

# Studies on Uranium Hydrogeochemistry in Kamalavathi River Basin, Gulbarga and Yadgir Districts, Karnataka

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## ABSTRACT

Uranium is a radioactive element and its presence in ground water, used for human consumption, forms a serious societal issue. The present study, deals with the occurrence and geochemistry of uranium in the aquifers underlying parts of Gulbarga and Yadgir districts of Karnataka State. The study area is confined to Kamalavathi river basin, where groundwater is the major source for domestic use. High uranium (>30 ppb) is found in the north eastern part of the study area. The present study deals with the geochemistry of uranium in ground water of Kamalavathi river basin. The data was subjected to statistical analysis to understand the inter relationship between various elements. The 2D and 3D maps of groundwater flow directions and uranium concentrations were used to understand the variation of uranium concentration along the groundwater flow. Detailed uranium geochemistry and Eh-pH diagrams were used to understand the lithological controls on occurrence of uranium in groundwater.

## INTRODUCTION

Quality of water plays an important role in the progress and development of the country as it has direct impact on health. Hence, it is extremely essential to sustain such natural resources present under the earth surface. Groundwater is the most widespread and accounts for nearly 50% of water used for irrigation, 80% is used for drinking purpose in several rural area and 50% of groundwater used for urban drinking and industrial needs (CGWB, 1977-78). High uranium concentrations are found in the groundwater of the study area, indicating high vulnerability of the local population to the ill-health effect. Understanding the uranium hydro-geochemistry may help in identifying the possible source of uranium concentrations. The groundwater flow directions are used to understand the variation of uranium concentration along the groundwater flow. In the present study, an integrated approach is used to understand the behavior of uranium in groundwater of study area and its impact on health.

## LOCATION OF STUDY AREA

The Kamalavathi river is a major tributary of Kagna river which is within the Bhima basin area. It flows in the northern direction and joins the Kagna river. The river basin is spread over two taluks, Sedam (Gulbarga district) and Yadgiri (Yadgiri district). Toposheet numbers that cover the area are 56 G/4, 56 G/8, 56 H/1 and 56 H/5. Study area falls within latitude of 16°53'00" to 17°13'00" and longitude 77°22'00" to 77°27'00", covering an area 540 sq. km (Fig. 1).

The main physiographic features in Kamalavathi river basin are hills and plains. The altitude ranges between 420 to 600 meters. The general slope of the area is towards north. The entire area is dissected by number of smaller streams and streamlets.

## GEOLOGY OF THE STUDY AREA

The study of geology of Bhima basin started in 1872 by King (Radhakrishna and Vaidyanadhan, 1994) with two fold classification. Later, Mahadevan (1947) classified the basin into three divisions. Presently the classification proposed by Jayaprakash (1999) reveals that the prominent litho-units of Bhima group are the limestone and shale, with a thin arenite and conglomerate at the base of the sequence exposed at several places marking the unconformity with the basement crystalline rocks (Radhakrishna and Vaidyanadhan, 1994). The study area occupies the eastern part of Bhima basin. As such geological exposures are scanty, except for a few which can be seen in the study area from Nachawar to Handarki road in the southwest region of Sedam, and to the north eastern side of Adki and Mudhol areas. Contacts between the rocks are obscured due to soil cover interspersed by vegetation and habitation in most of the area. Small mounds and highly elevated, irregular hillocks characterize an intrusive granitic rock type, which is considered as being equivalents to Closepet granite. Broadly, the studied area is characterised by basement granite, overlain by sandstone, shale, limestone and Deccan traps (Fig. 2). The sedimentary rocks of Bhima basin and the granitoids have been affected by intense faulting. Major faults across the basin define the structural boundaries of the different sectors (Kale and Peshwa, 1995).

## HYDROGEOLOGY OF THE STUDY AREA

The hydrogeological studies of the study area have been carried out by CGWB (2003-04). It is noted from the report that, groundwater in the study area occurs under phreatic as well as semi confined conditions in all geological formations. Whenever, these formations

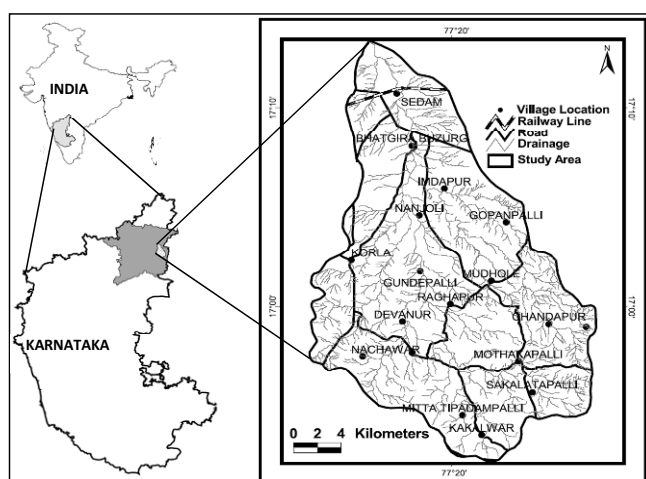


Fig.1. Location map of Kamalavathi river basin.

