Risk assessment of uranium in drinking water in Hisar district of Haryana, India

Vikas Duggal, Samriti Sharma and Amandeep Singh

ABSTRACT

The present study highlights uranium concentrations, associated health risks and physico-chemical properties of groundwater samples collected from the Hisar district of Haryana State, India. We found that uranium concentrations in 21 out of 68 (30.9%) samples exceeded the WHO provisional guideline value of 30 μg L^{-1}. The annual effective doses were estimated for different life stage groups. The highest dose was calculated for infants. From a radiological perspective, the mean cancer mortality risk and cancer morbidity risk were found to be 4.7×10^{-5} and 7.3×10^{-5}, respectively, which are lower than the permissible limit of 1.67×10^{-4} as prescribed by the Atomic Energy Regulatory Board, India. The lifetime average daily dose (LADD) of uranium ranged from 0.03 to 7.83 μg kg^{-1} day^{-1}. Approximately 23.5% of the samples showed significant chemical toxicity risk. A positive correlation between uranium and total dissolved solids (TDS) was observed.

Key words | annual effective dose, chemical toxicity, groundwater, radiological risk, uranium

Vikas Duggal (corresponding author)
Department of Physics,
DAV College,
#28464, St. No. 3A, S.A.S. Nagar, Bathinda 151001,
Punjab,
India
E-mail: *vikasduggal86@gmail.com*

Samriti Sharma
Amandeep Singh
Department of Applied Sciences,
Chandigarh University,
Gharuan-Mohali 140413, Punjab,
India

HIGHLIGHTS

- We report uranium concentrations in 68 groundwater samples collected from the Hisar district of Haryana, India.
- Uranium concentrations in 31% of the samples exceeded the WHO provisional guideline value of 30 μg L^{-1}.
- The infants have received relatively high mean annual effective doses compared to the other age groups.
- Approximately 23.5% of the samples showed $HQ > 1$, indicating chemical toxicity risk.

INTRODUCTION

Uranium is a naturally occurring radioactive element that is commonly present in groundwater. Studies show that the contribution of ingested uranium through food products accounts for 15%, whereas drinking water contributes to 85% of the ingested uranium. Hence, the health risk due to consumption of uranium-containing groundwater poses a greater risk compared to other causes (Cothern & Lappenbusch 1983; Rani *et al.* 2013; Adithya *et al.* 2019). Uranium concentration in groundwater depends on several factors, including lithological, geomorphologic and other geological conditions of the area. Uranium concentration can also result from human activities such as mining, combustion of coal and other fuels, the use of phosphate fertilizers, and nuclear power production (Kumar *et al.* 2016; Duggal *et al.* 2017).

Although uranium present in the environment, although it has no known positive metabolic functions and is regarded as a non-essential component, when accumulated in humans, it results in chemical and radioactive effects in the form of various health hazards (Sharma *et al.* 2019;

doi: 10.2166/ws.2020.313

Duggal *et al.* 2020). Most of the inhaled and ingested uranium is not absorbed and leaves the body in the feces. Absorbed uranium leaves the body in the urine. Some inhaled uranium can stay in the lungs for a long time. The uranium that is absorbed is deposited throughout the body; the highest levels are found in the bones, liver, and kidneys. Sixty-six percent of the uranium in the body is found in bones. It can remain in the bones for a long time; the half-life of uranium in bones is 70–200 days. Most of the uranium that is not in the bones leaves the body in 1–2 weeks (ATSDR 2013).

Physicochemical parameters (pH, total dissolved solids (TDS) and electrical conductivity (EC)) of groundwater are important in the sense that these parameters can provide important first hand in-situ information about the suitability of water for drinking purposes. TDS comprise inorganic salts (principally calcium, magnesium, sodium and potassium cations and carbonate, bicarbonate, chloride, sulphate and nitrate anions) and small amounts of organic matter that are dissolved in water. TDS in groundwater originate from natural sources, sewage, urban and agricultural run-off and industrial wastewater. pH is an another important monitoring parameter to assess the health of aquatic ecosystem, irrigation sources and discharges, livestock, drinking water sources, industrial discharges and intakes (Kumar *et al.* 2011; Bajwa *et al.* 2017).

There is a wide range of opinions on uranium standards, guidelines and health goals both nationally and internationally (Table 1). A review of the literature reveals that data on the concentration of uranium in groundwater is broadly available for many states of India, but no such study has been carried out earlier in the Hisar district of Haryana. This study aims to understand the uranium distribution in groundwater and compare the observed concentrations

