



Spatial distribution and risk assessment of naturally occurring uranium along with correlational study from Buldhana district of Maharashtra, India

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Abstract

The current study of the spatial distribution of uranium and water quality parameters along subsequent radiological impact due to uranium in the groundwater from the Buldhana district was undertaken. The chemo-radio toxicological dose owing to such dissolved uranium is estimated. The water quality parameters are compared with the World Health Organization and Bureau of Indian Standard's safe recommended limits and found well below. A correlation study was carried out between uranium and water quality parameters. Spatial distribution is mapped by GIS-based software. The chemo-radio toxicological risks due to uranium for different age groups were calculated. This finding in the study suggests that groundwater of the region is safe for drinking purposes based on a chemo-radiological point of view.

Keywords Uranium · Groundwater quality · Radiotoxicity · Chemical toxicity · Annual effective dose · Spatial distribution pattern

List of symbols

U	Uranium
pH	Potential of hydrogen
TDS	Total dissolved solids
EC	Electrical conductivity
DO	Dissolved oxygen
ORP	Oxidation reduction potential
TH	Total hardness
WHO	World health organization
BIS	Bureau of Indian Standards
GIS	Geographic information system
LULC	Land use land cover
DEM	Digital elevation model

USEPA	United States Environmental Protection Agency
EDTA	Ethylenediaminetetraacetic acid
AERB	Atomic Energy Regulatory Board
UNSCEAR	United Nations Scientific Committee on the Effects of Atomic Radiation

Introduction

Uranium is geogenic in nature and has three isotopes i.e. ^{234}U , ^{235}U , and ^{238}U , out of these, the most abundance is ^{238}U , which makes up about 99.27% mass of natural uranium [1, 2]. The sources of uranium in the aquatic environment are leaching from natural deposits like granite [3], releases from uranium industries [4], coal-burning [5], and excessive use of fertilizers containing phosphates [6, 7]. The adverse effects of uranium on human health are due to its radiological [8] and chemical properties [9, 10]. Radioactive element with heavy metal nature is possible to deliver a trace quantity of radiation dose to the population [11]. Due to its chemo-radiological characteristics, uranium contents are being regularly monitored by various health agencies and organizations throughout the world. United States Environmental Protection Agency (USEPA) in 1991 classified uranium as a category group A element and also suggested the

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complete nonappearance of uranium in drinking water as the safe limit for carcinogenic risk [12]. There is variation in the permissible limits for uranium concentration suggested by different agencies. For example, the World Health Organization has earlier suggested 15 $\mu\text{g/L}$ but now the limit has been raised to 30 $\mu\text{g/L}$ [13]. The physicochemical properties of water play a key role in assessing the quality and suitability of water for drinking purposes. Parameters like these are also supportive in studying and demonstrating the speciation of radionuclides and various pollutants due to geogenic and anthropogenic activities in the water ecosystem [14]. The origin of dissolved solids in the groundwater in different geological regions is due to the solubility of minerals [15]. The pH is an important monitoring parameter to assess the acidity or basicity of water to evaluate the health of the water ecosystem, to check the suitability of water for multiple usages. Anions, such as F^- , NO_3^- , SO_4^{2-} , and PO_4^{3-} present in water play a significant role in assessing the quality of water for drinking purposes. These anions must be monitored meticulously as their presence beyond the permissible limit can make water toxic and unfit for drinking purposes. The presence of fluoride in groundwater in a low amount protects from dental caries but in higher concentration ($> 2 \text{ mg/L}$) may cause dental fluorosis while the concentration less than (0.5 mg/L) may cause tooth decay [16]. Nitrate concentration over the recommended limit (50 mg/L as NO_3^-) [17] can cause blue baby syndrome [18]. Similarly, high sulfate content can cause diarrhea and intestinal disorders [19]. Hence, keeping in view these influences, the current work of chemo-radiological risk assessment due to naturally occurring uranium in groundwater was carried out with the following objectives as **1.** Spatial distribution of uranium and associated water quality parameters of Buldhana district. **2.** Impact and correlation of water quality with uranium concentration **3.** Assessment of chemo-radiological risk and associated radiation dose due to exposure of uranium to population over the study area.

Geology of study area

Buldhana district as a study area positioned in the western part of the Vidarbha region and northern head of Marathwada (Fig. 1a). This study area is spread in latitudes $19^\circ 51'$ to $21^\circ 17'$ in North, $75^\circ 57'$ to $76^\circ 59'$ longitudes towards East and it covers about 9670 km^2 of India. Buldhana had a population of 2,586,258 in 2011 [20]. It is bounded by the Madhya Pradesh state in the north, Aurangabad district in the west, Parbhani district to the south. Akola and Amravati districts are belonging to the east and northeast sides of Buldhana. The study area was occupied by the alluvium zone and basalts hard rocks area Fig. 1a. A major part of the district is covered by basaltic lava flows of Upper Cretaceous to Lower Eocene age [21]. The southern part of the study

area is grouped under the Sahyadri range, whereas the northern part is grouped under the Satpudas range (Fig. 1c). Deccan Trap Basalt found is an important formation to groundwater movement in the study area. Depositional vesicular massive units of dissimilar lava flows have provided the reason for rising the multi-layered aquifer system [21]. The groundwater movement capacity of Vesicular basalt mainly depends on the size, shape, density, and degree of interconnection of vesicles [22]. The northern part of the district is underlain by thick alluvial deposits of age from Pleistocene to Recent, which is called Purna Alluvium. The Alluvium is also observed in the minor patch in the southwest of the Malkapur district and east side of the Khambgaon district [21]. The Alluvial deposits, due to inter pore spaces in sand and gravel, provides them a high permeability and porosity and to make them a good groundwater reservoir. In the study area, a unique 'Lonar crater' is situated which is the only such crater in the world with basaltic nature and is believed to be created due to asteroid collision with earth's impact in the Pleistocene Epoch [23]. There is three major type soil found namely as Bharkali, Barad, and Morand. Morand soil is a mixture of silt and lime, heavy in texture and blackish which suitable for cotton production. In Chikhali and Mehkar taluka area, Black soil is found, but the depth of Topsoil is varying. Due to that, it is suitable for wheat cultivation, hence the majority area of the study region is under the cropland (Fig. 1b). Barad is a local name for sandy soil having a more sand percentage than silt and clay. Two main rivers are flowing through the study area namely Penganga and Purna. Katepunrana and Van are the tributaries of these rivers show in Fig. 1c [21]. The maximum temperature of the study area in summer ranges from 43 to 45°C respectively. The average annual rainfall recorded in the study area is 900 mm [24].