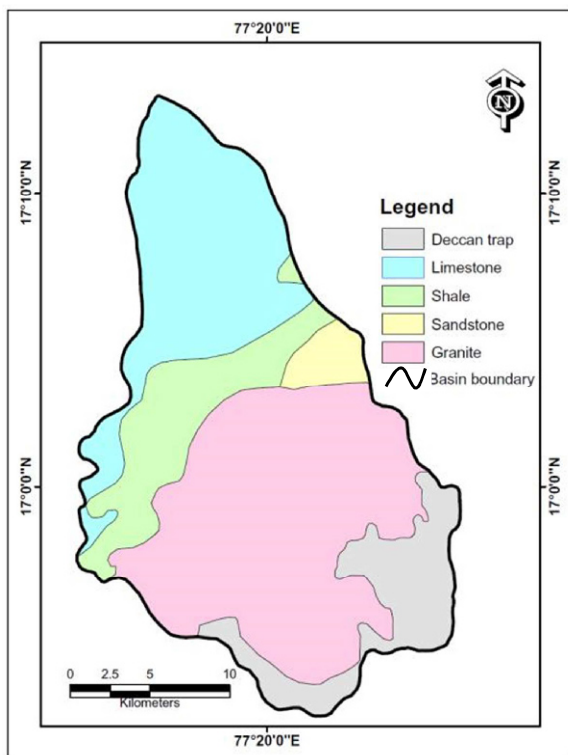


Fig.2. Geology map of Kamalavathi river basin

underlie the Deccan traps, ground water occurs under confined condition. The depth of weathering exceeding 20 m at places giving rise to good productive aquifers. The open wells are 4 to 15 m depth and yield upto 100 m<sup>3</sup>/day with sustain pumping of 1 to 5 hrs depending upon seasons. Borewells drilled at favorable locations may yield up to 5 lps. The presence of joints also helps in the occurrence and movement of ground water. Paleo-weathered beds are good for storing of groundwater (CGWB, AAP 2003-2004).

#### Depth to Water Level

The depth to water level (DTWL) varies from 2.47 m to 29.15 m bgl with an average of 8.60 m bgl in PRM<sup>1</sup>, 1.08 m to 29.2 m bgl with an average of 6.24m bgl in POM and 2.05 m to 25.82 m bgl with an

Table 1. Depth to water level in study area

DTWL (m bgl)	PRM <sup>1</sup>	POM	PRM <sup>2</sup>
<10	59(78%)	66(87%)	59(78%)
10 to 20	13(17%)	8(11%)	16(21%)
20 to 30	4(5%)	2(3%)	1(1%)

average of 7.76 m bgl in PRM<sup>2</sup>. In PRM<sup>1</sup> 78% of the water levels are less than 10m bgl, 17% are between 10 to 20 m bgl and 5% of water levels are 20 to 30m bgl. In POM 87% of the water levels are less than 10m bgl, 11% are between 10 to 20 m bgl and 3% of water levels are 20 to 30m bgl. In PRM<sup>2</sup> 78% of the water levels are less than 10m bgl, 21% are between 10 to 20 m bgl and 1% of water levels are 20 to 30m bgl.

#### Groundwater Flow Directions

The water table contours are used to generate groundwater flow lines using Surfer software. The 2D maps of water table along with groundwater flow directions have been generated to understand the nature of groundwater flow directions for the study area. It is observed from the 2D maps (Fig. 3a, 3b and 3c) that the groundwater flow directions show similar trend in all the three seasons, irrespective of seasonal variations. The general groundwater flow directions are towards north in all seasons. However, westerly and easterly flow directions with certain convergent areas are observed which may be due to the local structural controls and certain favorable configuration of the basin topography.

#### Recharge Conditions

The input component considered in GEC 1997 are rainfall recharge, recharge from canals, and from surface and ground water irrigation, from tanks and ponds and from water conservation structures. In the study area the recharge is considered mainly through rainfall infiltration, field irrigation and tanks.

