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Assessment of Age-Dependent Radiation Dose and Toxicological Risk of Uranium in Ground Water around Uranium Mines in Sikar, Rajasthan

Bhupesh Khyalia[a], Naresh Kumar[a], Rajesh Beniwal[a], Shakuntala[a], Amanjeet Panghal[a], Navish Kataria[b], Prikshit Gautam[c] & Ranjeet Dalal[a*]

[a]Centre of Radio Ecology & Department of Physics, Guru Jambheshwar University of Science and Technology, Hisar, Haryana 125 001, India

[b]Department of Environmental Science and Engineering, J.C. Bose University of Science and Technology, YMCA Faridabad, Haryana 21 006, India

[c]Department of Physics, Kirori Mal College, University of Delhi, Delhi 110 007, India

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Uranium concentration in forty-six groundwater samples collected from the region near the uranium deposits in the Royal village of Sikar district, Rajasthan, is determined using the LED fluorimetry technique. Age-dependent annual effective dose, radiological, and chemical toxicity risks are calculated to assess health risks associated with the groundwater for the people living in the nearby area. Uranium concentrations of different samples vary from 4.79–266.28 µg/L with an average value of 74.36 µg/L Overall 69% and 43% of samples exhibit uranium concentrations greater than recommended limits of WHO and AERB, respectively. Excess cancer risk values lie in the range from 1.36×10^{-5}-$7.54 \times 10^{-4}$and 2.10×10^{-5}-1.17×10^{-3} with mean values of 2.11×10^{-4} and 3.26×10^{-4} for mortality and morbidity, respectively. LADD value lies in the range of 0.35 µg/kg/day to 19.51 µg/kg/day with a mean value of 5.44 µg/kg/day. The hazard quotient value for 43.4% and 84.7% of samples is greater than unity according to AERB and WHO standards, respectively. The uranium retention and effective radiological dose to various body organs are estimated using biokinetic modelling. Physico-chemical parameters and their correlation with uranium concentration are also evaluated. A positive correlation is found between TDS and uranium concentration.

Keywords: Groundwater; LED Fluorimetry; Mining zone; Radiation; Uranium Concentration

1 Introduction

In India, people mainly depend on groundwater for drinking and irrigation due to its decentralized availability and easy accessibility. The groundwater is harnessed in large swathes of the countryside using hand pumps, tube wells, and open wells. When water flows through soil and rock formations, it amasses varying amounts of different compounds and minerals, including that of uranium[1]. The presence of uranium (which is geogenic in nature) in groundwater contaminates it and makes it unfit for various purposes. Anthropogenic activities also contribute to the rise in uranium levels in groundwater, like uranium mining, decrease in groundwater levels, use of pesticides and fertilizers, *etc*[2].

Uranium (U) is one of the heaviest naturally occurring radioactive materials, which is mainly found in bedrock. It is found in the earth's crust in the form of ^{238}U (99.27%),^{235}U (0.7%), and ^{234}U (0.005%)[3] isotopes. Uranium is naturally found in its oxide forms, such as uraninite and schoepite, and it is mainly found in its two oxidation states +4 and +6. Most of the uranium compounds with a +6-oxidation state as a uranyl ion (UO_2^{2+}) are soluble, while uranium compounds having tetravalent form are insoluble[4].

Due to the possible health hazards of uranium content in water, different health organizations place a safe limit value for uranium concentration in water. The World Health Organisation (WHO) has recommended that water having uranium concentration above 30 µg/L should not be used for drinking purposes[5]. In India, Atomic Energy Regulatory Board (AERB) has set this limit to 60 µg/L[6]. Humans are exposed to uranium mainly through food and water ingestion. It has been estimated that food contributes nearly 15% and water contributes about 85% to the ingestion of uranium in the human body[7]. Uranium has both

*Corresponding author
(E-mail: ranjeet@gjust.org)

radiological and chemical toxicity, and a high amount of uranium in water can cause several health problems, such as stomach cancer, kidney disease, lung cancer, and various other health issues[8]. The main target organs of uranium toxicity are kidneys and lungs[9]. As a result, measuring uranium conc. in groundwater becomes critical for assessing health risks.

The purpose of this study is to investigate the concentration of uranium in the groundwater of Sikar District in Rajasthan around the recently reported uranium deposits in the Royal village of Sikar district. This is a semi-arid area with no nearby river or canal. Hence, people living in this region are dependent on groundwater for drinking and irrigation purposes. The major crops in the studied area are *bajra* (pearl millet), pulses (legumes), barley, and cotton. Rocks of this region are mainly calcerous. In the past, very few studies have been done in the Rajasthan state. Rani *et al.*[10] quantified uranium in Northern Rajasthan and observed that 44% of water samples exhibited higher uranium compared to the WHO limit. Later, Duggal *et al.*[11] measured the uranium in the Sikar district of Rajasthan and reported that nearly 25% of water samples contained uranium higher than the WHO standard. Similarly, Kaur *et al.*[12] also reported higher uranium concentration values compared to the WHO limit in the Sikar district in 27% of water samples. These reports indicate high uranium concentration values in Northern Rajasthan groundwater. The reports of possible uranium deposits in the Sikar district necessitate the estimation of uranium concentration in the groundwater of this area. Although the uranium mining process has not started, the possible mining and milling operation can contribute toward groundwater contamination. Thus, it becomes necessary to measure the uranium concentration in the groundwater of the surrounding area before any mining process starts. This would be helpful in monitoring the change in the radioactivity level of groundwater due to the mining process. The studies reported in the past in Rajasthan state have covered a very wide area, and only one or two samples were taken around the reported uranium deposits. As a result, more research is required to assess the uranium concentration in the region surrounding the uranium deposit site. We conducted a careful analysis of the natural radioactivity present in the groundwater of the region surrounding uranium deposits since a significant uranium presence is expected in this area.

In this study, the age-dependent annual effective dose is estimated along with the radiological and chemical toxicity risks. The retention of uranium and radiological doses to various human organs and tissues is also estimated in this study using the biokinetic and dosimetric model given by ICRP[13]. These models are used because of their effectiveness and their wide acceptance in the scientific community[14]. Other physico-chemical characteristics of water, such as pH, TDS, salinity, etc., are also examined in addition to U conc. These variables inform us of the water's appropriateness for domestic, commercial, industrial, and agricultural uses[15].The presence of hydrogen and hydroxyl ions in water is determined by pH value of water and is vital for body balance and for the regulation of metabolic processes. The TDS value of a water sample refers to the concentration of dissolved substances in water. The TDS value of drinking water should be within a certain level as a high TDS value indicates a higher concentration of dissolved salts and elements, which in turn are harmful to the body. High TDS will give water a bitter taste too. The measured uranium concentration and physico-chemical parameters are compared with the recommended limits proposed by various health organizations.