Experimental technique

Sample collections

The targeted study area is divided into a systematic $6 \times 6 \text{ km}^2$ grid to cover the entire area and avoid bias in the sampling. The geomorphological and Land use land cover map of the study region were prepared using ArcGIS 10.8 software [25], which is shown in Fig. 1a, b. A total of 102 samples were collected from different hand pumps and bore wells of various water depths. These sources were dug in either the residential areas or agricultural fields and were being used for drinking and irrigation purposes. Wherever the groundwater source was not available, surface water was collected as a sample source. Before the collection of samples, the water sources were kept running for 2–5 min, and then water samples were collected in

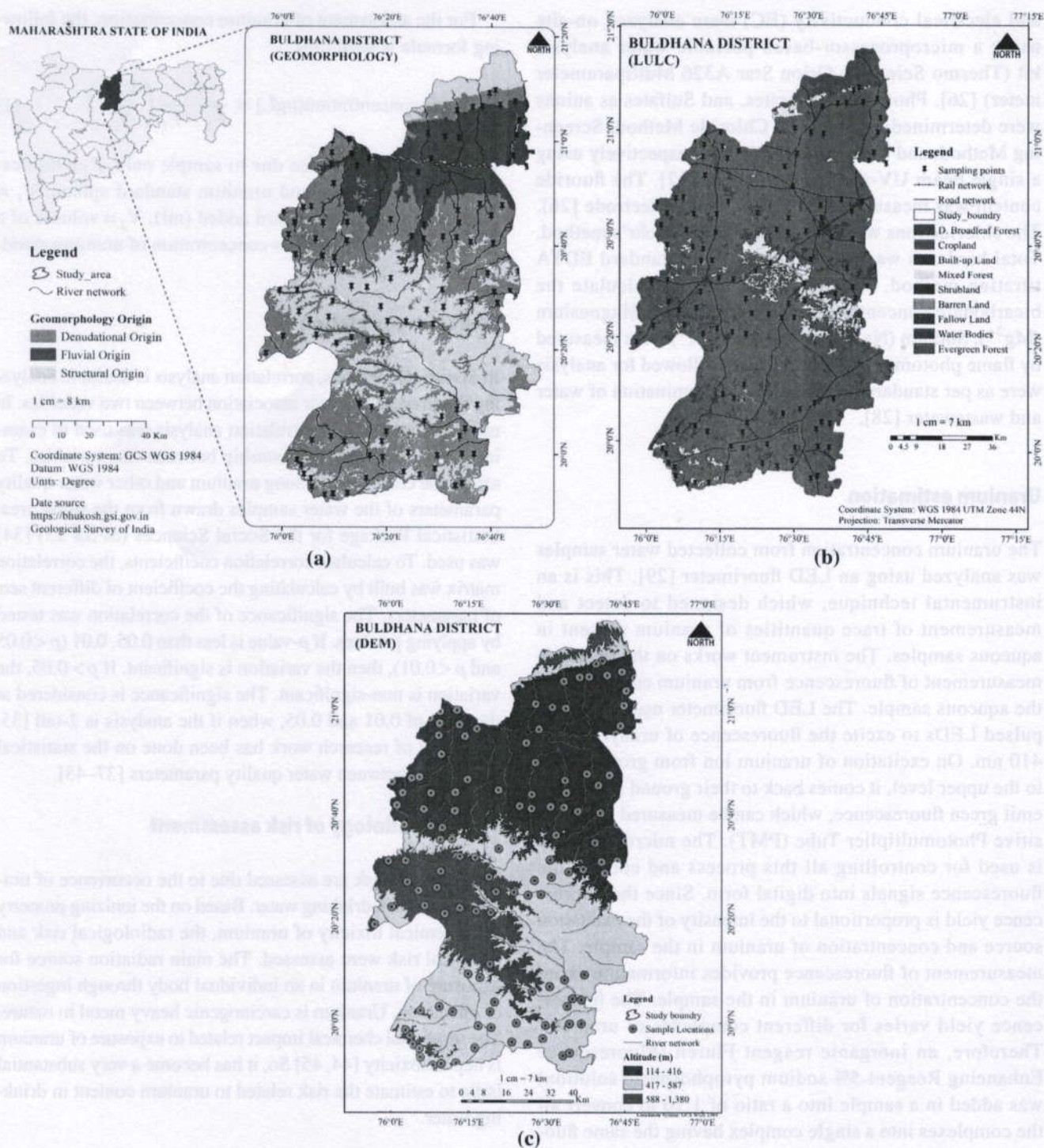


Fig. 1 a Geomorphology map of Buldhana district as a study area. b Land use land cover (LULC) map of the study area. c Digital elevation model (DEM) of Buldhana district

pre-cleaned (10% nitric acid) polyethylene bottles. After analyzing the in-situ parameters, the one-liter capacity bottle is filled with samples, well labeled with primary data; send to ex-situ analysis.

Major ions and field measurement analyses

The in-situ parameters like pH, temperature, oxygen reduction potential (ORP), total dissolved solids (TDS),

and electrical conductivity (EC) were analyzed on-site using a microprocessor-based portable water analysis kit (Thermo Scientific Orion Star A326 Multiparameter meter) [26]. Phosphates, Nitrates, and Sulfates as anions were determined by Stannous Chloride Method, Screening Method, and Turbidimetry Method respectively using a single beam UV-spectrophotometer [27]. The fluoride content was measured with Ion-selective Electrode [26]. The chloride ions were determined using Mohr's method. Total hardness was estimated using the standard EDTA titration method. Alkalinity was used to calculate the bicarbonate concentration. Calcium (Ca^{2+}), Magnesium (Mg^{2+}), Sodium (Na^+) and Potassium (K^+) were measured by flame photometry. All procedures followed for analysis were as per standard methods for the examination of water and wastewater [28].

Uranium estimation

The uranium concentration from collected water samples was analyzed using an LED fluorimeter [29]. This is an instrumental technique, which designed to detect and measurement of trace quantities of uranium present in aqueous samples. The instrument works on the principle measurement of fluorescence from uranium complexes in the aqueous sample. The LED fluorimeter uses a bank of pulsed LEDs to excite the fluorescence of uranyl ions at 410 nm. On excitation of uranium ion from ground level to the upper level, it comes back to their ground state, they emit green fluorescence, which can be measured by a sensitive Photomultiplier Tube (PMT). The microcontroller is used for controlling all this process and convert the fluorescence signals into digital form. Since the fluorescence yield is proportional to the intensity of the excitation source and concentration of uranium in the sample. The measurement of fluorescence provides information about the concentration of uranium in the sample. The fluorescence yield varies for different complexes of uranium. Therefore, an inorganic reagent Fluren (Fluorescence Enhancing Reagent-5% sodium pyrophosphate solution) was added in a sample into a ratio of 1:10 to convert all the complexes into a single complex having the same fluorescence yield. To assess the uranium concentration in an aqueous solution various methods are available. The main purpose to select the LED fluorimeter cost-effective with precision than others. The main benefit of this method viz., there is no need to sample preparation and has a very good detection limit, however, it has a lower dynamic range of detection. The detection limit of this method is 0.2 $\mu\text{g/L}$ [30–32]. Before uranium content measurement, an LED fluorimeter was calibrated with the standard solution provided by Quantalase Enterprises Pvt. Ltd., Indore, India.