Table 1 | Standards and guidelines for uranium in drinking water

Country/Bodies	Value ($\mu g\ L^{-1}$)		Reference
Atomic Energy Regulatory Board, India	RBL	60	AERB (2004)
Australia	GV	17	NHMRC (2011)
Bulgaria	ML	60	European Food Safety Authority (2009)
Canada	MAC	20	Health Canada (2019)
European Union	ML	Not specified	European Union Council Directive (2013)
Finland	RV	100	European Food Safety Authority (2009)
Germany	Limit	10	Bundesministerium für Gesundheit (2011)
Indonesia	ML	15	Peraturan Menteri Kesehatan Republik Indonesia (2010)
Japan	TV	2	Japan Ministerial Ordinance Concerning Drinking-water Quality Standards (2010)
Malaysia	MAV	2	Ministry of Health Malaysia (2004)
New Zealand	MAV	20	Ministry of Health New Zealand (2008)
Oman	ML	15	Omani Standard (2012)
Peru	MPL	15	Ministerio de Salud Lima-Perú (2011)
Singapore	MPQ	15	Environmental Public Health Regulations Singapore (2008)
Slovenia	ML	6.8	European Food Safety Authority (2009)
South Africa	GV	15	South African National Standard (2011)
The Russian Federation	MAC	100	The Russian Federation (2003)
Uganda	ML	15	Uganda Standard (2008)
United States Environmental Protection Agency	MCL	30	USEPA (2011)
World Health Organization	PGV	30	WHO (2011)

RBL, Radiological based limit; GV, Guideline value; ML, Maximum limit; MAC, Most acceptable concentration; RV, Recommended value; TV, Target value; MAV, Maximum acceptable value; MPL, Maximum permissible limit; MPQ, Maximum prescribed quantity; MCL, Maximum contaminant level; PGV, Provisional guideline value.

with drinking water quality guidelines/standards, to compute age-dependent annual effective doses (AEDs) and to determine radiological and chemical toxicity risks to humans due to ingestion of uranium in drinking water. The physicochemical parameters were also quantified to measure the groundwater quality and to find the correlation, if any, with the determined uranium concentrations.

GEOLOGY OF STUDY AREA

Haryana State is situated in North India. Hisar is the west central most district of Haryana State with a total geographical area of 3,860 km^2 and lies between the north latitudes 28°56′00″ to 29°38′30″ and east longitudes 75°21′12″ to 76°18′12″. Figure 1 shows the geographic location of the Hisar district on the map of Haryana as well as the location of the sampling sites. The area forms a part of the Indo-Gangetic plain. The geological formations met within the district comprise unconsolidated alluvial deposits of quaternary age. The area falls in the Yamuna sub-basin of the Ganga Basin. The area is irrigated by shallow tube wells, the network of Bhakra canal systems and western Yamuna canal system. Groundwater occurs in the alluvium under a water table and is semi-confined to confined conditions. The district is divided into two geographic regions, that is, upload plain and sand dune clusters. The soils of the area are of three types: arid brown solonized, sierozem and desert soils. The study region is bounded by the Fatehabad district in the north, Hanumangarh district of

