.054	7.21	40.054	40.054	7.176	39.864	39.864
2	2.509	13.936	53.99	2.509	13.936	53.99	2.316	12.864	52.729
3	1.499	8.328	62.318	1.499	8.328	62.318	1.647	9.15	61.879
4	1.419	7.883	70.201	1.419	7.883	7.883	1.498	8.322	70.201

**Figure 4** | Screen plot and component plot in rotated space for factor analysis.



**Table 3** | Rotated component matrix of factor analysis for physico-chemical variables of groundwater of Bastar district**Rotated Component Matrix<sup>a</sup>**

Component	Depth level	Gamma radiation	pH	TDS	EC	ORP	Temp.	Salinity	DO	Cl <sup>-</sup>	NO <sub>3</sub> <sup>-</sup>	SO <sub>4</sub> <sup>2-</sup>	PO <sub>4</sub> <sup>2-</sup>	U	TH	CaH	MgH	TA
1	-0.324	-0.035	0.451	<b>0.976</b>	<b>0.976</b>	0.005	0.05	<b>0.974</b>	0.148	<b>0.818</b>	0.088	0.190	-0.127	-0.078	<b>0.973</b>	<b>0.936</b>	<b>0.731</b>	<b>0.947</b>
2	0.379	-0.035	-0.277	-0.035	-0.035	0.040	0.101	-0.037	-0.333	0.406	<b>0.867</b>	0.394	0.306	<b>0.888</b>	0.028	0.065	-0.082	0.040
3	0.573	<b>0.795</b>	-0.176	0.028	0.028	-0.660	0.229	0.022	-0.212	-0.046	-0.015	0.316	-0.006	-0.066	-0.064	-0.065	-0.038	-0.064
4	-0.173	-0.018	-0.059	-0.026	-0.026	-0.198	<b>0.777</b>	-0.010	0.471	-0.123	0.122	-0.083	<b>0.683</b>	0.182	0.046	-0.032	0.238	0.044

Extraction method: principal component analysis.

<sup>a</sup>. Rotation converged in six iterations. Rotation method: Varimax with Kaiser Normalization.0–0.49 weak loading, 0.50–0.74 moderate loading, 0.75–0.95 strong loading, , –1 highest loading (Sahu *et al.* 2018).

with total dissolved solid, electrical conductivity, salinity, and total hardness while chloride, calcium hardness, and total alkalinity contributing to strong loading. Only magnesium hardness contributed to moderate loading at factor 1. This factor explained the mineralization process in groundwater due to calcium and magnesium hardness. The same trends was also observed in the groundwater of Dongergaon block (Sahu *et al.* 2017). Factor analysis 2 holds a 12.864% variance of the total cumulative variance that does not have the highest loading, but both uranium and nitrate contributed to strong loading. The uranium contained in the groundwater was due to natural occurrences of uranium ore deposited in the bed rock of the aquifer. The highest loading of uranium and nitrate in factor 2 suggested that dissolution from uranium minerals such as uranyl nitrate ( $\text{UO}_2(\text{NO}_3)_2$ ). Saikia *et al.* (2021) also found the similar trends between uranium and nitrate in the groundwater of Nalbari district of Assam. Factor analysis 3 has 9.150% variance of the total cumulative variance and shows strong loading with gamma radiation while depth level contributed to moderate loading, suggesting that the deep bedrock of the aquifer was loaded with a signification amount of radioactive elements. The physico-chemical parameter did not show the highest loading in factor 4, but only temperature contributed to the strong loading, whereas moderate loading with phosphate and factor 4 has 8.322% variance of the total cumulative variance 70.201. Temperature is the most important parameter that influenced the water chemistry. Factor 4 signified that the groundwater temperature played a significant role in mineralization increasing with increase in the temperature.