For the assessment of uranium concentration, the following formula is used [33].

$$\text{Uranium concentration}(\mu\text{g/L}) = \frac{F_1}{F_2 - F_1} \left(\frac{V_1 C}{V_2} \right) \quad (1)$$

where F_1 = fluorescence due to sample only, F_2 = fluorescence due to sample and uranium standard spiked, V_1 = volume of uranium standard added (ml), V_2 = volume of a sample taken (ml), and C = concentration of uranium standard solution ($\mu\text{g/L}$).

Pearson's correlation

In Statistical methods, correlation analysis is useful in analyzing the strength of linear association between two variables. In many research fields, correlation analysis was used to examine getting the linear relationship between two variables. To assess the correlation among uranium and other water quality parameters of the water samples drawn from the study area, Statistical Package for the Social Sciences (SPSS 23) [34] was used. To calculate correlation coefficients, the correlation matrix was built by calculating the coefficient of different sets of parameters. The significance of the correlation was tested by applying p values. If p -value is less than 0.05, 0.01 ($p < 0.05$ and $p < 0.01$), then the variation is significant. If $p > 0.05$, the variation is non-significant. The significance is considered at the level of 0.01 and 0.05, when if the analysis is 2-tail [35, 36]. A lot of research work has been done on the statistical correlation between water quality parameters [37–43].

The methodology of risk assessment

Two types of risk are assessed due to the occurrence of uranium content in drinking water. Based on the ionizing property and chemical toxicity of uranium, the radiological risk and chemical risk were assessed. The main radiation source for exposure of uranium in an individual body through ingestion or inhalation. Uranium is carcinogenic heavy metal in nature. The most vital chemical impact related to exposure of uranium is nephrotoxicity [44, 45] So, it has become a very substantial issue to estimate the risk related to uranium content in drinking water.

Radiological risk estimation

The radiological risk also known as *Excess cancer risk* (ECR) is the probability of developing cancer to an individual in life due to exposure to potentially carcinogenic compounds like uranium [46, 47]. It is calculated using the following equation:

$$\text{Excess cancer risk(ECR)} = U_c \times R \quad (2)$$

where U_c is uranium concentration in water expressed in Bq/L and calculated as

= concentration of U($\mu\text{g/L}$) \times conversion factor(0.02528 Bq/L)

R is the risk factor, calculated by the following equations:

$$(R) = r \times IR \times TEP \quad (3)$$

where r = uranium risk coefficient for mortality and morbidity was taken 1.19×10^{-9} and 1.84×10^{-9} Bq/L respectively [48], IR = Ingestion Rate (L day^{-1}) [48] and TEP = 65 years i.e. 23,725 days [13].

Chemical toxicity risk estimation

Uranium in drinking water affects the kidney and bones of humans due to its chemo-toxicological properties. The chemical toxicity of uranium is estimated through the term Lifetime Average Daily Dose (LADD). The LADD is assessed by the following equation [13].

$$\text{LADD}(\mu\text{g/L kg}^{-1} \text{ day}^{-1}) = \frac{U \times IR \times EF \times LE}{BW \times AT} \quad (4)$$

where U = uranium concentration in water sample ($\mu\text{g/L}$), IR = Ingestion Rate (L day^{-1}), EF = Exposure Frequency 365 (days year^{-1}) [48]. LE = life expectancy was taken 70 years, IR 4.05 L day^{-1} , BW is body weight (kg), which is taken as 70 kg. AT is an average time (Days) of exposure, which is taken as 25,550 days for adults.

Hazard Quotient (HQ) is used to estimate the degree of effect due to the ingestion of uranium through water consumption. Whenever HQ value is found to be less than 1 (< 1), then there are no adverse health issues probable through the exposure of uranium in water. [49] It was calculated using the following equation:

$$\text{HQ} (\mu\text{g/L kg}^{-1} \text{ day}^{-1}) = \frac{\text{LADD}}{\text{Rfd}} \quad (5)$$

where Rfd is the Reference Dose. Its value is $4.53 \mu\text{g kg}^{-1} \text{ day}^{-1}$ [50, 51]

Assessment of annual effective dose (AED)

The annual effective dose is to assess how an individual is unsafe from the exposure to the radiation. Worldwide, about 87% of the total radiation dose acquired by a human is due to natural sources. [52] and in India, about 97.7% of radiation dose received by an average Indian populace is observed due to natural radiation sources [52]. The radiation dose owed by the ingestion of uranium through drinking water for different age groups is estimated by using the following conversion factor suggested by the International Atomic Energy Agency [53].

$$\text{AED} = Ac \times F \times I \times 365 \quad (6)$$

where Ac = average concentration of uranium (Bq/L), F = Effective per unit intake ($\mu\text{Sv year}^{-1} \text{ Bq L}^{-1}$) is taken 4.4×10^{-8} [53] and I = Age-dependent Daily water intake (L day^{-1}) for different age groups are suggested by Dietary Reference Intakes [54].

Spatial distribution of uranium

The Interpolation method was adopted for spatial distribution mapping of uranium and other water quality parameters. It is a well-known geostatistical tool used for the elimination of countless defects and limitations [55]. The interpolation method is very important in the geospatial analysis or distribution modeling of any parameter. IDW is an algorithm to interpolate the spatial data based on weighted average values of surrounding sample points. The interpolation method is assuming the values are nearer to one another are more alike than those which are farther away. The inverse distance weighted (IDW) method in the spatial analyst-modeling tool (ArcGIS 10.8) is used to interpolate the unknown uranium concentration from the known place.

Results and discussion

Result analysis and interpretation

To fulfill, the ever-increasing demand for the food supply of a large population, chemical-based fertilizers are used to increase the production of crops. However, this additional use of fertilizers and other chemical-based compounds are washed away with rains, percolates through the soil, and affects the quality of groundwater. According to health agencies like WHO about 80% of diseases that occur in humans are due to unsafe drinking water. Therefore, it is an important aspect to regularly monitor the quality status of water and cross-check them to find out whether they are contradicting the standard limits prescribed by WHO/BIS [56, 57]. The descriptive statistics of all parameters are presented in Table 1 and represented in the forms of a box plot (Fig. 2a, b). Based on the analysis, the pH is observed slightly alkaline in nature with a mean value of 7.4 in the study area due to the dissolution of soil nutrients and minerals. The pH value of most of the collected groundwater samples (97%) is observed within the recommended permissible limit by WHO/BIS i.e. 6.5 to 8.5 [50] and only about 3% of samples are crossing the recommended limits. The electrical conductivity of collected samples is recorded in the range from 285 to 6031 $\mu\text{S/cm}^1$ with an average of 1586 $\mu\text{S/cm}^1$. At some locations, the higher values of EC are inferred that these areas of Purna River Alluvium are

Table 1 Statistical summary of physico-chemical parameters of groundwater samples (n = 102)