#### DATA USED AND METHODOLOGY

The groundwater samples were collected in three different seasons to understand the seasonal variation of hydro-geochemistry in the study area. A total of 78 groundwater samples were collected in pre-monsoon, April-2010 (PRM<sup>1</sup>), 75 samples in post-monsoon, January-2011 (POM) season and 75 samples were collected in pre-monsoon, June-2011 (PRM<sup>2</sup>). Locations of wells were chosen to have a good

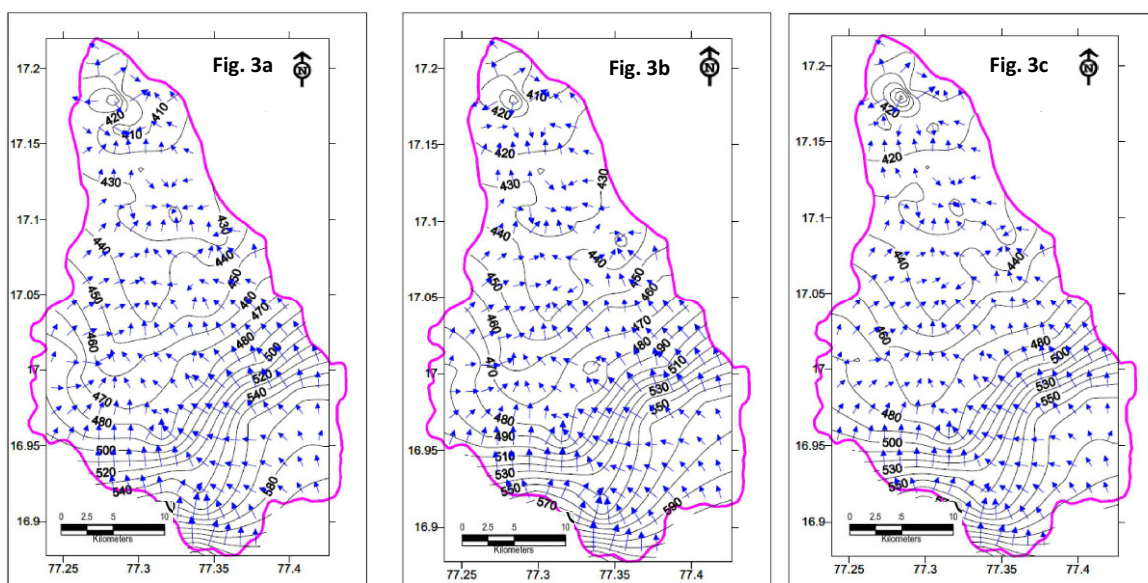


Fig.3a-c. Groundwater flow direction map.

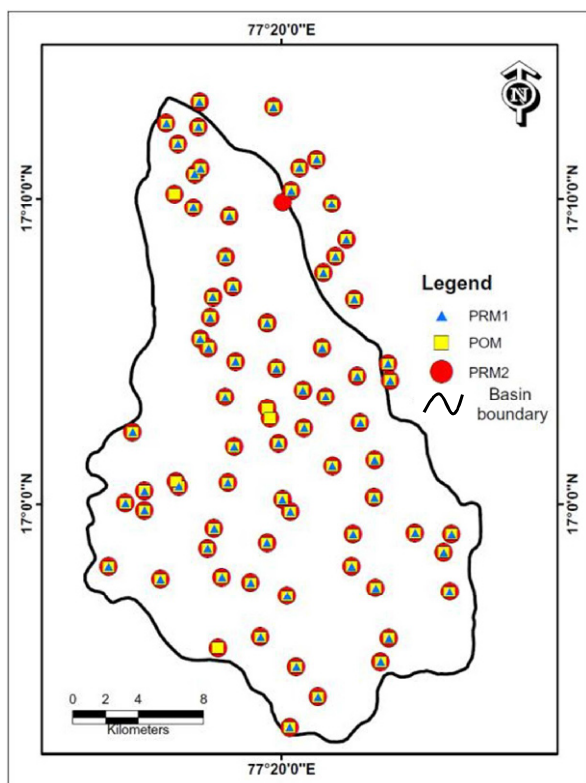


Fig.4. Sample location map

spread over the entire area (Fig. 4). The data is collected from the all available borewells of each villages in the study area. The density of the sampling is 1 sample for every 14 sq.km. All deep borewells are chosen for groundwater sample collection representing the deeper aquifer systems. In-situ measurements of temperature, pH, salinity, electrical conductivity, total dissolved solids and Eh were measured for groundwater samples using Systronics field analysis kit. Water samples were collected in 1000 ml fresh polythene bottles for three different seasons from the borewells as well as from open wells. The samples for uranium analysis are collected in separate 500 ml polythene bottle. The samples collected for uranium are filtered with 0.45micron filter paper and acidified with 0.5ml of concentrated hydrochloric acid to prevent its adsorption onto the container wall. These samples were analyzed to determine the diverse range of major ionic constituents as per the APHA (1995) guidelines. Different instruments such as flame photometer, UV Spectrophotometer, Hach colorimeter were used along with regular volumetric (titration) methods, to analyze the geochemical constituents of samples. The statistical analysis of major ionic constituents was carried out to summarize the general composition of the groundwater samples. Hydrogeochemical data were brought into GIS platforms. Different softwares such as, Map Info Professional 8.5, Arc GIS 9.1, Surfer8 and SPSS were used to generate hydro-geochemical information system. Geochemical simulation model software WATEQ4F, WATCLAST etc., were used to determine the solubility and equilibrium.