### 3.4. Spearman's correlation matrix

To determine the relationship between the physico-chemical parameters, Spearman's correlation matrix (two-tailed) was run at 0.01 and 0.05 levels of  $\alpha$  in SPSS (Version 22) and is presented in Table 4. In this correlation matrix, statistically significant relationships for uranium were found between nitrate, chloride and sulfate. This indicates the existence of uranium as a dissolved salt in the form of uranium chloride. A similar trend was reported by many researchers (Singh *et al.* 2003; Selvi *et al.* 2016; Sharma *et al.* 2019). Chloride and nitrate are uranium carriers and therefore show a positive correlation with uranium, consequently, their assurance analysis and correlation are important (Sar *et al.* 2017).

### 3.5. Hierarchical cluster analysis

Hierarchical cluster analysis is an efficient data mining strategy that divides variables into groups by their similarities and differences within each category. Classification of physico-chemical variables was performed using cluster analysis. Ward's linkage method was applied to calculate the similarity of physico-chemical data and the results provided visually meaningful dendrograms and differentiated groups. The dendrogram classified the 18 variables into six broad classes, each of which was further subdivided into smaller clusters, as seen in Figure 5(a). The entire physico-chemical parameter was divided into two macro-clusters, which showed the maximum distance of 25 scales in the Rescale Distance Cluster Combined Scale. Cluster 1 is further divided into two micro-clusters, pH, DO,  $\text{PO}_4^{2-}$ ,  $\text{NO}_3^-$ , and uranium in micro-cluster 1.1 and  $\text{SO}_4^{2-}$ ,  $\text{Cl}^-$ , magnesium hardness, temperature, and ORP included in mini-cluster 1.2. Macro-cluster 2 is further divided into two micro-clusters with a disparity level 13 scale, in which EC contributed to micro-cluster 2.2, while micro-cluster 2.1 again splits into three mini-clusters containing TDS, and salinity show similarity and formed mini-cluster 2.1.1. The second mini-cluster 2.1.2 was showing to be similar between groups, encompassing calcium hardness, total alkalinity, and total hardness, while depth and gamma radiation contributed to mini-cluster 2.1.3. Cluster analysis was also used by Sahu *et al.* (2018), to classify the physico-chemical variable of groundwater samples.

Ward's linkage method was also applied to calculate the level of similarity in the concentration of uranium released from the geology of the respective groundwater sampling locations presented in Figure 5(b), providing visually meaningful dendrograms, which divided all 70 sampling loci into 10 sub-clusters. Sub-clusters 1 to 8 are minor branches of cluster 1, while sub-clusters 9 and 10 are both major branches of cluster 2. The dissolution of uranium in groundwater was similar within the groups/clusters, while dissimilarity was measured between groups/clusters measured as rescaled distance cluster combine scale, present in Figure 5(b).