2 Geology of the Studied Area

The studied region lies in the Sikar district of Rajasthan state, India. Fig. 1 shows the geographic location of the studied area in the Sikar district, along with the sample location. The map is formed using ARC GIS 10.7.1 software. Sikar district lies between 27° 21' to 28° 12'North latitudes and 74° 44' to 75° 25'East longitudes. The selected region lies in the

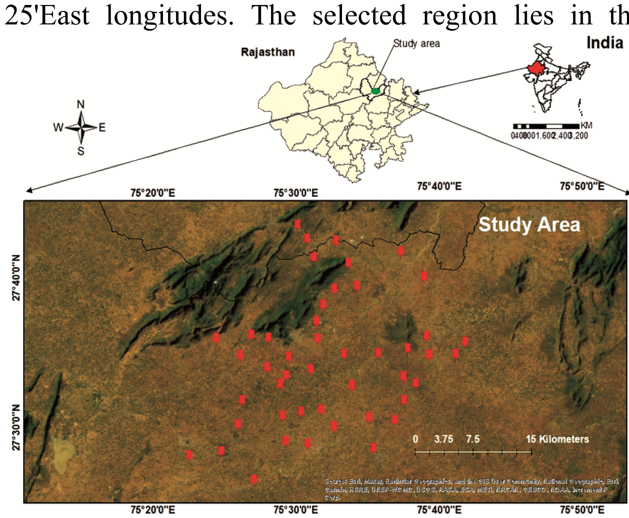

Fig. 1 — Map showing the sampling location. (*The map is only intended to be used as a visual aid and do not indicate any view on the legal positionof any country or territory or the delimitation frontiers or boundaries.)

north-central part of the Sikar district. A few samples were also taken from the Jhunjhunu district. The weather in this area has summer, winter, and monsoon seasons as well. The air is usually dry except for the rainy season. The average temperature in the area is 35 °C, with a maximum of up to 48 °C and a minimum of up to 0 °C.

The major aquifer in the selected area is the hard rock aquifer which constitutes quartzite, schist, phyllite, gneiss, and amphibolite of Delhi Super Group. Compared to alluvium aquifers, the water-yielding capacity of these hard rocks is poor. The general flow of groundwater in the studied region is from the southwest to the northeast direction, and the water level is, in general, less than 50 m.b.g.l. (meters below ground level). Long-term trends of hydrographs indicate a decline in groundwater level. Due to this decline in water level, alluvium is no longer the yielding aquifer[16].

The rock type in the studied area belongs to the Delhi subgroup, which is divided into Alwar and Ajabgarh groups. The rocks in the studied area mainly belong to the Ajabgarh group of Delhi subgroups. The rocks of the Ajabgarh group are characterized by a large portion of calcareous rocks. The main rock types are Mica Schist, Dolomite, Marble, and Phyllites[17]. The soil of the studied region is generally well-drained, light-textured older alluvium soil. There is no major irrigation project in this region, and only groundwater is the main source of irrigation.

The studied area neither has any major industrial development nor any large-scale mining activities. Recently, Atomic Minerals Directorate for Exploration and Research (AMD) found uranium oxide deposits in the Sikar district of Rajasthan. Anew uranium mining project is expected to start in the Royal village of Sikar district by Uranium Corporation of India Limited (UCIL)[18]. The mining operation will be carried out at latitude 27° 33′ 25″ N and longitude 75° 29′ 25″ E. Mining will be carried out within an area of 247.8 ha[19].

3 Materials and Method

3.1 Sampling procedure and water sample preparations

Figure 1 depicts the collection of 46 water samples from various locations in the studied area. Groundwater samples were mostly collected from hand pumps (HP) and bore wells (BW). The samples were taken in250 mlpolypropylene bottles that had been acid washed. Prior to use, these bottles were also cleaned with the help of double distilled water and dried. For sample collection from hand pumps (HP) and bore wells (BW), water was continuously pumped out for about 5-10 minutes until the temperature, EC, and pH were stabilized before fetching inside the bottles. After collecting the water sample, all the samples were filtered with the help of Whatman filter paper No. 42 to get rid of the suspended sediments, and then nitric acid was added to acidify them so that they would last longer.

3.2 LED Fluorimeter technique

The U conc. in water samples is measured using Light Emitting Diode (LED) Fluorimeter (LF-2a). The LED Fluorimeter uses UV radiation of a suitable wavelength to illuminate the liquid solution made up of the intended water sample. The UV light excites the uranium complexes present in the sample. These excited complexes returned to their ground state by emitting a green fluorescence light which is detected by Photo Multiplier Tube (PMT). The minimum uranium detection limit of the instrument in a water sample is 0.2 μg/L. The liquid solution is prepared by mixing the water sample and a buffer solution named Fluren (Fluorescence Enhancing Reagent). This buffer solution is formed by adding 5g of Sodium Pyrophosphate powder to 100 ml of double distilled water and then acidified by adding orthophosphoric acid (H_3PO_4) in it drop by drop until the pH of the solution reaches a value of 7[20]. Thereafter, a cuvette containing a mixture of 6 ml of sample and 10% (0.6 ml) of this buffer solution is placed inside the fluorimeter for measurement. The instrument provides us with an estimate of U conc. by measuring the fluorescence produced by the sample. To estimate the uranium content in groundwater samples, the following formula is used[20].

$$\mathrm{CF} = \frac{C_U}{F_{St} - F_{dw}}$$

Uranium conc. in groundwater sample = CF × (F_S – F_{dw}).

Where C_U= The conc. of U in the standard solution, CF = Calibration Factor, F_{St} = Fluorescence from standard solution, F_{dw} = Fluorescence from distilled water, F_S = Fluorescence from the sample. The instrument carries out all these calculations by itself.

3.2.1 Absorbed dose due to U conc. in groundwater

The following calculation is used to calculate the yearly effective dose D_w (μSv/year) caused by uranium ingestion through drinking water[21].

$$D_w = U_w \times DCF \times 10^3 \times W \quad \ldots(1)$$

Where U_w is activity conc. of U in water (Bq/L), DCF stands for Dose Conversion Factor (mSv/Bq), and WI defines the age-dependent annual Water Intake (L/year).