Parameters	pH	ORP	EC	TDS	DO	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ⁻²	PO ₄ ⁻³	U	TH	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	HCO ₃ ⁻
Mean	7.4	181	1586	778	6.2	0.2	309	99 (80–117)	234	15 (13–16)	2.5	499	85 (77–92)	55 (51–58)	88 (80–95)	4 (4–4)	532 (492–571)
(LCL–UCL)	(7.32–7.48)	(178–185)	(1414–1758)	(693–862)	(6–6.39)	(0.18–0.21)	(246–371)		(216–251)		(2.1–2)	(453–544)					
* 95% CL																	
WHO PL	8.5	–	–	1500	–	1.5	600	50	400	–	30	500	200	150	200	–	–
% of sam-ples above WHO PL	2.94	–	–	3.92	–	0	22.55	50.98	5.88	–	0	45.1	0	0	1.0	–	–
BIS PL	8.5	–	–	2000	–	1.5	1000	–	400	–	60*	600	200	100	–	–	600
% of sam-ples above BIS PL	3.0	–	–	3.0	–	–	1.0	–	6.0	–	–	37	0	3.0	–	–	35

All values are in mg/L except pH, EC (μS/cm¹) ORP (mV), and U (μg/L)

*LCL lower control limit, UCL upper control limit, CL confidence level

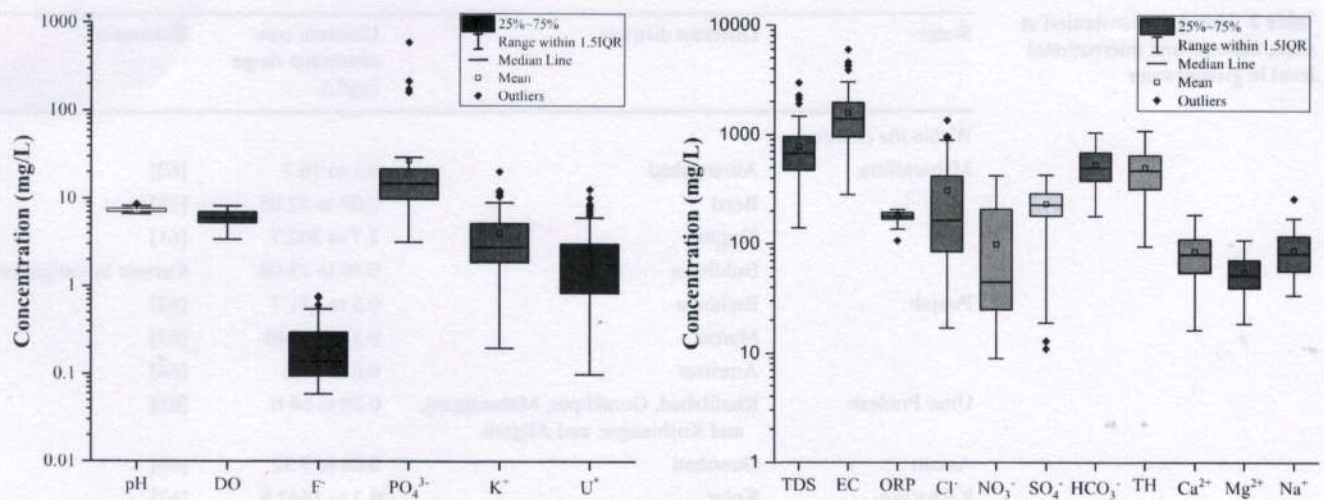


Fig. 2 Representation through the box plot of physio-chemical parameters of the study area

affected by internal salinity problem due to diagenetically different meteoric water having lengthier residence period, high rate of evapotranspiration and it is restricted to the sandy aquifers inter-layered with clayey beds due to which less recharge of groundwater is taking place. The TDS values were observed in the range from 140 to 2966 mg/L with a mean value of 778 mg/L. According to WHO (1500 mg/L) and BIS (2000 mg/L) recommended limits, 3.9%, and 3% of total samples, TDS values were found to exceed the permissible limit. The excessive dissolved solids, water taste like a metallic and it is the source for corrosion of pipes. In this study, TDS concentration was high due to the presence of ions like bicarbonates, carbonates, sulfates, chlorides, and calcium. Based on alkaline pH and anionic study of groundwater it is has been concluded that the bicarbonates were dominating in comparison with other anions. The observed concentration of dissolved bicarbonates is in the range of 180 to 1050 mg/L with an average value of 532 mg/L. The fluoride concentration is observed in the range from 0.06 to 0.77 mg/L with an average of 0.20 mg/L and no sample was crossing the recommended limit i.e. 1.5 mg/L [56]. NO_3^- in drinking water causes adverse health effects when it exceeds the maximum permissible limit (50 mg/L) recommended by WHO. The observed concentration of NO_3^- varies in the range from 9 to 425 mg/L with a mean value of 99 mg/L. From the total number of samples, 50.98% exceeds the maximum permissible limit suggested by WHO. These observed high concentrations are indicative of anthropogenic pollution by human activity like excessive use of chemical-based fertilizers. The same results are found by the Central groundwater Board in 2011 [21]. It is a very significant issue for further study in this regard. The SO_4^{2-} concentrations are observed in the range from 11 to 430 mg/L with an average value of 234 mg/L. About

6% of the total samples were found to cross the maximum permissible limit suggested by WHO/BIS. The probable sources for sulfate are discharge of untreated domestic and industrial waste [58]. The PO_4^{3-} concentrations are observed in the range from 3 to 30 mg/L with an average value of 15 mg/L. These values are within the range of WHO/BIS standards. The observed uranium concentration is in the range 0.1 to 13.1 $\mu\text{g/L}$ with an average value of 2.5 $\mu\text{g/L}$ which is well below the maximum permissible limit suggested by the AERB and WHO [13, 49]. The lower amount of uranium observe might be due to leaching of uranium from adjoining / host aquifer granite rich rock formations. The anthropogenic activities, urbanization, and excessive use of chemical-based fertilizers and pesticides, which are accountable for the upsurge in the TDS of the region, might be another cause. However, generally, it seems that the probable source of uranium observed in the study area may be geogenic in nature. The observed results of uranium distribution in the current study are compared with local, state, national, and international levels already reported in the literature and the comparison is presented in Table 2. It has been found that the observed level of uranium in drinking water at the local level is much lower than the reported value [38, 39, 59] represented in Table 2. The total hardness of all collected groundwater samples ranged between 95 to 1090 mg/L with an average value of 499 mg/L. Around 45% of the total sample crossed the permissible limits suggested by WHO [13] i.e. 500 mg/L. In the current study the order of ions in the groundwater is $\text{Na}^+ > \text{Ca}^{2+} > \text{Mg}^{2+} > \text{K}^+$ and $\text{HCO}_3^- > \text{SO}_4^{2-} > \text{Cl}^- > \text{NO}_3^-$, respectively. The Na^+ ion concentrations in the study area vary from 34 to 261 mg/L with an average of 88 mg/L. All samples are observed in the permissible limit by WHO [13] except one. The ionic concentration of Ca^{2+} ranges from 16 to 186 mg/L had an