## RESULTS AND DISCUSSIONS

### Hydrogeochemistry

The general water chemistry is studied through statistical analysis (Range, Mean, Median, Mode and Standard deviation) of results of chemical analysis of groundwater samples for major ionic constituents in the study area for all the three seasons. The results of chemical analysis is checked for their consistency and given in the Table 2. The groundwater temperature in the study area varies from 28.4 to 38.7°C

in PRM<sup>1</sup>, 27 to 31°C in POM and 27.7 to 30.4°C in PRM<sup>2</sup> seasons. The pH of the groundwater samples varies from 7.87 to 11.0 in PRM<sup>1</sup>, 8 to 11 in POM and 7.6 to 11.12 in PRM<sup>2</sup> season indicating the alkaline nature of groundwater in the study area. The EC and TDS is found higher during both premonsoon (PRM<sup>1</sup> and PRM<sup>2</sup>) seasons as compared to postmonsoon season. The Eh values varies from -254 to -0.75 in PRM<sup>1</sup>, -249 to -21 in POM and -154 to -28 in PRM<sup>2</sup> seasons indicating the reduced conditions of the groundwater in study area. The major constituents are well within the permissible limit of drinking purpose and show an increase or decrease in concentrations

Among the different seasons depending on dilution or precipitation of elements. The fluoride in the study area is found to higher than the permissible limit (1.5 mg/L) in all the three seasons. Uranium in the groundwater samples of the study area is found to be more than permissible limit (30ppb) in some villages in north eastern part of study area. The list of villages showing higher uranium concentration in groundwater is given in Table 3.

### Uranium Geochemistry

Uranium (a primordial radionuclide) occurs in a dispersed state in the earth's crust. Uranium salt is the most soluble of the long-lived radionuclides and forms ions with oxidation states of +4 (UO<sub>2</sub> and U<sup>4+</sup>) and +6 (UO<sub>3</sub> and UO<sub>2</sub><sup>2+</sup>) (Banks et al., 1995). Uranium will bond with oxygen to form the uranyl ion, or uranium dioxide, which is soluble in ground water under aerobic conditions. As uranium is a natural lithophile element and is contained almost in all natural waters and its concentration in groundwater depends on lithology, geomorphology and other geological conditions of the region (Sridhar Babu et al., 2008). Uranium present in the earth, migrates to water, plants, subsequently reaching human beings. Uranium has both chemical and radiological toxicity on organs of human beings viz., kidneys and lungs.

The uranium concentration in groundwater of the study area varies from < 1 ppb to 800 ppb in PRM<sup>1</sup>, < 1 ppb to 760 ppb in POM and < 1 ppb to 473 ppb in PRM<sup>2</sup> seasons. The concentration of uranium in some of the wells is higher than the permissible limit (<30ppb) prescribed by United State Environmentatl Protection Agency (EPA – US, 2003) (Fig. 5).

The spatial distribution map of uranium is presented to understand the variation of uranium concentration in the study area. Anomalous values of uranium in groundwater are limited to a small area while a major part of the area is under permissible limit (Fig. 6a-c).

Spatial distribution of Uranium on a 3D map, along with the map of groundwater flow directions (Fig. 7a-c), indicate that the concentrations are low(below permissible limit of EPA) in southern, western and central region of the study area, while higher concentrations are seen only in north eastern parts. Hence, there is no significant increase observed along the groundwater flow direction. The observed higher concentration in the north eastern part could be due to the structural connectivity across the boundary of the study area.

High uranium concentrations are found in the north eastern part of the basin only during one seasonal data and in the rest, the values have remained low. The seasonal variations are observed in the uranium concentration showing the dilution effect from PRM<sup>1</sup> through POM to PRM<sup>2</sup>.

### Rock - Water Interaction

The stoichiometric approach is adopted to understand the process of mineral and ionic chemistry of groundwater in the study area with respect to uranium. The uranium compounds that are likely to precipitate with the present water composition are, rutherfordin (UO<sub>2</sub>(CO<sub>3</sub>), schoepite (UO<sub>2</sub>(OH)<sub>2</sub>), uranophane (Ca(UO<sub>2</sub>)<sub>2</sub>(SiO<sub>3</sub>)<sub>2</sub>(OH)<sub>2</sub>·5H<sub>2</sub>O and gummite etc., which are determined using