A conversion factor of 1μg/L = 0.02528 Bq/Lis used to convert the conc. of U in drinking water from μg/L to Bq/L[1]. The average water intake rates for different age groups people are different and the sensitivity of their organs varies too. Thus, age-dependent annual effective dose measurement is imperative.

3.2.2 Radiological Toxicity Risk Assessment

The radiological risk (ECR) due to the intake of uranium via drinking water is calculated using the method provided by USEPA[22].

$$ECR = U_w \times R \quad \ldots(2)$$

Where ECR refers to Excess Cancer Risk, U_w stands for activity conc. of U (Bq/L), and R represents the risk factor (per Bq/L).

The risk factor (R) is calculated using the relation[22] given in eq. 3.

$$R = r \times IR \times ED \quad \ldots(3)$$

Where r = risk coefficient for ingestion (1.19×10^{-9} $Bq^{-1}$for mortality and 1.84×10^{-9} Bq^{-1} for morbidity obtained from literature[23,24]), IR is ingestion rate of drinking water (4.05 L/Day)[25],and ED refers to exposure duration which equals the life expectancy of people *i.e* 63.7 years (or 23250 Days)[25].

3.2.3 Chemical Toxicity Risk Assessment

The Lifetime Average Daily Dose (LADD) of uranium received due to the consumption of water is used to define the chemical toxicity risk caused by uranium. LADD is defined as the daily intake of uranium per kilogram of body weight and can be determined using the equation[26].

$$LADD = \frac{EPC}{AT} \times \frac{IR}{BW} \times EF \times LE \quad \ldots(4)$$

Where LADD stands for Lifetime Average Daily Dose (μg/kg/day),IR refers to the ingestion rate of water (4.05 L/day)[25], and EPC stands for exposure point conc. of U in drinking water (μg/L), LE is the life expectancy (63.7 years)[25], EF defines the exposure frequency (350 days/year)[27],AT is the average time (63.7 × 365 = 23250 days), BW stands for the body weight (53 kg for an Indian adult[28]).

3.2.4 Calculation of Hazard Quotient

The Hazard Quotient quantifies the amount of damage caused by U ingestion through drinking water (HQ).

$$HQ = \frac{LADD}{R_f} \quad \ldots(5)$$

Where HQ is the Hazard Quotient, LADD represents the lifetime average daily dose as defined above and R_f stands for reference dose (4.4 μg/kg/day (AERB)[6] and 1.2 μg/kg/day (WHO)[5]).

3.3 Uranium retention and effective dose to different human organs

To estimate the uranium retention and effective dose to different human organs two significant models given by ICRP[29,30] *i.e.*, the Biokinetic and Dosimetric model are used. In the hair compartment Biokinetic model, a systematic framework is provided for the migration, distribution, and retention of uranium in different body organs as well as how uranium gets excreted from the body via urine, faeces, and hairs[13,31]. The intake of uranium in the nearby population for the age group of 60 was established in this paper utilising the biokinetic model. For the estimation of uranium retention in different organs, parameters like breathing rate (0.633m^3/h), body weight (68.83 Kg), kidney mass (310g), water consumption rate (1.4 L/d), and urine volume (1.38 L/d) are considered as steady. Effective doses to different organs are calculated by using the dose coefficient model given by Li *et al.*[31] by considering the nuclear transformation in the vicinities of the target organs and the deposition of energy in organs and tissues. The complete formulation of the Biokinetic and dosimetric model are summarised by Pragin *et al.*[14].

3.4 Physico – Chemical Parameters:

Physico-Chemical characteristics of water such as pH, TDS, EC, Temperature, and Salinity are also measured with the help of Combo pH/Conductivity/TDS Tester (Low Range) - HI98129 meter. The measurements of these physico-chemical parameters are done on-site at the time of sampling. The measurement of these parameters is also important as they provide us with information about the feasibility of water for different purposes such as drinking, irrigation, domestic use, *etc*[32].These measured values of the physico-chemical parameters are compared with the standard limits recommended by WHO and BIS (Bureau of Indian Standards).

4 Results and Discussion

4.1 Uranium concentration analysis in different water samples

The LED fluorimeter measurements for uranium concentration of 46 different water samples are tabulated in Table 1, along with their physico-chemical parameters measured at the time of sampling. The uranium concentration of samples varies between 4.79 µg/L (min. value) to 266.28 µg/L (max. value) with an average value of 74.36 µg/L. Fig. 2 depicts the number of samples in various U conc. ranges in the form of a pie chart. The reason for such high concentration in the studied area may be

Table 1 — Uranium Concentration of different samples along with physico - chemical parameters