### 3.6. Non-carcinogenic risk assessment

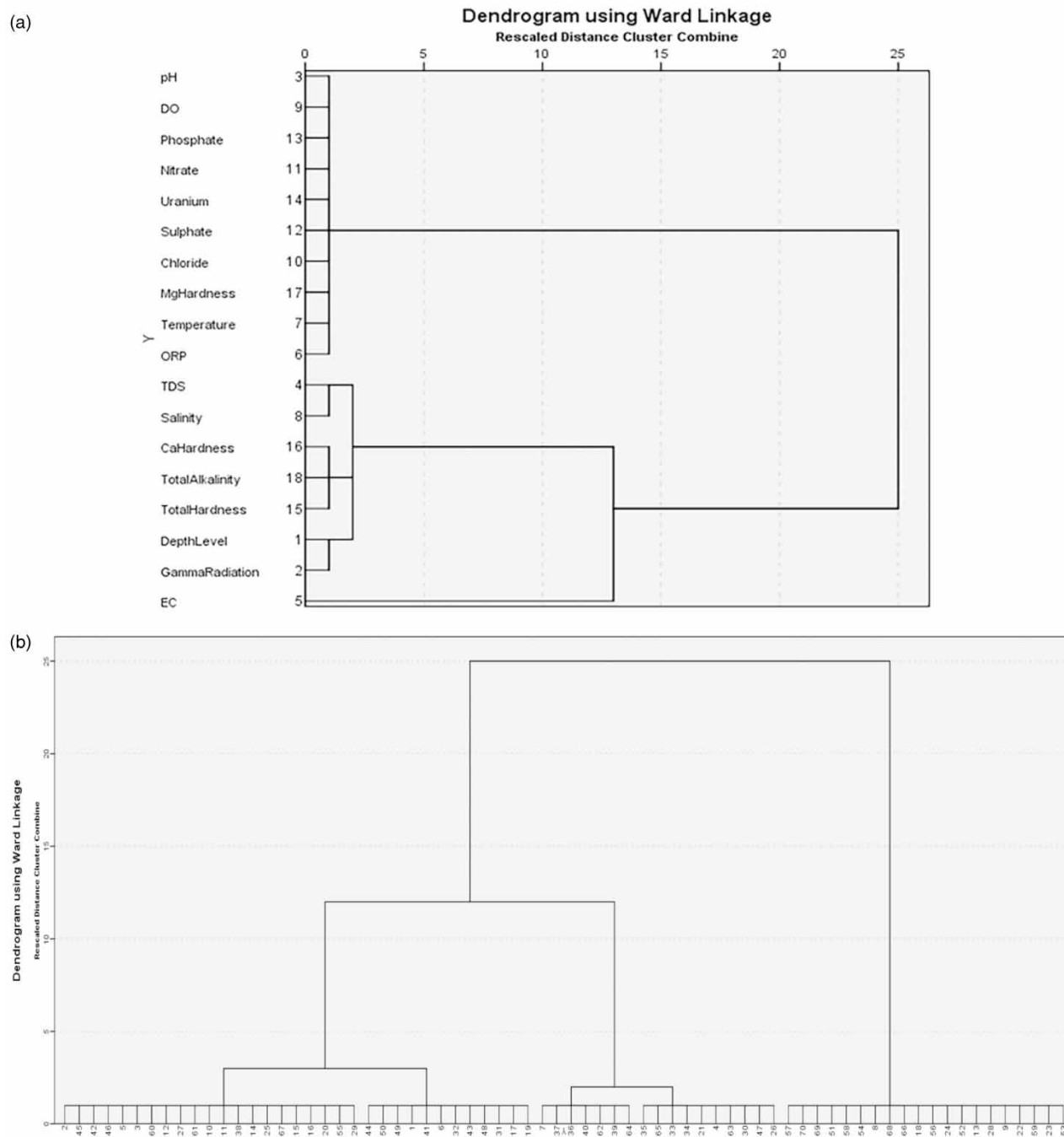
Non-carcinogenic risk was determined by analyzing CDI by potable water and uranium concentration in drinking water for different age groups (young children, children, and adults) of residents living in the study region. Figure 6 compares the RfD with the analyzed CDI value for young children, children and adult residents in Bastar district and Figure 7 compares the recommended HQ doses with the analyzed HQ values for young children, children and, adult residents in Bastar district. The observed CDI values for young children, children, and adults in the Bastar district ranged from 0.04 to 1.89 with an average of 0.50 for young children, 0.04 to 1.98 with an average of 0.52 for children, and 0.05 to 2.64 with an average of 0.70 for