Table 2 Uranium distribution at state, national and international level in groundwater

States	Different districts	Uranium concentration range (µg/L)	References
<i>Within the country</i>			
Maharashtra	Aurangabad	0.1 to 16.7	[60]
	Beed	0.03 to 32.85	[38]
	Nagpur	2.7 to 302.7	[61]
	Buldhana	0.10 to 13.06	Current investigation
Punjab	Bathinda	0.5 to 571.7	[62]
	Mansa	0.13 to 1340	[63]
	Amritsar	0.6 to 65.3	[64]
Uttar Pradesh	Khalilabad, Gorakhpur, Maharajganj, and Kushinagar, and Aligarh	0.20 to 64.0	[65]
Assam	Guwahati	0.08 to 5.32	[66]
Karnataka,	Kolar	0.3 to 1442.9	[67]
Haryana	Hisar, Mahendragarh, Panipat, Sonipat	0.39 to 290	[68]
<i>At global level</i>			
Korea (Okchun belt)		0.5 to 263	[69]
Nigeria (Ogun state)		20 to 267	[70]
Germany		0.001 to 16.0	[71]
France		0.18 to 37.2	[72]
Argentina		0.04 to 11.0	
Hungary		0.08 to 75.3	
Finland		0.04 to 12,097	
Southern Finland		6 to 3400	[73]
Switzerland		0.05 to 92.02	[74]
Morocco		4.46 to 308.4	[75]

average value of 85 mg/L. The Mg^{2+} ions concentration is in the range between 19 and 109 mg/L with a mean value of 55 mg/L. The K^+ and HCO_3^- ionic concentrations are observed in the range 0.1 to 21 mg/L and 180 to 1050 mg/L, it is found that all the sampling locations fall under the safe category.

Pearson's correlation

To assess the correlation between the water quality parameters ($n = 17$) and uranium concentration, the Statistical Package for the Social Sciences (SPSS) software was used. The main reason for chose Karl Pearson's correlation method for the correlation matrix because the water quality data is normally distributed. The extracted correlation data from the software is presented in Table 3. Based on extracted data a strong positive correlation was observed between uranium to TDS, EC, and chloride with a 0.01 confidence level and it is suggesting that groundwater samples having high TDS contents denotes more tendency to interfere with uranium concentration with ionic species [76]. Also, it was observed that uranium and bicarbonates are strongly correlated with each other (0.01 confidence level) due to the possibility of the formation of a uranyl bicarbonate derivative [77]. TDS

and EC are strongly positively correlated with each other. pH with ORP, DO to fluoride, chloride to nitrate, and sulfate to phosphate are negatively correlated with 0.01 confidence level. The ORP is significantly correlated with fluoride and potassium. It is suggesting that OPR is playing a significant role in groundwater quality and affect the movement of contaminant species. A weak positive correlation was observed in uranium to ORP, calcium, magnesium, and bicarbonate ions. This finding itself suggesting that the existence of uranium may be due to the soluble mineral salts as well as an anthropogenic source like excessive use of phosphate-containing fertilizers, the same finding is reported by many researchers [63]. Based on the result it is very important to minimize the use of chemical-based fertilizer and discharge the radioactive waste into water bodies. The calcium and magnesium ions also positively correlated with each other. It is suggesting that the weathering of bedrock is the main source of the cations. The nitrate to hardness, calcium was shown a weak negative correlation.

Radiological risk estimation

The radiological risk assessment due to intake of uranium through drinking water for adults is performed using the

term Excess Cancer risk (ECR) and presented in Table 4. The ECR was calculated from Eq. 2. An estimated ECR value for adults is observed well below the permissible limit suggested by AERB i.e. $1.67 \times 10^{-4} \mu\text{g kg}^{-1} \text{day}^{-1}$ [49], hence on observed values there no excess cancer risk due to intake of uranium contained water.

Assessment of chemical risk

The chemical risk expressed as Lifetime Average Daily Dose (LADD) due to uranium in groundwater is calculated by using Eq. 4. The observed values are presented in Table 4 and vary from 6.12×10^{-3} to $7.99 \times 10^{-1} \mu\text{g kg}^{-1} \text{day}^{-1}$ with an average $1.51 \times 10^{-1} \mu\text{g kg}^{-1} \text{day}^{-1}$ for adult. The mean values of LADD are well below the permissible limit i.e. $4.53 \mu\text{g kg}^{-1} \text{day}^{-1}$ recommended by AERB [49]. To assess the chemical toxicity, Hazard Quotient was also calculated to estimate the threat that might happen due to the consumption of uranium-containing drinking water to the human body. The estimated values of HQ are observed in ranges 1.35×10^{-3} to $1.76 \times 10^{-1} \mu\text{g kg}^{-1} \text{day}^{-1}$ with mean 3.34×10^{-3} for adult. According to the mean observed value of HQ for all age groups, it showing much less than

the recommended value by health agencies and representing that water of the study area is fit for drinking purposes [49].

Annual Effective Dose (AED)

Annual effective dose (AED) is calculated for all age groups and presented in Table 5. AED is varying for all age groups due to daily water intake. The mean AED for all life stage groups were found to be lower than the recommended value of $100 \mu\text{Sv y}^{-1}$ [13]. Based on the calculated values of LADD, HQ, and AED presenting the groundwater of the study region contained a trace amount of uranium, and it does not pose any harmful impact on human health.

Spatial distribution of uranium

In the present study for predicting the correlation among the water quality parameters and spatial distribution patterns, the interpolation method (Inverse distance weighted) is applied. The main purpose to choose the Inverse distance weighted (IDW) interpolation method is that this method is a deterministic estimation method where values at unknown locations are determined by a linear combination of values at nearby known locations. This method

Table 4 Extracted statistical data of chemo-radiological risk of uranium in the study area

Life stage group	Statistical entity	Uranium ($\mu\text{g/L}$)	Conc. of U in (Bq/L)	ECR (Mortality) $\mu\text{g kg}^{-1} \text{day}^{-1}$	ECR (Morbidity) $\mu\text{g kg}^{-1} \text{day}^{-1}$	(LADD) $\mu\text{g kg}^{-1} \text{day}^{-1}$	HQ $\mu\text{g kg}^{-1} \text{day}^{-1}$
Adult	Min	0.10	0.00	5.71E-11	9.30E-11	6.12E-03	1.35E-03
	Max	13.06	0.33	7.46E-09	1.21E-08	7.99E-01	1.76E-01
	Mean	2.48	0.06	1.41E-09	2.30E-09	1.51E-01	3.34E-02
	Median	1.77	0.04	1.01E-09	1.65E-09	1.09E-01	2.40E-02