**Table 2.** Statistics of water chemistry from the study area

Variable	Range			Mean			Median			Mode			Standard Deviation		
	PRM <sup>1</sup>	POM	PRM <sup>2</sup>	PRM <sup>1</sup>	POM	PRM <sup>2</sup>	PRM <sup>1</sup>	POM	PRM <sup>2</sup>	PRM <sup>1</sup>	POM	PRM <sup>2</sup>	PRM <sup>1</sup>	POM	PRM <sup>2</sup>
Temp(°C)	28.40 to 38.70	27 to 31	27.7 to 30.4	30.98	28.9	28.1	30.75	28.9	28.7	30.40	28.5	28.4	1.72	0.82	0.64
pH	7.87 to 11.0	8.0 to 11.0	7.6 to 11.12	9.28	8.9	9.13	9.31	8.8	9.05	9.36	8.08	8.54	0.66	0.59	0.80
EC(μS)	120 to 8580	246 to 5370	331 to 6090	1518.31	1390.2	1412	1130.00	1170.0	1170	1490.00	1500	1170	1378.94	820.74	882
TDS(mg/l)	159 to 5360	132 to 2830	171 to 3190	810.44	770.9	736	590.00	660.0	610	640.00	660	730	777.57	442.91	465
Eh(mV)	-0.75 to -254	-21 to -249	-28 to -154	-87.36	-82.5	-87	-77.00	-72.0	-89	-83.00	-45	-104	52.20	47.91	26.5
Ca(mg/l)	30 to 320	20 to 470	10 to 270	106.64	111.6	90.9	100.00	90.0	80	90.00	30	80	38.46	91.59	50.7
Mg(mg/l)	10 to 180	10 to 290	20 to 440	46.83	130.5	151	40.00	120.0	140	30.00	100	140	28.18	63.47	87.9
Na(mg/l)	12.7 to 196.10	36 to 227	0 to 205	83.03	116.4	90.4	68.20	107.2	81.9	-	50	0	48.87	50.37	53.5
K(mg/l)	0.2 to 161	0 to 207	2.3 to 137	14.58	26.4	17.7	5.15	19.0	8.2	1.20	18	3.7	28.04	37.59	25.5
Cl(mg/l)	27.23 to 889.37	50 to 670	0 to 470	175.02	205.3	132	145.38	179.9	140	145.38	179.94	0	122.17	130.50	135.9
CO <sub>3</sub> (mg/l)	10 to 170	0 to 220	0 to 260	55.10	66.7	51.2	50.00	60.0	40	50.00	40	0	29.87	57.64	63.7
HCO <sub>3</sub> (mg/l)	100 to 690	220 to 730	210 to 880	353.23	394.3	443.2	360.00	360.0	430	360.00	350	330	83.91	117.39	140
SO <sub>4</sub> (mg/l)	0 to 18.75	0 to 22	3.5 to 229.4	4.15	5.8	54.5	2.47	3.3	32.5	0.00	1.466	-	4.72	5.72	54.6
Fe(mg/l)	0 to 3.96	0 to 1	0 to 7.13	0.14	0.2	0.29	0.02	0.2	0.005	0.00	0.194	0	0.53	0.12	0.95
NO <sub>3</sub> (mg/l)	0 to 16	0 to 36	-	3.79	16.4	-	2.55	18.5	-	1.60	0	-	3.89	14.20	-
F(mg/l)	0.17 to 8.68	0 to 9	0.48 to 9.12	1.48	1.4	1.75	1.14	1.2	1.59	2.30	1.38	3.2	1.37	1.20	1.17
Uranium (ppb)	1 to 800	1 to 760	1 to 473	28.4	18.4	15.08	4	4	5	1	1	3	122.6	87.4	54.7

**Table 3.** List of villages showing higher uranium in groundwater

Sl. No.	Village Name	PRM <sup>1</sup>	POM	PRM <sup>2</sup>
1	Ashanagar, CCI	800	760	473
2	Bidarched	42	41	34
3	Gopanpalli	32	35	31
4	Mudhol cross	-	37	37
5	Kurkunta	78	-	-
6	Huda	720	-	-
7	Indiranagar	-	59	73
8	Kadtal	-	35	-
9	Rangwar road	-	31	-

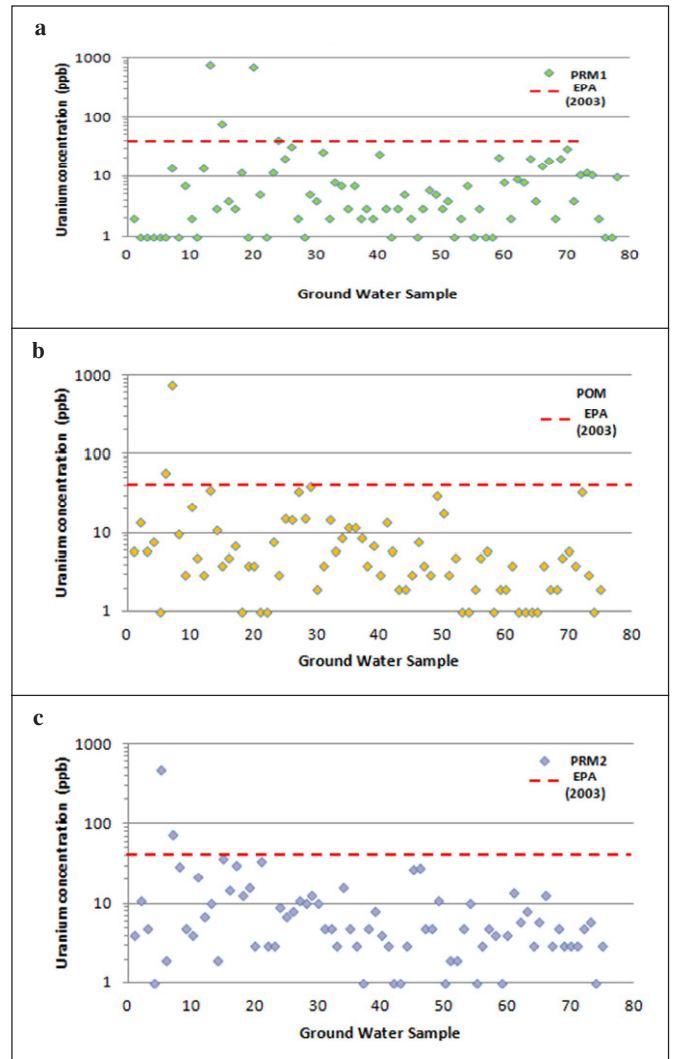
WATEQ4F software. These compounds show a negative value of saturation indicating, the uranium compounds are in state of under-saturation (Table 4).