**Table 4** | Spearman's correlation coefficient within physicochemical parameters of Bastar district ( $n=70$ )

	DL (feet)	GR (ns/ h)	pH	TDS (mg/l)	EC ( $\mu$ S/ cm)	ORP (mV)	Temp. (oC)	Salinity (mg/l)	DO (mg/l)	Cl <sup>-</sup> (mg/l)	NO <sub>3</sub> <sup>-</sup> (mg/l)	SO <sub>4</sub> <sup>2-</sup> (mg/l)	PO <sub>4</sub> <sup>2-</sup> (mg/l)	U ( $\mu$ g/ l)	TH (mg/l)	CaH (mg/l)	MgH (mg/l)	TA (mg/l)
Depth level	1																	
Gamma radiation	0.384 <sup>b</sup>	1																
pH	-0.317	-0.033	1															
TDS	-0.279	-0.031	0.401 <sup>b</sup>	1														
EC	-0.279	-0.031	0.401 <sup>b</sup>	1	1													
ORP	-0.181	-0.277	0.099	0.011	0.011	1												
Temp.	-0.070	0.134	-0.064	0.032	0.032	-0.211	1											
Salinity	-0.284	-0.040	0.401 <sup>b</sup>	0.999 <sup>b</sup>	0.999 <sup>b</sup>	0.005	0.038	1										
DO	-0.218	-0.115	0.153	0.143	0.143	0.077	0.138	0.151	1									
Cl <sup>-</sup>	-0.139	-0.039	0.319 <sup>a</sup>	0.788 <sup>b</sup>	0.788 <sup>b</sup>	0.058	0.015	0.785 <sup>b</sup>	-0.036	1								
NO <sub>3</sub> <sup>-</sup>	0.225	0.057	-0.111	0.030	0.030	-0.043	0.163	0.029	-0.096	0.447 <sup>b</sup>	1							
SO <sub>4</sub> <sup>2-</sup>	0.237 <sup>a</sup>	0.097	-0.089	0.169	0.169	-0.046	0.128	0.160	-0.056	0.224 <sup>a</sup>	0.182	1						
PO <sub>4</sub> <sup>2-</sup>	0.348 <sup>a</sup>	0.207	-0.056	0.084	0.084	0.163	0.002	0.078	-0.054	0.107	0.160	0.149	1					
U	<b>0.212<sup>a</sup></b>	-0.059	-0.182	-0.121	-0.121	-0.054	0.205	-0.117	-0.215	<b>0.259<sup>a</sup></b>	<b>0.748<sup>b</sup></b>	<b>0.263<sup>b</sup></b>	0.070	1				
TH	-0.338	-0.088	0.375 <sup>b</sup>	0.920 <sup>b</sup>	0.920 <sup>b</sup>	0.037	0.048	0.918 <sup>b</sup>	0.159	0.768 <sup>b</sup>	0.112	0.154	0.071	-0.050	1			
CaH	-0.290	-0.091	0.334 <sup>a</sup>	0.899 <sup>b</sup>	0.899 <sup>b</sup>	0.046	0.001	0.892 <sup>b</sup>	0.172	0.769 <sup>b</sup>	0.153	0.159	0.072	-0.047	0.967 <sup>b</sup>	1		
MgH	-0.346	-0.049	0.354 <sup>b</sup>	0.658 <sup>b</sup>	0.658 <sup>b</sup>	0.001	0.155	0.666 <sup>b</sup>	0.069	0.499 <sup>b</sup>	-0.037	0.085	0.045	-0.040	0.738 <sup>b</sup>	0.543 <sup>b</sup>	1	
TA	-0.304	-0.103	0.353 <sup>b</sup>	0.894 <sup>b</sup>	0.894 <sup>b</sup>	0.034	0.031	0.891 <sup>b</sup>	0.139	0.723 <sup>b</sup>	0.117	0.150	0.083	-0.040	0.969 <sup>b</sup>	0.934 <sup>b</sup>	0.722 <sup>b</sup>	1