S. No.	Sample Location	Latitude	Longitude	U Conc. (µg/L)	pH	EC (mS/cm)	TDS (ppm)	Salinity (ppt)
1	Berughat	27°43'31"	75°30'5"	98.53	6.6	1.36	680	0.67
2	Bagora	27°42'32"	75°30'47"	249.38	6.6	2.48	1240	1.25
3	Dev ji ki dhani	27°42'25"	75°32'49"	32.09	7.08	1.36	680	0.68
4	Bhaira with luharwas	27°41'18"	75°31'16"	5.83	7.25	0.71	355	0.35
5	Kothri luharwas	27°40'55"	75°33'40"	19.57	7.33	0.94	470	0.47
6	Kerpura	27°39'23"	75°34'17"	219.80	7.85	0.65	325	0.32
7	Lalsar	27°39'12"	75°32'40"	54.95	7.28	1.52	760	0.74
8	Rampura	27°37'1"	75°31'27"	34.86	7.12	1.21	605	0.61
9	Khandela PG college	27°35'52"	75°31'30"	93.19	7.52	1.28	640	0.64
10	Seel ki bara	27°34'49"	75°33'21"	86.48	7.83	0.87	435	0.44
11	Bhojpura	27°32'45"	75°33'54"	111.76	8.1	1.13	565	0.56
12	Ralawat	27°30'35"	75°35'7"	69.34	8.05	0.82	410	0.4
13	Shri Madhopur	27°28'33"	75°35'26"	72.29	7.74	1.13	565	0.57
14	Kalyanpura	27°30'25"	75°36'56"	11.18	7.8	1.65	825	0.82
15	Kanchanpura	27°31'44"	75°37'35"	11.55	7.84	1.18	590	0.59
16	Joravar Nagar	27°32'51"	75°38'24"	41.48	7.82	0.96	480	0.48
17	Hamirpur	27°33'20"	75°37'32"	182.81	7.52	1.65	825	0.83
18	Piplodh ka bas	27°34'53"	75°35'45"	39.15	7.51	1.65	825	0.82
19	Balwar	27°35'14"	75°37'50"	98.06	7.59	1.11	555	0.55
20	Bhadwari	27°34'48"	75°39'20"	29.49	7.54	1.57	785	0.78
21	Garhi Khanpur	27°36'1"	75°39'8"	32.39	7.68	1	500	0.49
22	Kaliyawas	27°34'49"	75°41'9"	13.59	8.04	0.44	220	0.22
23	Kanwat	27°35'39"	75°41'51"	35.29	7.64	0.59	295	0.3
24	Chala	27°40'1"	75°38'59"	150.85	8.02	0.58	290	0.29
25	Guhala	27°41'41"	75°37'20"	27.14	8.3	0.55	275	0.28
26	Meharpur ki dhani	27°34'41"	75°29'31"	120.55	8.22	1.16	580	0.58
27	Gurara	27°35'58"	75°28'4"	6.43	7.35	0.71	355	0.35
28	Fatehpura	27°34'44"	75°26'6"	134.58	7.87	1.17	585	0.59
29	Gurara	27°36'7"	75°26'54"	4.79	7.28	0.87	435	0.44
30	Hatyaz	27°35'55"	75°24'26"	266.28	7.57	1.36	680	0.68
31	Madani	27°24'31"	75°24'54"	22.88	7.97	0.62	310	0.31
32	Thikariya	27°26'25"	75°27'4"	10.54	8.5	0.75	375	0.37
33	Palsana	27°28'3"	75°22'33"	38.00	7.67	1.04	520	0.52
34	Bhadla ki dhani	27°28'18"	75°24'46"	31.58	7.9	1	500	0.5
35	Fatehpur(govindpura)	27°30'8"	75°25'57"	111.97	7.7	1.9	950	0.94
36	Bahadurpura	27°31'45"	75°26'14"	22.60	8.26	0.82	410	0.4
37	Gokul ka bas	27°33'56"	75°27'59"	133.29	7.75	1.48	740	0.74
38	Royal	27°32'48"	75°28'53"	38.15	7.52	1.51	755	0.76
39	Suhagpur	27°33'25"	75°29'19"	164.00	7.8	1.08	540	0.54
40	Goriya	27°30'43"	75°29'4"	28.79	7.75	1.41	705	0.71
41	Pujari ka bas	27°29'0"	75°29'17"	37.76	8.12	1.15	575	0.57
42	Jajod	27°28'51"	75°30'49"	146.17	7.93	1.67	835	0.84
43	Shola	27°30'58"	75°30'21"	53.70	7.95	1.66	830	0.82
44	Nimera	27°31'7"	75°31'46"	25.71	7.56	2.54	1270	1.27
45	Jankipura	27°29'59"	75°32'41"	62.02	7.96	1.06	530	0.53
46	Barsinghpura	27°33'52"	75°31'2"	139.57	7.69	1.12	560	0.56
	Min			4.79	6.6	0.44	220	0.22
	Max			266.28	8.5	2.54	1270	1.27
	Mean			74.36	7.7	1.18	592	0.59

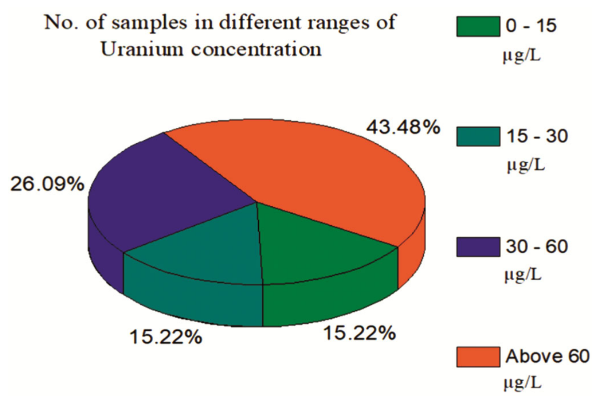

Fig. 2 — Pie Chart showing the number of water samples at various U conc. intervals.

attributed primarily to the presence of the high amount of uranium deposits in this area, which might have significantly elevated the level of uranium in the groundwater of the surrounding area. The presence of Aravalli hills may also influence the geological formation of the studied area[10], and the host rocks of the studied region may contain uranium-bearing minerals[11].

From a safety point of view, a safe limit for the concentration of uranium in drinking water is recommended by many health organisations. The safe limit prescribed by WHO[5], and U. S. Environmental Protection Agency (USEPA)[32] is 30 µg/L. International Commission on Radiological Protection (ICRP)[34] and the United Nations Scientific Committee on Effects of Atomic Radiation (UNSCEAR)[35] prescribe much lower safe limit values of1.9 µg/L and 9 µg/L, respectively. In India, the safe limit stipulated by AERB[6] is 60 µg/L. Interestingly, in the present study, it is found that the concentration of uranium in most of the water samples are greater than the safe limit values of various health organizations. Uranium conc. in about 69% of samples is found to be more than the recommended limit prescribed by WHO and USEPA, whereas 43% of samples exceeded the recommended limit of AERB. The maximum value of uranium concentration found is 266.28µg/L in the village of Hatyaz which is nearly 9 times greater than the safe limit value of WHO. Nearly 93% of samples were found to be having a concentration greater than the limit prescribed by UNSCEAR and all the groundwater samples show concentrations far above the recommended limit of ICRP. The measured uranium concentration in one or two of the locations also matches quite well with the previously done study[11] on those locations in the studied area.

Table 2 — Age-dependent annual effective dose

S. No.	Life stage	Age group (years)	DWI[12,39] (L/day)	DCF[12] (mSv/Bq)
1.	Infant	0-0.5	0.7	3.4×10^{-4}
		0.5-1	0.8	3.4×10^{-4}
2.	Children	1-3	1.3	1.2×10^{-4}
		4-8	1.7	8×10^{-5}
3.	Male	9-13	2.4	6.8×10^{-5}
		14-18	3.3	6.7×10^{-5}
		>18	3.7	1.3×10^{-5}
4.	Female	9-13	2.1	6.8×10^{-5}
		14-18	2.3	6.7×10^{-5}
		>18	2.7	1.3×10^{-5}
5.	Pregnancy	19-50	3.0	1.3×10^{-5}
6.	Lactation	19-50	3.8	1.3×10^{-5}

The value of pH for all the water samples is found to be varying in between 6.6 to 8.5 with a mean value of 7.69, indicating that pH values of all groundwater samples lie well within the recommended limit of BIS (2012)[36]. EC varies from 0.44 mS/cm to 2.54 mS/cm with an average value of 1.18 mS/cm. It is found that EC values in 23.9 % of samples exceed the recommended level of 1.5 mS/cm prescribed by WHO (1993)[37] and 63.8 % of samples exhibit that EC value lies above the water act (1956)[38] permissible limit of 1 mS/cm.