**Lithological Controls on Uranium Geochemistry**

Uranyl carbonate shows considerable solubility in sodium carbonate solution, indicating the formation of stable complex ion (Blake et al., 1956). Considering sodium-uranium-carbonate balance and the neutral pH, the presence of bicarbonate, bicarbonate-carbonate or hydroxyl-carbonate complexes seem unlikely. Polymerize hydroxyl bicarbonate complexes however cannot be excluded. These would be isomeric with hydrated carbonate complexes and the data are not

**Table 4.** Saturation Indices of Uranium minerals

Minerals	PRM <sup>1</sup> Range		POM Range		PRM <sup>2</sup> Range	
	Min	Max	Min	Max	Min	Max
Gummite	-9.127	-3.745	-9.634	-4.284	-10.078	-6.068
Rutherfordin	-9.09	-1.092	-7.458	-1.57	-7.204	-3.479
Schoepite	-3.752	0.931	-4.143	1.202	-4.571	-0.6
B-UO <sub>2</sub> (OH) <sub>2</sub>	-3.841	0.834	-4.248	1.097	-4.679	-0.702
UO <sub>3</sub> (c)	-6.496	-1.851	-6.965	-1.616	-7.404	-2.016
Uranophane	-	-	-6.272	3.354	-4.961	-2.916



**Fig.5a-c.** Uranium concentration in groundwaters of study area

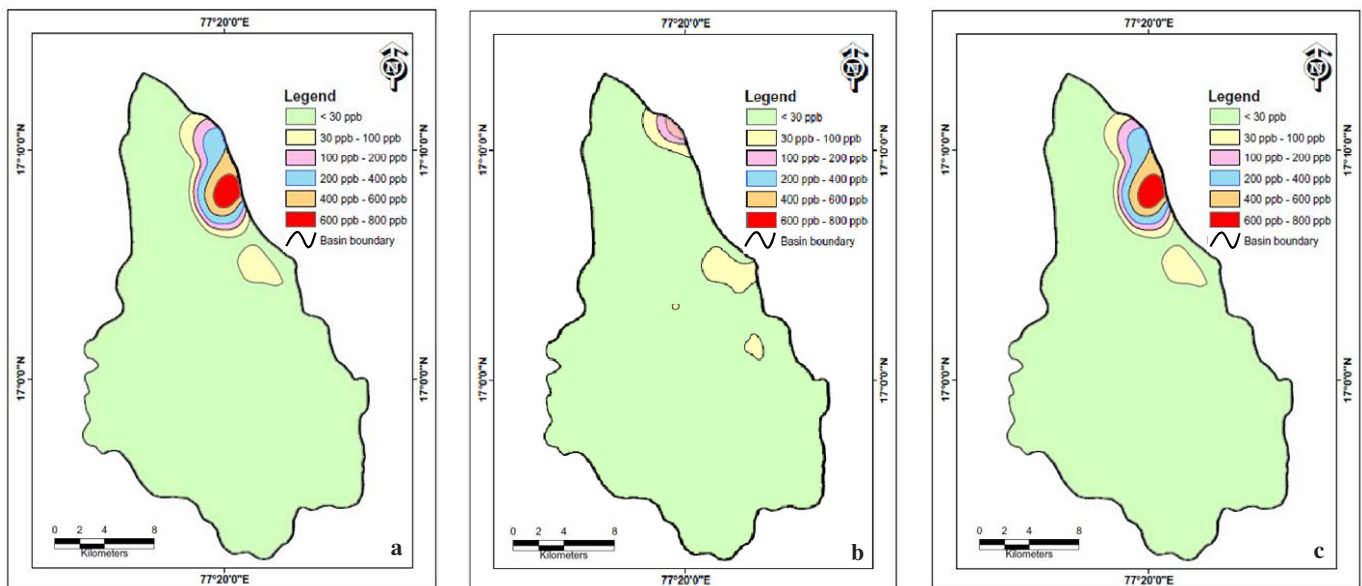


Fig.6. Spatial distribution map of U (a) PRM<sup>1</sup>. (b) POM. (c) PRM<sup>2</sup>.