Table 5 Age-dependent annual effective doses of the collected water sample

Life stage group	Age group (Years)	DWI ($\text{L}^{-\text{day}}$)	Min ($\mu\text{Sv y}^{-1}$)	Max ($\mu\text{Sv y}^{-1}$)	Average ($\mu\text{Sv y}^{-1}$)
Infants	0–0.5	0.7	2.8E-08	3.7E-06	7.0E-07
	0.6–1	0.8	3.2E-08	4.2E-06	8.0E-07
Children	1 to 3	1.3	5.3E-08	6.9E-06	1.3E-06
	4 to 8	1.7	6.9E-08	9.0E-06	1.7E-06
Males	9 to 13	2.4	9.7E-08	1.3E-05	2.4E-06
	14 to 18	3.3	1.3E-07	1.7E-05	3.3E-06
	Adult > 19	3.7	1.5E-07	2.0E-05	3.7E-06
Female	9 to 13	2.1	9.3E-08	1.2E-05	2.3E-06
	14 to 18	2.3	9.3E-08	1.2E-05	2.3E-06
	Adult > 19	2.7	1.1E-07	1.4E-05	2.7E-06
Pregnancy	14 to 18	3	1.2E-07	1.6E-05	3.0E-06
	19 to 50	3	1.2E-07	1.6E-05	3.0E-06
Lactation	14 to 18	3.8	1.5E-07	2.0E-05	3.8E-06
	19 to 50	3.8	1.5E-07	2.0E-05	3.8E-06

Table 3 Extracted data of Karl Pearson's correlation of water quality parameters of the study area

	pH	TDS	EC	ORP	DO	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ⁻	HCO ₃ ⁻	TH	Ca ²⁺	Mg ²⁺	Na ⁺	K ⁺	U	
pH	1																	
TDS	-0.157	1																
EC	-0.157	1.000**	1															
ORP	-.396**	0.117	0.117	1														
DO	-0.117	0.166	0.166	0.093	1													
F ⁻	-.396**	-0.055	-0.055	-.208*	0.03	1												
Cl ⁻	-0.026	.795**	.795**	0.117	0.086	.222*	1											
NO ₃ ⁻	0.193	-0.103	-0.103	-0.051	0.086	.222*	- .283**	1										
SO ₄ ²⁻	0.056	-0.015	-0.014	-0.051	-0.182	0.042	0.114	0.018	1									
PO ₄ ⁻	-0.143	.383**	.383**	-0.021	-0.051	-0.015	.311**	0.025	-0.087	1								
HCO ₃ ⁻	0.007	.686**	.686**	-0.023	-0.032	0.086	.532**	0.183	.215*	.381**	1							
TH	-0.055	.787**	.787**	0.131	0.034	0.014	.726**	- .223*	0.174	.381**	.567**	1						
Ca ²⁺	-0.055	.787**	.787**	0.131	0.034	0.014	.726**	- .223*	0.174	.381**	.567**	1.000**	1					
Mg ²⁺	0.007	.686**	.686**	-0.023	-0.032	0.086	.532**	0.183	.215*	.381**	1.000**	.567**	.567**	1				
Na ⁺	.305**	0.091	0.091	0.157	-0.033	.371**	0.17	-0.029	0.074	-0.082	0.021	.204*	.204*	0.021	1			
K ⁺	-0.135	0.163	0.163	.232*	0.047	0.052	0.123	-0.09	0.05	0	0.077	0.158	0.158	0.077	0.107	1		
U	-0.089	.555**	.554**	-0.029	0.155	0.048	.467**	-0.036	0.057	0.185	.433**	.458**	.458**	.433**	0.096	-0.012	1	

All values are in mg/L except pH, EC (µS/cm¹) ORP (mV), and U (µg/L)

**Correlation is significant at the 0.01 level (2-tailed)

*Correlation is significant at the 0.05 level (2-tailed)

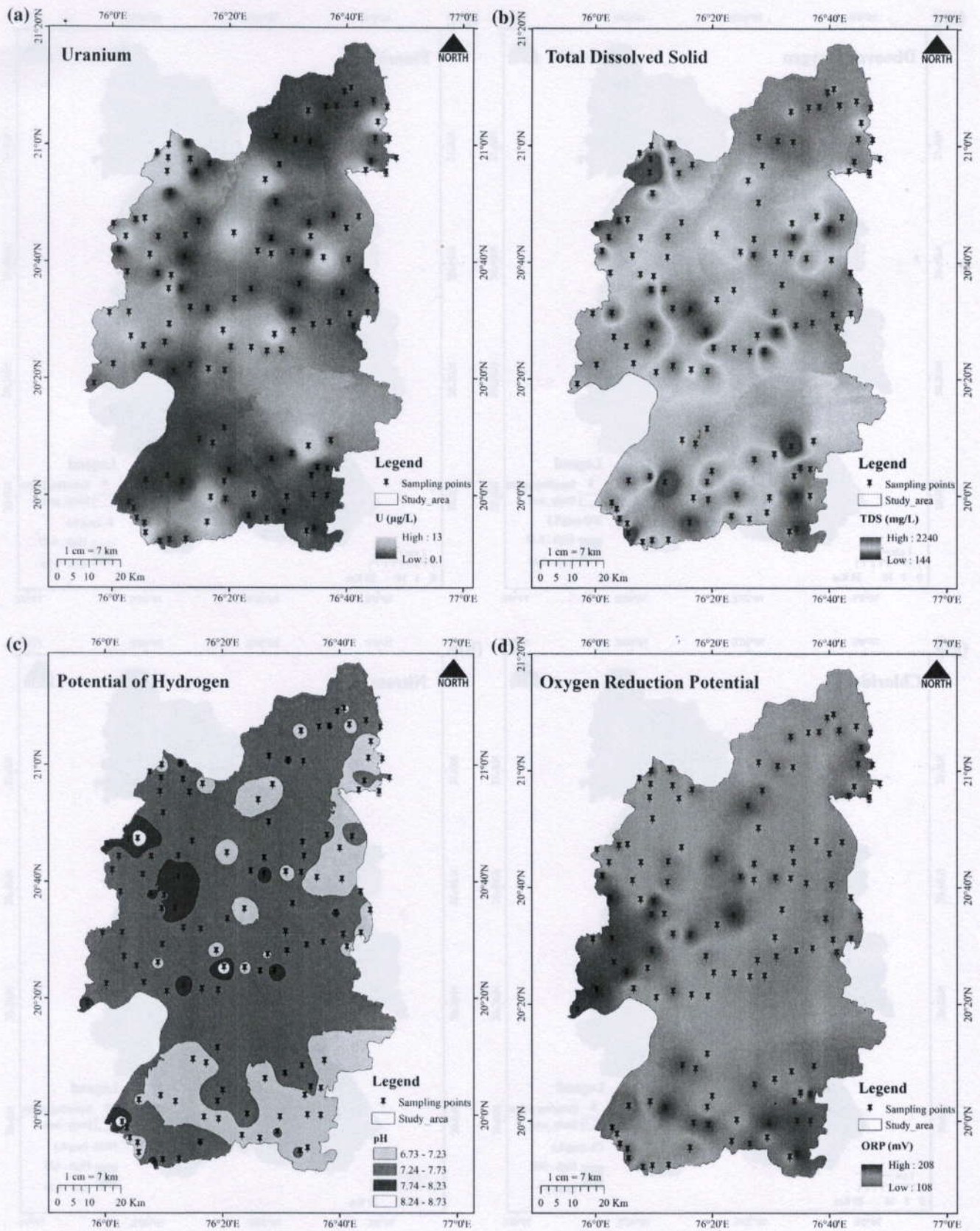


Fig. 3 a–p Spatial distribution maps of uranium along with water quality parameters