These EC values fall in the good to permissible categories except for two samples in which EC values lie in the doubtful region. TDS value varies from a minimum of 220 ppm to a maximum of 1270 ppm, with an average value of 592.06 ppm. No sample exceeds the safe limit of 2000 ppm recommended by BIS 2012[36].

4.2 Age-dependent Annual Effective Dose Analysis

As daily water intake by people of various age groups is different, so it becomes necessary to measure the yearly effective dose for people of different age groups. The daily water intake for people of various age groups and their corresponding dose conversion factors are tabulated in Table 2. It is found that infants of age 7-12 months have the highest mean value of effective dose (186.62 µSv/ year), as shown in Fig. 3 below. The effective dose is found to be higher than the recommended limit for infants in nearly 50% of the samples, which can be attributed to the fact that they consume more water compared to their body mass and due to immature organ development[40,41]. In the case of children, for the age groups of 1-3 years and 4-8 years, nearly 39.1% and 36.9% of samples, respectively, exceed the recommended level of 100 µSv/ year of WHO[42]. For

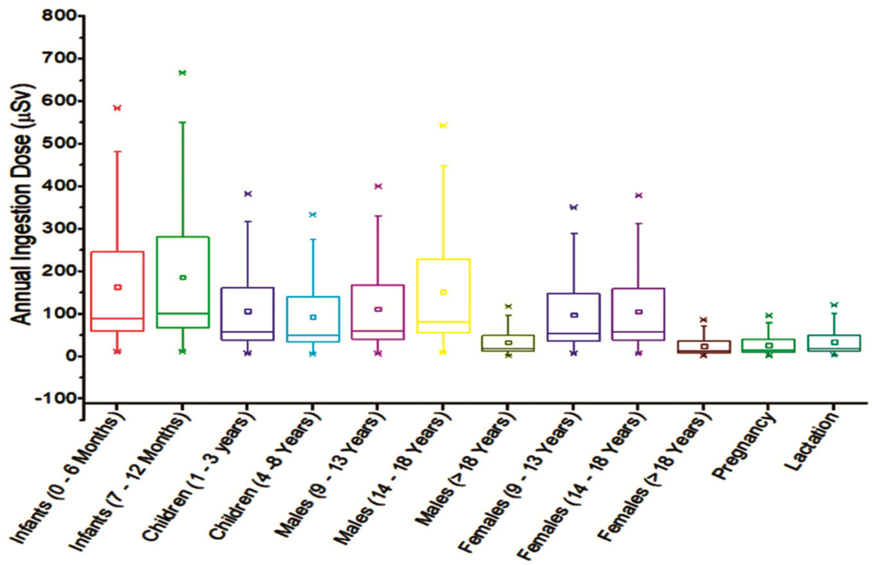

Fig. 3 — Box and Whisker plot for Age-Dependent annual effective dose.

males of age group 9 -13 years and 14-18 years, 41.3% and 47.8% of samples exceed the recommended level of 100 µSv/ year, respectively, while for males of the age group above 18 years, only two sample lies above the recommended limit.

Similarly, for females of age group 9-13 years and 14-18 years, 36.9% and 39.1% of samples lie above the recommended safe limit of 100 µSv/ year, respectively. However, for females of age > 18 years, no sample exceeds the safe limit of WHO. As women consume more water during the lactation phase, 3 samples have ingestion doses higher than the prescribed limit while, during the pregnancy phase, no sample exhibited ingestion doses greater than the recommended limit of WHO.

When compared to individuals over the age of 18, the effective dose value in newborns and teenagers is about ten times higher. The observed high effective dose for the age range 14-18 years could be attributed to the body producing a large amount of sexual hormones when puberty occurs at this age. According to the findings of this study, those under the age of 18 are more affected by groundwater in the study area, and hence consuming this water is not recommended for infants and teens. However, the risk for those over the age of 18 appears to be insignificant and hence can be regarded as safe for consumption by adults.

4.3 Radiological and Chemical toxicity risk analysis in water samples

The number of cancer-related fatalities per 100,000 persons is referred to as the cancer mortality rate. The excess cancer risk (ECR) value for mortality for adult humans lies in the range from 1.36×10^{-5} to 7.54×10^{-4} with an average value of 2.11×10^{-4}. The term "cancer morbidity" describes the condition of becoming ill as a result of cancer. The ECR value for morbidity lies in the range of 2.10×10^{-5} to 1.17×10^{-3} with an average value of 3.26×10^{-4}. The cancer risk for mortality lies well below the recommended limit[1,43,44] of 10^{-3}. However, two samples exhibit a cancer risk value for morbidity greater than the prescribed limit[1,43,44] of 10^{-3}. Uranium also possesses chemical toxicity (non-carcinogenic) risks along with radiological toxicity (carcinogenic) risks. Uranium is more deleterious because of its chemical toxicity which is determined in terms of LADD value. In this study, it is found that the LADD value varies between 0.35 µg/kg/dayto19.51 µg/kg/day with an average value of 5.44 µg/kg/day which is higher than the reference dose of both WHO (1.2 µg/kg/day)[5] and AERB (4.4 µg/kg/day)[6]. ECR, LADD, and HQ values are shown in Table 3.

The HQ value is calculated by dividing the LADD value by the reference dose value of 1.2 µg/kg/day and 4.4 µg/kg/day in accordance with the WHO and AERB standards, respectively. According to the AERB standard, 43.4 % of samples exceed the prescribed limit of 1 while according to WHO standards, 84.7 % of samples have HQ values higher than 1. The fact that HQ values are greater than 1 in so many of the samples shows that there is a danger of chemical toxicity in the drinking water of the research area.