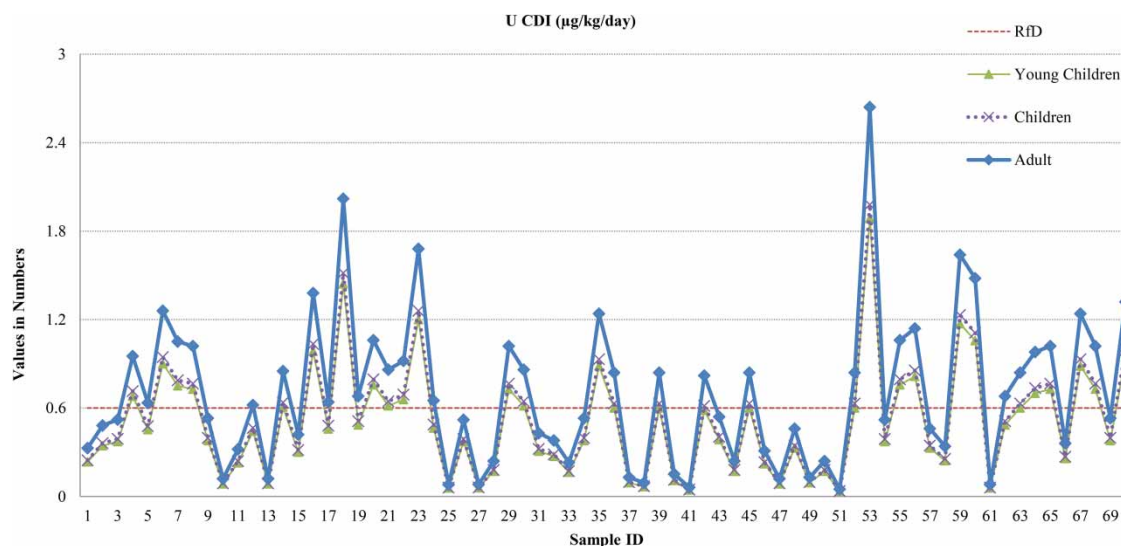
0–0.49 weak correlation, 0.5–0.74 moderate positive correlation,  $\geq 0.75$  strong correlation, -1 highest correlation (Singh *et al.* 2021).

Correlation is significant (a) at the 0.05 level and (b) at the 0.01 level.

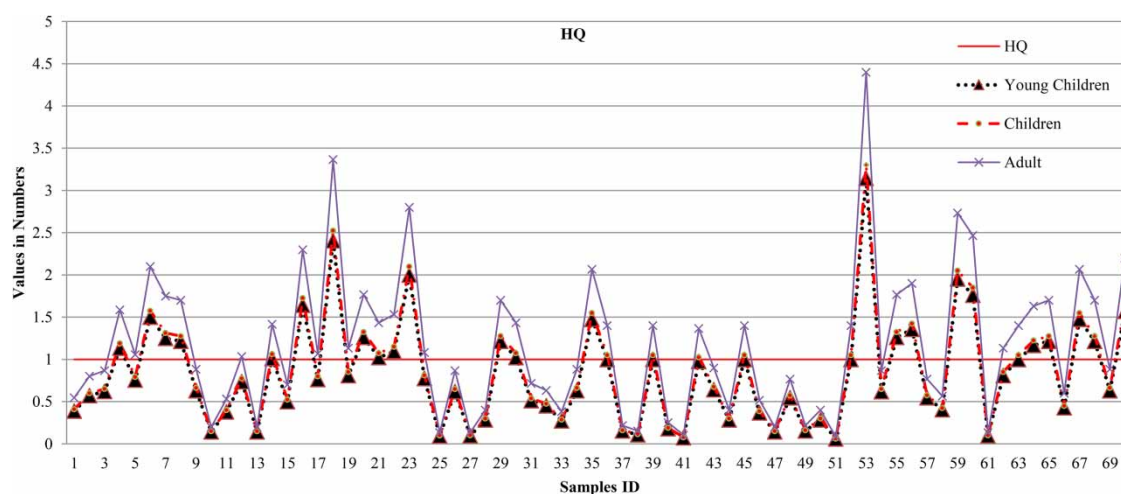


**Figure 5** | Dendrograms or tree plot of cluster analysis for physico-chemical variables (a) and samples-wise (b).

adults shown in Table 5. The assessed value of the CDI has been found to exceed the reference dose in 24 samples for young children, 30 samples for children, and 36 samples for adults. WHO has suggested that a maximum of  $0.6 \mu\text{g/kg/day}$  uranium may be acceptable, beyond the recommended values would pose potential health risks to humans and other living beings, such as gene mutations, defects in developing fetuses and infants etc. Numerous researchers have also conducted uranium exposure risk assessments in different parts of Chhattisgarh (Sar *et al.* 2018; Sahu *et al.* 2019; Singh *et al.* 2021). CDI was previously studied by Singh *et al.* in the Bastar district in 2021, and ranged from 0.03 to 1.67, which is the lowest in comparison to the present study.