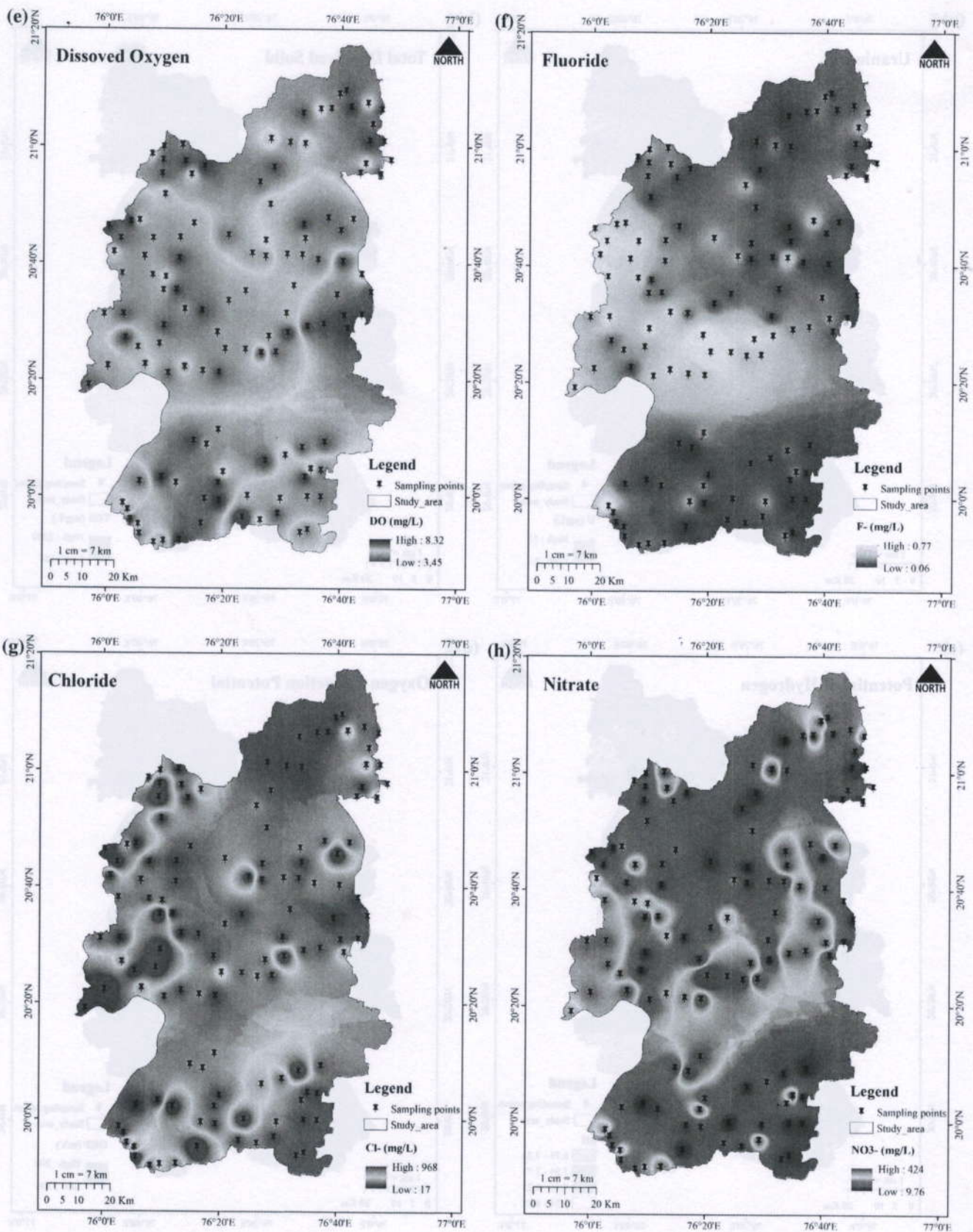


Fig. 3 (continued)