Table 3 — Radiological risk, chemical toxicity risk, and hazard index for all samples

S. No.	Sample Location	ECR (Mortality)	ECR (Morbidity)	LADD (µg/kg/day)	HQ (WHO)	HQ (AERB)
1	Berughat	2.79E-04	4.32E-04	7.22	6.02	1.64
2	Bagora	7.06E-04	1.09E-03	18.27	15.23	4.15
3	Dev ji ki dhani	9.09E-05	1.41E-04	2.351	1.96	0.53
4	Bhaira with luharwas	1.65E-05	2.55E-05	0.427	0.36	0.10
5	Kothri luharwas	5.54E-05	8.57E-05	1.434	1.19	0.33
6	Kerpura	6.23E-04	9.63E-04	16.11	13.42	3.66
7	Lalsar	1.56E-04	2.41E-04	4.026	3.36	0.92
8	Rampura	9.88E-05	1.53E-04	2.555	2.13	0.58
9	Khandela PG college	2.64E-04	4.08E-04	6.829	5.69	1.55
10	Seel ki bara	2.45E-04	3.79E-04	6.337	5.28	1.44
11	Bhojpura	3.17E-04	4.89E-04	8.189	6.82	1.86
12	Ralawat	1.96E-04	3.04E-04	5.081	4.23	1.15
13	Shri Madhopur	2.05E-04	3.17E-04	5.297	4.41	1.20
14	Kalyanpura	3.17E-05	4.90E-05	0.819	0.68	0.19
15	Kanchanpura	3.27E-05	5.06E-05	0.846	0.71	0.19
16	Joravar Nagar	1.18E-04	1.82E-04	3.04	2.53	0.69
17	Hamirpur	5.18E-04	8.01E-04	13.4	11.16	3.04
18	Piplodh ka bas	1.11E-04	1.71E-04	2.869	2.39	0.65
19	Balwar	2.78E-04	4.29E-04	7.185	5.99	1.63
20	Bhadwari	8.35E-05	1.29E-04	2.161	1.80	0.49
21	Garhi Khanpur	9.18E-05	1.42E-04	2.373	1.98	0.54
22	Kaliyawas	3.85E-05	5.95E-05	0.996	0.83	0.23
23	Kanwat	1.00E-04	1.55E-04	2.586	2.15	0.59
24	Chala	4.27E-04	6.61E-04	11.05	9.21	2.51
25	Guhala	7.69E-05	1.19E-04	1.989	1.66	0.45
26	Meharpur ki dhani	3.41E-04	5.28E-04	8.833	7.36	2.01
27	Gurara	1.82E-05	2.81E-05	0.471	0.39	0.11
28	Fatehpura	3.81E-04	5.89E-04	9.862	8.22	2.24
29	Gurara	1.36E-05	2.10E-05	0.351	0.29	0.08
30	Hatyaz	7.54E-04	1.17E-03	19.51	16.26	4.43
31	Madani	6.48E-05	1.00E-04	1.677	1.40	0.38
32	Thikariya	2.99E-05	4.62E-05	0.773	0.64	0.18
33	Palsana	1.08E-04	1.66E-04	2.784	2.32	0.63
34	Bhadla ki dhani	8.95E-05	1.38E-04	2.314	1.93	0.53
35	Fatehpur(govindpura)	3.17E-04	4.90E-04	8.205	6.84	1.86
36	Bahadurpura	6.40E-05	9.90E-05	1.656	1.38	0.38
37	Gokul ka bas	3.78E-04	5.84E-04	9.767	8.14	2.22
38	Royal	1.08E-04	1.67E-04	2.795	2.33	0.64
39	Suhagpur	4.65E-04	7.18E-04	12.02	10.01	2.73
40	Goriya	8.16E-05	1.26E-04	2.11	1.76	0.48
41	Pujari ka bas	1.07E-04	1.65E-04	2.767	2.31	0.63
42	Jajod	4.14E-04	6.40E-04	10.71	8.93	2.43
43	Shola	1.52E-04	2.35E-04	3.935	3.28	0.89
44	Nimera	7.28E-05	1.13E-04	1.884	1.57	0.43
45	Jankipura	1.76E-04	2.72E-04	4.544	3.79	1.03
46	Barsinghpura	3.95E-04	6.11E-04	10.23	8.52	2.32

4.4 Uranium retention and annual effective doses to various organs and tissues

The retention and excretion of uranium to different organs estimated by Biokinetic modelling are tabulated in Table 4.

The mean concentration of uranium in Blood is 0.049 µg with a minimum value of 0.003 µg and a maximum value of 0.177 µg. The mean value of uranium concentration in Cortical bone surface, cortical bone volume, trabecular bone surface, and trabecular

Table 4 — Retention of uranium in different body organs.