**Figure 6** | Comparatively demonstration of chronic daily intake of uranium, recommended reference dose and hazardous quotient for different age groups of inhabitants of Bastar district.



**Figure 7** | Comparatively demonstration of hazardous quotient for different age groups of inhabitants of Bastar district.

**Table 5** | CDI and HQ values for non-carcinogenic risk assessment in different age groups of inhabitants of the Bastar district

District	Age group	Statistics	Intake rate (liter)	Exposure duration (year)	Exposure frequency (day)	Body weight (kg)	Average time (day)	Reference dose (µg/kg/day)	U level in GW (µg/l)	U CDI (µg/kg/day)	Hazard quotient
Bastar	Young children (3–6 years)	Mean	1	4	365	14	1,460	0.6	6.97	0.5	0.83
		Min	1	4	365	14	1,460	0.6	0.5	0.04	0.06
		Max	1	4	365	14	1,460	0.6	26.4	1.89	3.14
	Children (7–18)	Mean	3	12	365	40	4,380	0.6	6.97	0.52	0.87
		Min	3	12	365	40	4,380	0.6	0.5	0.04	0.06
		Max	3	12	365	40	4,380	0.6	26.4	1.98	3.3
	Adults (>18)	Mean	6	30	365	60	10,950	0.6	6.97	0.7	1.16
		Min	6	30	365	60	10,950	0.6	0.5	0.05	0.08
		Max	6	30	365	60	10,950	0.6	26.4	2.64	4.4



HQ values for young children, children and adults in the study area ranged from 0.06 to 3.14 with an average of 0.83 for young children, 0.06 to 3.30 with an average of 0.87 for children, and 0.08 to 4.40 with an average of 1.16 for adults. The measured values of HQ in 24 water samples for young children were found to exceed the recommended value of HQ in 32 water samples for children and 36 water samples for adults, the HQ exceeded the recommended value of 1, which indicated a significant possibility of chemical toxicity (WHO 2008). HQ in the Bemetara district was previously studied by Sahu *et al.* (2019), and ranged from 0.04 to 6.04 which was higher compared with our study. Uranium risk assessment studies were also conducted in several districts of Chhattisgarh state such as Durg, Balod, Bemetara, Kanker, but there was no population difference in different age groups and lack of data or no such research in Bastar district of Chhattisgarh state (Sar *et al.* 2017; Sahu *et al.* 2019; Sahu *et al.* 2020).

#### 4. CONCLUSION

The objective of this study was accomplished to assess the quality and health risks of uranium-rich groundwater by ingestion of uranium through drinking water in age groups of residents of Bastar district, Chhattisgarh, Central India. In total, 58 groundwater samples in the district were found to have high concentrations of uranium in excess of the standard (1.9 µg/L) recommended by the ICRP, which may be a cause for concern. Uranium shows a significant affinity with nitrate, chloride and sulfate ions. The measured concentration of pH, TDS, salinity, total hardness, magnesium hardness, total alkalinity,  $\text{Cl}^-$ ,  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and,  $\text{PO}_4^-$  were well within the drinking water standards of WHO and BIS, but it was observed for calcium hardness concentration, that is exceeds WHO and BIS drinking water standards. Factor analysis calculated four factors in the 17 groundwater parameters, extracting 70.201% of the cumulative variance in the data set. Cluster analysis was successfully performed with better results for the uranium releasing nature in the geology of the district across all sampling locations. The CDI of uranium from drinking water was found to exceed the reference value. The HQ was found to be >1 for young children, children, and adults. The study strongly suggested, for vital consideration, in initiation of a study on temporal variation and adoption preventive actions or installed remedial technologies in the prone area.

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#### DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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