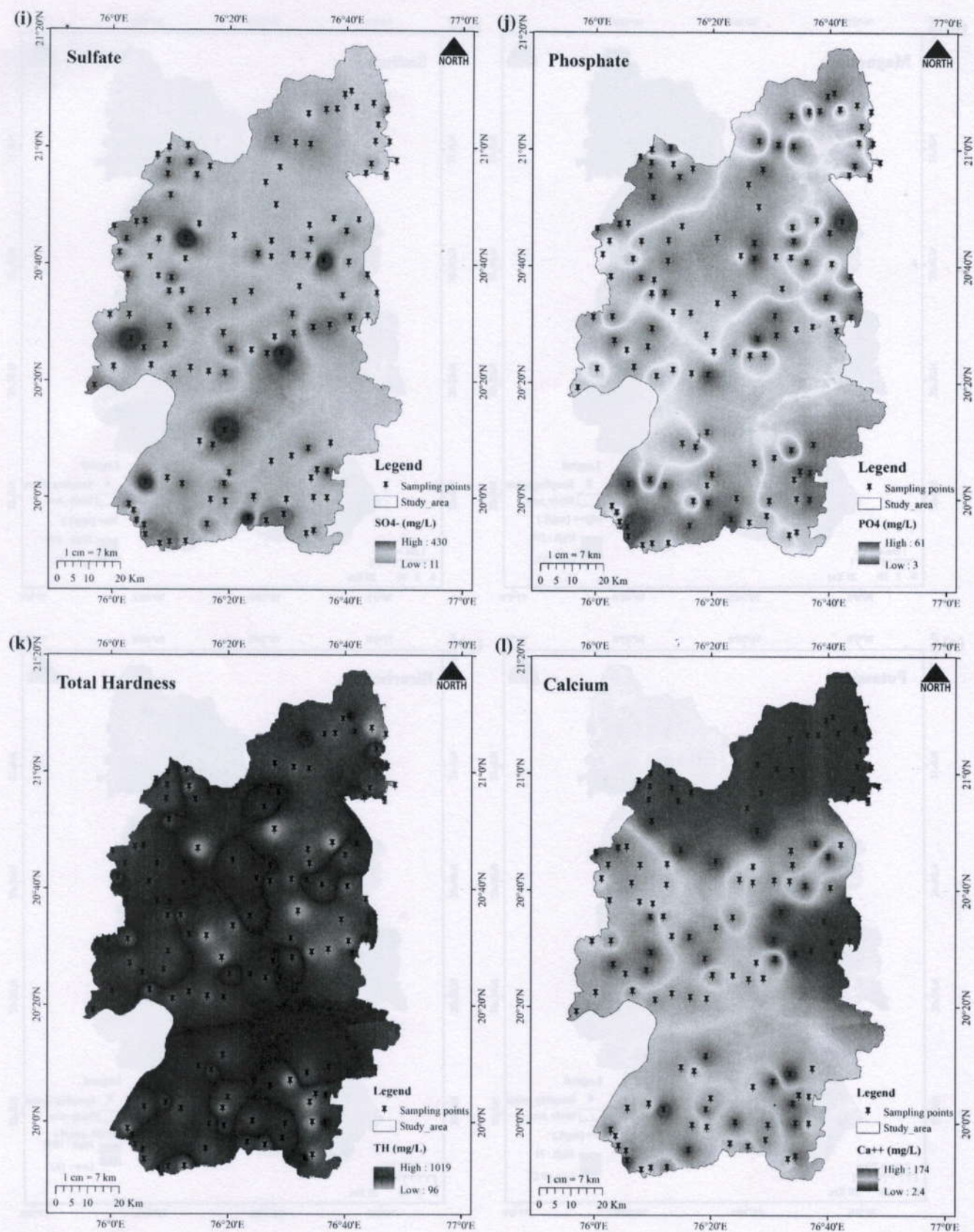


Fig. 3 (continued)

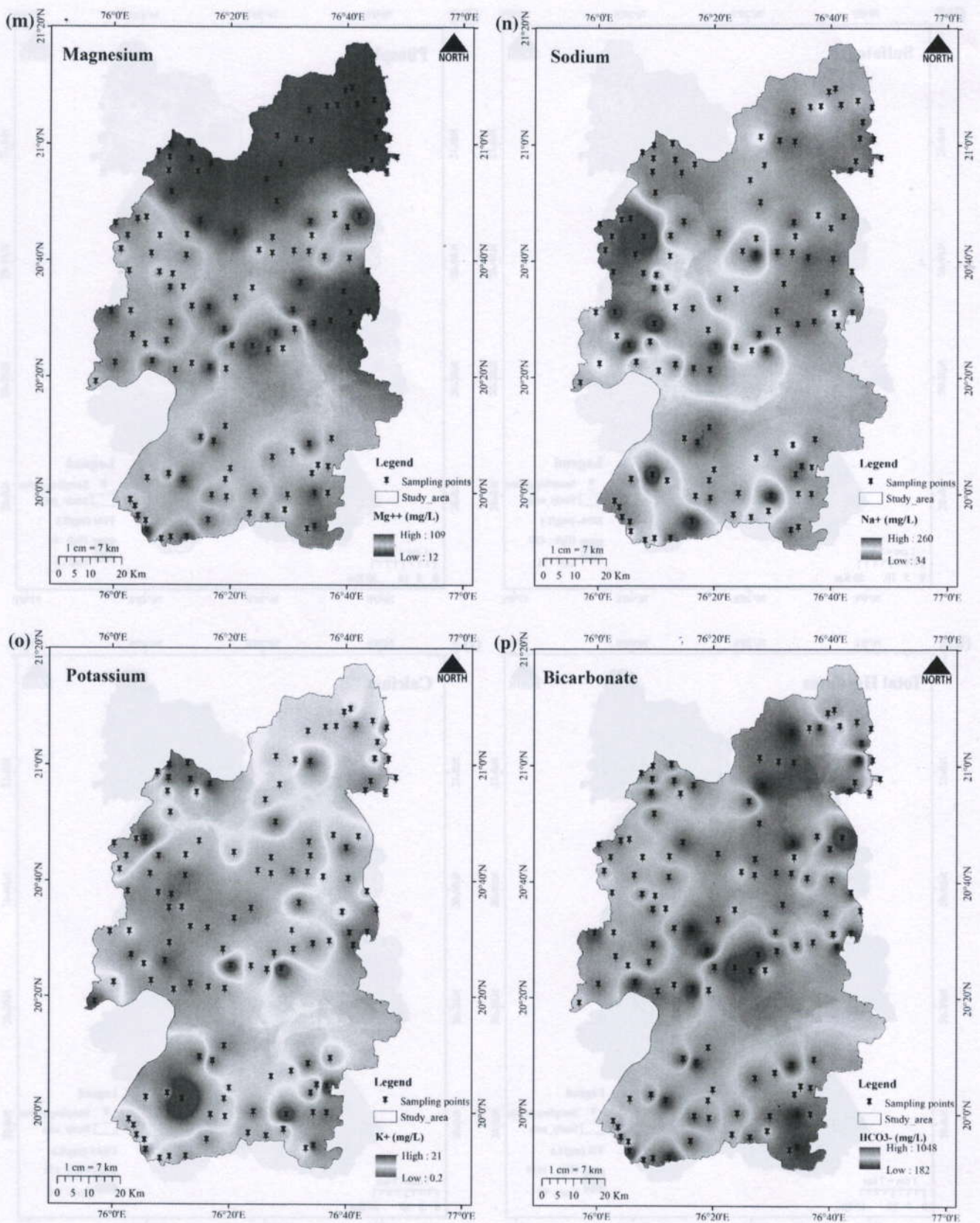


Fig. 3 (continued)

is helpful in visualizations of how the parameters look like over the study area. The resulting maps are shown in Fig. 3a–p that are showing the spatial distribution of water quality parameters throughout the study area. In the spatial distribution of uranium, there is color coding (green to yellow) to representing the lower to a higher concentration of uranium represented in Fig. 3a. In the case of other water quality parameters, the interpolation maps were presented from Fig. 3b–p.

Summary

In the current study, spatial distribution of uranium and associated water quality parameters and subsequent radiological and chemo toxicological dose due to uranium were assessed in the Buldhana district. The observed mean uranium concentration ($2.5 \mu\text{g/L}$) is well below the recommended level by national and international health agencies [13, 49]. The radio-chemo toxicological risk due to naturally occurred uranium for an adult is estimated in the form of ECR and LADD is observed well below the safe limit i.e. $1.67 \times 10^{-4} \mu\text{g kg}^{-1} \text{day}^{-1}$ and $4.53 \mu\text{g kg}^{-1} \text{day}^{-1}$ respectively is suggested by national and international health agencies like AERB and WHO. The lower amount of uranium is observed might be due to leaching of uranium from adjoining/host aquifer granite rich rock formations. The anthropogenic activities, urbanization, and excessive use of chemical-based fertilizers and pesticides, which are accountable for the upsurge in the TDS of the region, might be another cause. However, generally, it seems that the probable source of uranium observed in the study area may be geogenic in nature. It will be safe to state that the groundwater's from the study region are safe for drinking, domestic and irrigation purposes as for uranium associated risks are concerned.

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Compliance with ethical standards

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

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