S. No.	U (μg/L)	Blood (μg)	Skeleton (μg)				Kidney		Liver (μg)	OST (μg)	GI tract (μg)			
			C_bone_s	C_bone_v	t_bone_s	t_bone_v	Retention (μg)	Concentration (μg /g)			st_cont	si_cont	uli_cont	lli_cont
1	98.53	0.066	0.72	128.32	0.90	34.53	1.70	0.0054	5.24	51.81	5.75	22.85	76.18	137.12
2	249.38	0.166	1.83	324.77	2.29	87.39	4.30	0.0138	13.27	131.13	14.55	57.84	192.81	347.05
3	32.09	0.021	0.24	41.79	0.29	11.25	0.55	0.0017	1.71	16.87	1.87	7.44	24.81	44.66
4	5.83	0.004	0.04	7.59	0.05	2.04	0.10	0.0003	0.31	3.07	0.34	1.35	4.51	8.11
5	19.57	0.013	0.14	25.49	0.18	6.86	0.34	0.0011	1.04	10.29	1.14	4.54	15.13	27.23
6	219.80	0.146	1.61	286.24	2.02	77.03	3.79	0.0122	11.70	115.58	12.82	50.98	169.94	305.88
7	54.95	0.037	0.40	71.56	0.50	19.26	0.95	0.0030	2.92	28.89	3.21	12.74	42.48	76.47
8	34.86	0.023	0.26	45.40	0.32	12.22	0.60	0.0019	1.86	18.33	2.03	8.09	26.95	48.51
9	93.19	0.062	0.68	121.36	0.86	32.66	1.61	0.0051	4.96	49.00	5.44	21.61	72.05	129.69
10	86.48	0.058	0.63	112.62	0.79	30.31	1.49	0.0048	4.60	45.47	5.04	20.06	66.86	120.35
11	111.76	0.074	0.82	145.54	1.03	39.17	1.93	0.0062	5.95	58.77	6.52	25.92	86.41	155.53
12	69.34	0.046	0.51	90.30	0.64	24.30	1.19	0.0038	3.69	36.46	4.04	16.08	53.61	96.50
13	72.29	0.048	0.53	94.14	0.66	25.33	1.25	0.0040	3.85	38.01	4.22	16.77	55.89	100.60
14	11.18	0.007	0.08	14.56	0.10	3.92	0.19	0.0006	0.60	5.88	0.65	2.59	8.64	15.56
15	11.55	0.008	0.08	15.04	0.11	4.05	0.20	0.0006	0.61	6.07	0.67	2.68	8.93	16.07
16	41.48	0.028	0.30	54.02	0.38	14.54	0.71	0.0023	2.21	21.81	2.42	9.62	32.07	57.73
17	182.81	0.122	1.34	238.07	1.68	64.06	3.15	0.0101	9.73	96.13	10.66	42.40	141.34	254.41
18	39.15	0.026	0.29	50.98	0.36	13.72	0.67	0.0021	2.08	20.59	2.28	9.08	30.27	54.48
19	98.06	0.065	0.72	127.70	0.90	34.36	1.69	0.0054	5.22	51.56	5.72	22.74	75.81	136.47
20	29.49	0.020	0.22	38.40	0.27	10.33	0.51	0.0016	1.57	15.51	1.72	6.84	22.80	41.04
21	32.39	0.022	0.24	42.18	0.30	11.35	0.56	0.0018	1.72	17.03	1.89	7.51	25.04	45.08
22	13.59	0.009	0.10	17.70	0.12	4.76	0.23	0.0007	0.72	7.15	0.79	3.15	10.51	18.91
23	35.29	0.023	0.26	45.96	0.32	12.37	0.61	0.0019	1.88	18.56	2.06	8.18	27.28	49.11
24	150.85	0.100	1.11	196.45	1.38	52.86	2.60	0.0083	8.03	79.32	8.80	34.99	116.63	209.93
25	27.14	0.018	0.20	35.34	0.25	9.51	0.47	0.0015	1.44	14.27	1.58	6.29	20.98	37.77
26	120.55	0.080	0.88	156.99	1.11	42.25	2.08	0.0067	6.42	63.39	7.03	27.96	93.20	167.76
27	6.43	0.004	0.05	8.37	0.06	2.25	0.11	0.0003	0.34	3.38	0.38	1.49	4.97	8.95
28	134.58	0.090	0.99	175.26	1.24	47.16	2.32	0.0074	7.16	70.77	7.85	31.21	104.05	187.29
29	4.79	0.003	0.04	6.24	0.04	1.68	0.08	0.0002	0.25	2.52	0.28	1.11	3.70	6.67
30	266.28	0.177	1.95	346.78	2.44	93.32	4.59	0.0147	14.17	140.02	15.53	61.76	205.87	370.57
31	22.88	0.015	0.17	29.80	0.21	8.02	0.39	0.0012	1.22	12.03	1.33	5.31	17.69	31.84
32	10.54	0.007	0.08	13.73	0.10	3.69	0.18	0.0005	0.56	5.54	0.61	2.44	8.15	14.67
33	38.00	0.025	0.28	49.49	0.35	13.32	0.65	0.0021	2.02	19.98	2.22	8.81	29.38	52.88
34	31.58	0.021	0.23	41.13	0.29	11.07	0.54	0.0017	1.68	16.61	1.84	7.32	24.42	43.95
35	111.97	0.074	0.82	145.82	1.03	39.24	1.93	0.0062	5.96	58.88	6.53	25.97	86.57	155.82
36	22.60	0.015	0.17	29.43	0.21	7.92	0.39	0.0012	1.20	11.88	1.32	5.24	17.47	31.45
37	133.29	0.089	0.98	173.58	1.22	46.71	2.30	0.0074	7.09	70.09	7.78	30.91	103.05	185.49
38	38.15	0.025	0.28	49.68	0.35	13.37	0.66	0.0021	2.03	20.06	2.23	8.85	29.50	53.09
39	164.00	0.109	1.20	213.58	1.51	57.47	2.83	0.0091	8.73	86.24	9.57	38.04	126.79	228.23
40	28.79	0.019	0.21	37.49	0.26	10.09	0.50	0.0016	1.53	15.14	1.68	6.68	22.26	40.07
41	37.76	0.025	0.28	49.17	0.35	13.23	0.65	0.0020	2.01	19.86	2.20	8.76	29.19	52.55
42	146.17	0.097	1.07	190.36	1.34	51.22	2.52	0.0081	7.78	76.86	8.53	33.90	113.01	203.42
43	53.70	0.036	0.39	69.93	0.49	18.82	0.93	0.0029	2.86	28.24	3.13	12.45	41.52	74.73
44	25.71	0.017	0.19	33.48	0.24	9.01	0.44	0.0014	1.37	13.52	1.50	5.96	19.88	35.78
45	62.02	0.041	0.45	80.77	0.57	21.73	1.07	0.0034	3.30	32.61	3.62	14.38	47.95	86.31
46	139.57	0.093	1.02	181.76	1.28	48.91	2.40	0.0077	7.43	73.39	8.14	32.37	107.91	194.23

bone volume are found to be 0.55 μg, 96.83 μg, 0.68 μg, 26.06 μg, respectively. As cortical bone has a large volume fraction, the uranium concentration is found more in cortical bone. The value of uranium in the kidney is found to be lying in the rile 0.08 – 4.59 μg with an average value of 1.28 μg. Similarly, uranium concentration in the liver is found to be 0.025 – 14.17 μg with a mean value of 3.96 μg.

The gastrointestinal tract (GIT) is the first human organ through which uranium enters the human body. So, it is essential to measure the uranium concentration in GIT. GIT has four sections, stomach wall, small intestine wall, upper large intestine, and lower large intestine, which are attributed as St wall, SI wall, ULI, and LLI, respectively. The mean value of U conc. in all these four sections are found to be 4.34 μg, 17.25 μg, 57.49 μg, and 103.48 μg, respectively. The U conc. in other soft tissues is also important to measure as these are the neighbouring sites of the target organs. The study shows that the mean concentration of U in these soft tissues is 39.10 μg.

The annual effective radiological dose to different human organs, *i.e.*, Kidney, Liver, Gonads (Testes and Ovaries), Red marrow, Bone surface, *etc*., are shown in Table 5. The mean effective dose is found to be 12.31 μSv (0.79 – 44.08 μSv). The uranium dose to kidneys is found to be lying in the range of 3.72 – 206.83 μSv with an average value of 57.76 μSv. The mean dose values of 21.78 μSv (1.40 – 77.99 μSv) in the liver, 5.59 μSv (0.36 – 20.01 μSv) in the ovaries, 5.49 μSv (0.35 – 19.67 μSv) in testes, and 17.04 μSv (1.10 – 61.03 μSv) in red marrow have been noticed.