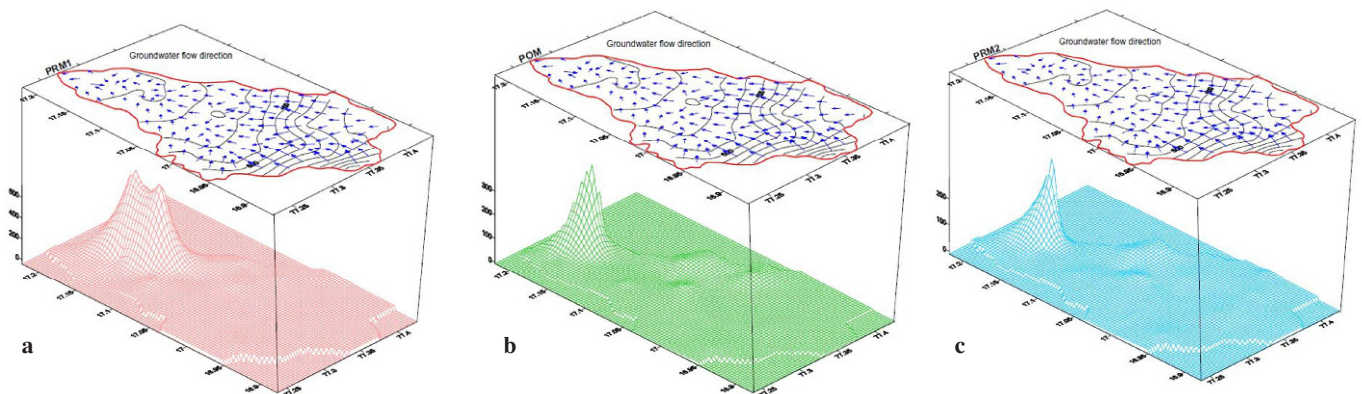
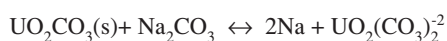
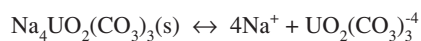


Fig.7. 3D map of uranium concentration (ppb) with groundwater flow (a) PRM<sup>1</sup>. (b) POM. (c) PRM<sup>2</sup>.

sufficient to distinguish between the two types. For simplicity the complex ion may be considered as  $\text{UO}_2(\text{CO}_3)_2^{-2}$  with the corresponding expression for the dissolution of uranyl carbonate in sodium carbonate solutions.



A dissolution reaction for sodium uranyl tricarbonate may be written as,



The reaction shows the dissociation from tricarbonate to bicarbonate as,



### Eh-pH studies

The radioactivity of uranium in groundwater is highly variable and is dependent upon the conditions of formation of the water and its geochemical nature (Bhimasankaran, 1974). The conditions of oxidation and reduction have a great bearing in determining the amount of uranium that can be held in solution by groundwater.

The solubility of uranium in the groundwater is very sensitive to change in redox conditions. Under oxidizing conditions, the uranium content of groundwater varies from  $n \times 10^{-6}$  to  $n \times 10^{-5}$  gm/l whereas in reducing conditions the uranium may be found in the order of  $10^{-7}$  gm/l (Bhimasankaran, 1974).

The variations in Eh – pH values are studied to understand the speciation. Using Eh – pH diagram, reported by Langmuir (1978), the water samples of the study area are found to be stable under carbonate complexes  $[\text{UO}_2(\text{CO}_3)]^4$ . This means that uranium circulation in ground waters is controlled by higher pH values of the study area. Eh – pH diagrams also support the view that uranyl complex is likely to be under carbonate species (Fig.8).