4.5 Pearson's correlation coefficient analysis

In the present work, the correlation of uranium concentration with various physico-chemical parameters of water is also estimated. It is observed that uranium exhibits a slight positive correlation with EC (r = 0.277). The correlation between uranium and TDS is 0.277, and the U-salinity correlation is 0.282, while a slight negative correlation with pH value (r = -0.128) is observed. This confirms the fact that the groundwater samples with high TDS value show a high uranium concentration as the water which has high TDS will have more ionic species and consequently will have a higher tendency to interact with the uranium in groundwater.

Table 5 — Annual effective dose to different body organs.

S.No.	Locations	Kidney (μSv)	Liver (μSv)	Gonads		Red Marrow (μSv)	Ef μSv
				Testes (μSv)	Ovaries (μSv)		
1	Berughat	17.09	6.44	1.62	1.65	5.04	3.64
2	Bagora	24.52	9.25	2.33	2.37	7.24	5.23
3	Dev ji ki dhani	76.54	28.86	7.28	7.40	22.58	16.31
4	Bhaira with luharwas	193.70	73.04	18.42	18.74	57.16	41.28
5	Kothri luharwas	24.93	9.40	2.37	2.41	7.36	5.31
6	Kerpura	4.53	1.71	0.43	0.44	1.34	0.96
7	Lalsar	15.20	5.73	1.45	1.47	4.48	3.24
8	Rampura	170.73	64.37	16.23	16.51	50.38	36.38
9	Khandela PG college	42.68	16.09	4.06	4.13	12.59	9.10
10	Seel ki bara	27.08	10.21	2.57	2.62	7.99	5.77
11	Bhojpura	72.39	27.29	6.88	7.00	21.36	15.43
12	Ralawat	67.17	25.33	6.39	6.50	19.82	14.32
13	Shri Madhopur	86.81	32.73	8.25	8.40	25.62	18.50
14	Kalyanpura	53.86	20.31	5.12	5.21	15.89	11.48
15	Kanchanpura	56.15	21.17	5.34	5.43	16.57	11.97
16	Joravar Nagar	8.68	3.27	0.83	0.84	2.56	1.85
17	Hamirpur	8.97	3.38	0.85	0.87	2.65	1.91
18	Piplodh ka bas	32.22	12.15	3.06	3.12	9.51	6.87
19	Balwar	142.00	53.54	13.50	13.73	41.90	30.26
20	Bhadwari	30.41	11.47	2.89	2.94	8.97	6.48
21	Garhi Khanpur	76.17	28.72	7.24	7.37	22.47	16.23
22	Kaliyawas	22.90	8.64	2.18	2.22	6.76	4.88
23	Kanwat	25.16	9.49	2.39	2.43	7.42	5.36
24	Chala	10.56	3.98	1.00	1.02	3.11	2.25
25	Guhala	27.41	10.33	2.61	2.65	8.09	5.84
26	Meharpur ki dhani	117.17	44.18	11.14	11.33	34.58	24.97
27	Gurara	21.08	7.95	2.00	2.04	6.22	4.49

(Contd.)

Table 5 — Annual effective dose to different body organs.

S.No.	Locations	Kidney (μSv)	Liver (μSv)	Gonads		Red Marrow (μSv)	Ef μSv
				Testes (μSv)	Ovaries (μSv)		
28	Fatehpura	93.63	35.30	8.90	9.06	27.63	19.95
29	Gurara	4.99	1.88	0.47	0.48	1.47	1.06
30	Hatyaz	104.54	39.42	9.94	10.11	30.85	22.28
31	Madani	3.72	1.40	0.35	0.36	1.10	0.79
32	Thikariya	206.83	77.99	19.67	20.01	61.03	44.08
33	Palsana	17.77	6.70	1.69	1.72	5.24	3.79
34	Bhadla ki dhani	8.19	3.09	0.78	0.79	2.42	1.75
35	Fatehpur(govindpura)	29.51	11.13	2.81	2.85	8.71	6.29
36	Bahadurpura	24.53	9.25	2.33	2.37	7.24	5.23
37	Gokul ka bas	86.97	32.79	8.27	8.41	25.66	18.53
38	Royal	17.56	6.62	1.67	1.70	5.18	3.74
39	Suhagpur	103.53	39.04	9.84	10.01	30.55	22.06
40	Goriya	29.63	11.17	2.82	2.87	8.74	6.31
41	Pujari ka bas	127.39	48.03	12.11	12.32	37.59	27.15
42	Jajod	22.37	8.43	2.13	2.16	6.60	4.77
43	Shola	29.33	11.06	2.79	2.84	8.65	6.25
44	Nimera	113.54	42.81	10.80	10.98	33.50	24.20
45	Jankipura	41.71	15.73	3.97	4.03	12.31	8.89
46	Barsinghpura	19.97	7.53	1.90	1.93	5.89	4.26

5 Conclusions

In the present study, the U conc. in forty-six groundwater samples taken from the region around the alleged uranium reserves is estimated using the LED fluorimeter technique. In 69% and 43% of samples, Uranium concentrations exceed the WHO and AERB recommended limits, respectively. Uranium concentration varies from 4.79 µg/L to 266.28 µg/L with a mean value of 74.36 µg/L. The average value of U conc. is also found to be m recommended value of all health organisations. This elevated level of U conc. in the studied area may be due to the presence of Aravalli hills and the high amount of uranium deposits in this area.

An attempt has also been made to measure uranium retention and effective dose in different body organs. The liver, bone surface, and kidney display the maximum retention of uranium, indicating that uranium is being accumulated in these organs. Annual effective dose is also higher in kidneys than other organs, indicating that kidneys are the primary target organs of uranium toxicity. The estimated average value of excess cancer risk for mortality is 2.11×10^{-4} while for morbidity, the average value is 3.26×10^{-4}. The value of cancer risk for mortality is found to be lying well within the safe limit for all samples but for morbidity two of the samples exhibit higher cancer risk. The average value of LADD is 5.44µg/kg/day which is nearly 5 times the normal value which indicates the high value of chemical toxicity of uranium in the groundwater. The average value of HQ index (4.54 (WHO) and 1.24 (AERB)) also turned out to be above the recommended safe limit of 1. The yearly effective dose value come across to be highest for infants as they consume more water compared to their body mass. For teens, the effective dose is also found to be higher compared to adults which may be due to the production of sexual hormones during puberty. Also, the dose value is found to be higher for males compared to females in the respective age groups. From the observed measurements, it can be concluded that the groundwater in many portions of the region under study is unfit for human consumption. Government should also take precautionary steps during future mining project so that the uranium concentration of underground water does not increase any further. A further study of more of the surrounding area can be done to check the suitability of groundwater for various purposes.

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