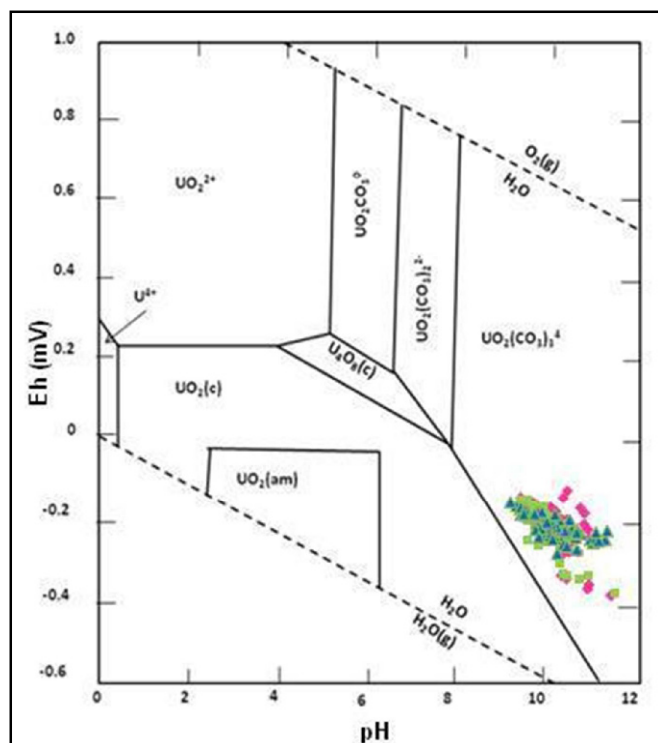
According to Boberg and Runnels (1971), Eh is always positive near an ore deposit but becomes increasingly negative as the distance increases from the ore deposit. Near the ore deposits the groundwater is always found to be in acidic condition with less pH values and vice versa.

### Statistical Analysis

The major-ion chemistry of the groundwater is controlled by the mineral dissolution resulting in enrichment of uranium in groundwater. The SPSS software is used to get the results of statistical analysis for the groundwater samples of the study area to understand the inter relationship of uranium with other chemical constituents of the groundwater in the study area. In PRM<sup>1</sup>, Uranium is showing positive correlation with T, Eh, Ca, Na, alkalinity, Cl,  $\text{SO}_4$  and Total Alkalinity, whereas negative correlation is found with pH, EC, Mg,  $\text{CO}_3$ , K and Fe. In POM positive correlation is found with Eh, EC, TDS, Ca, Na, K,  $\text{SO}_4$  and Fe whereas, negative correlation is observed between T, pH, Mg,  $\text{CO}_3$ , Cl and Total alkalinity. In PRM<sup>2</sup> a positive correlation

**Table 5.** Correlation coefficient of Uranium and other parameters in groundwaters of study area

	T	pH	Eh	EC	Ca	Mg	Na	K	CO <sub>3</sub>	HCO <sub>3</sub>	Cl	SO <sub>4</sub>	Fe	Alkalinity	Water Table
PRM <sup>1</sup>	0.223	-0.65	0.001	-0.037	0.68	-0.41	0.034	-0.084	-0.094	0.038	0.013	0.132	-0.039	0.004	-1.39
POM	-0.004	-0.051	0.050	0.026	0.53	-1.17	0.222	0.018	-0.05	-0.75	-0.90	0.051	0.001	-0.112	-0.118
PRM <sup>2</sup>	-0.98	0.83	-0.038	-0.151	-0.31	-0.98	0.194	-0.093	-0.037	-0.175	-0.173	0.102	0.404	0.112	-0.221



**Fig.8.** Eh – pH diagram for uranium in ground water of the study area.

is found between uranium and pH, Na, SO<sub>4</sub> and Fe and a negative correlation is found between Uranium and T, Eh, EC, Ca, Mg, K, CO<sub>3</sub>, HCO<sub>3</sub> and Cl and D.O (Table 5).

The groundwater table is also considered as a parameter to check the correlation with uranium and it is observed that negative correlation exist between uranium and groundwater levels in all three seasons (Table 5).

Correlation of uranium with other constituent and elements in natural waters proved to be commonly erratic or poor and is found to be varying for different season data.

## CONCLUSION

The groundwater in the study area are found to be in alkaline conditions. The Eh value indicate the reduced condition of the groundwater. The major ionic chemistry of groundwater is found to be well within the permissible limit for drinking purpose except Fluoride and Uranium. De-fluoridation can take care of fluoride issue in the study area. As the uranium is radioactive element, the present study was carried out to understand the uranium and its behavior in groundwater of the study area. The higher uranium in different seasons are found in north eastern parts of the study area. The Stoichiometric studies are carried out to know the saturation indices of uranium for all the three seasons and the results suggest that, with the present water composition the uranium compounds such as Rutherfordin, Schoepite, Uranophane and Gummite etc., are likely to form. The groundwater of study area are under saturation with respect to

uranium carbonate salts indicating the dissolution state of uranium minerals. The variation in concentration of uranium is may be due to dilution or concentration of groundwaters. Hence, in the present reduced environment of groundwater the uranium minerals are likely to be in dissolution form. The increase uranium concentration along the groundwater flow directions are also seen in present study indicating that the source of uranium is along the groundwater flow direction towards the upstream side of the study area as uranium deposit doesnot readily release the uranium into water under reduced conditions. Consumption of groundwater would not have a telling effect on human beings for over a very long use. The results of correlation of uranium with other ionic constituents does not serve the purpose of the study as there is no clear correlation observed. High Uranium content in groundwater alone does not serve as criteria for uranium prospecting and the study does not affiliated with any known source of uranium mineralization.

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