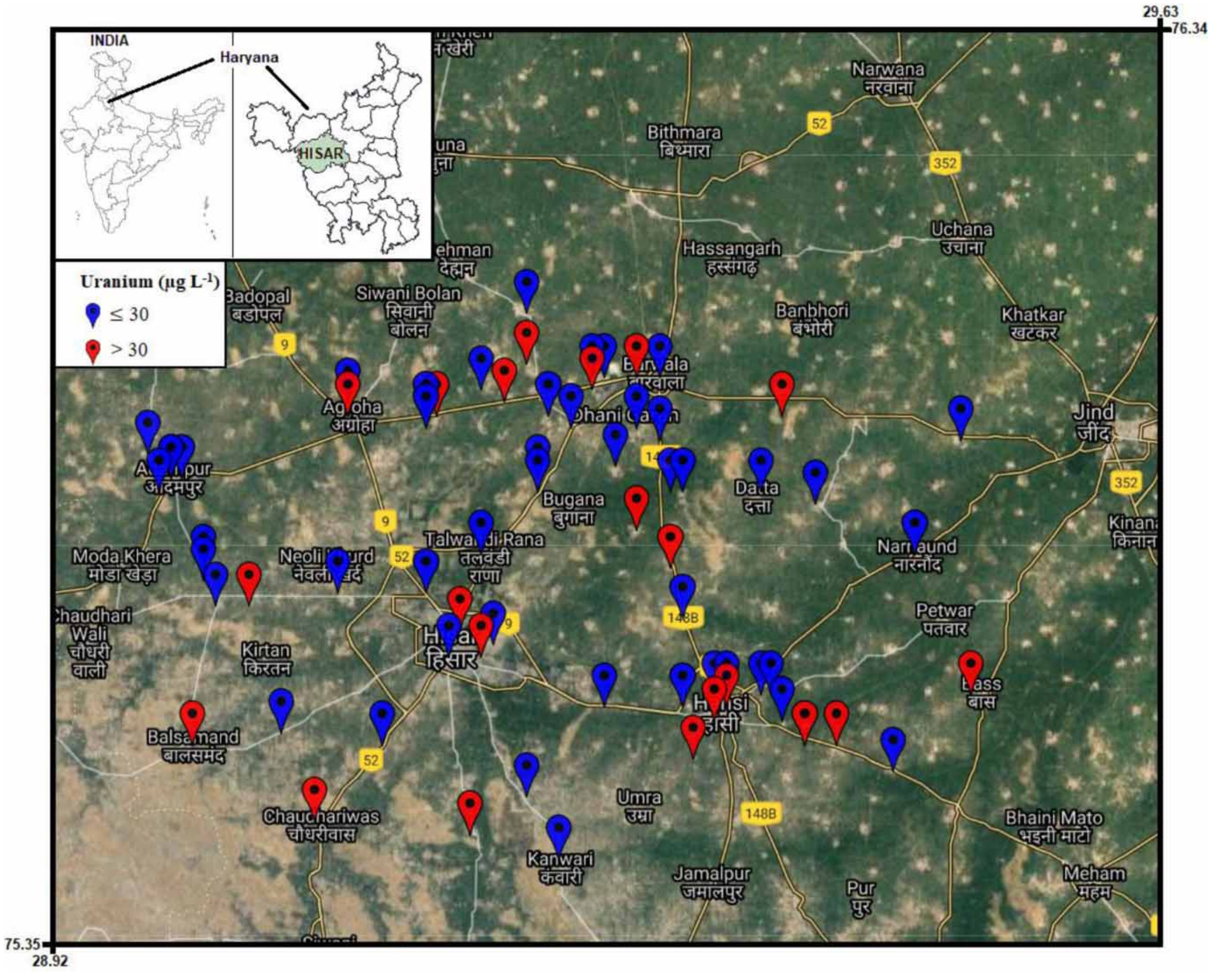

Figure 1 | Map showing the sample locations in the Hisar district of Haryana.

Rajasthan in the west, Jind district in the east, and Bhiwani and Rohtak districts of Haryana in the south and southeast, respectively (CGWB Hisar 2017; Sharma *et al.* 2017).

MATERIALS AND METHODS

Sample collection and physicochemical parameters

A total of 68 water samples were collected from the Hisar district of Haryana (Figure 1). The sources of water comprise bore wells, electric motors, tube wells and hand pumps. Sampling sites were chosen where water is continuously used for human consumption as well as animals and in crop production. The sampling sites were also chosen in such a manner that the study area is uniformly represented. The position of each sampling site was determined by using portable global positioning system (GPS). The water freshness was maintained by pumping out enough water for about 10 min before sampling. Before collection, the water samples were filtered using 0.45 μm Whatman filter paper to remove suspended matter/sediments and then stored in pre-cleaned acid-washed polyethylene bottles until analysis. Physico-chemical parameters such as pH, TDS, and EC in groundwater samples were determined *in situ* using a portable microcontroller water analysis kit NPC 362D. The instrument was calibrated using standard solutions that bracketed the expected values.

Instrumentation

The LED fluorimeter model LF-2a developed by Quantalase Enterprises Private Limited, Indore, India, was used for the analysis of uranium in drinking water. The instrument has lower and upper detection limits of 0.5 μg L^{-1} and 1,000 μg L^{-1}, respectively, with an accuracy of ±10%. The instrument consists primarily of three components: light-emitting diodes (LEDs) as a source of excitation, a sample section, and a photomultiplier tube (PMT) as a detector (Figure S1 in the Supplementary Material). The LED source emits ultraviolet radiation with wavelength 400 nm carrying 20 μJ energy and pulse duration of 20 μs at a repetition rate of 1,000 pulses per second excites the uranyl ions present in the water sample placed in the sample section. On de-excitation, green fluorescence emitted by uranyl ions is measured by sensitive PMT.

Sodium pyrophosphate ($Na_4P_2O_7.10H_2O$) solution (5%) was prepared in double-distilled water and its pH was adjusted to 7 by dropwise addition of orthophosphoric acid. This solution acts as a fluorescence-enhancing reagent. To give an estimate the concentration of uranium in a given sample, the instrument takes an average of 1,280 pulses. After every five samples, blank measurements are repeated to arrest memory effects and contamination. Details about the calibration of the instrument, analytical method, and quality control are given elsewhere (Kumar *et al.* 2016; Prasad *et al.* 2019). The concentration of uranium was estimated using the following equations (Sharma *et al.* 2019):

$$\text{Calibration factor (CF)} = \frac{\text{Uranium concentration in standard solution}}{\text{Fluorescence of standard} - \text{fluorescence of water}} \tag{1}$$

$$\text{Uranium concentration in unknown sample} = \text{CF} \times (\text{Fluorescence from sample} - \text{fluorescence from water}) \tag{2}$$

Age-dependent dose assessment

The uranium activity concentration was determined using a unit conversion factor of 1 μg L^{-1} = 0.02528 Bq L^{-1} (Sahoo *et al.* 2010; Rani *et al.* 2013). The annual effective dose for different age groups from ingestion of uranium in water was determined as (Bronzovic & Marovic 2005):

$$\text{Ingestion dose (Sv y}^{-1}) = U_a \times \text{DWI} \times \text{DCF} \times 365 \tag{3}$$

where U_a is the uranium activity concentration (Bq L^{-1}), DWI is the age-dependent daily water intake (L day^{-1}) prescribed by the Institute of Medicine of the National Academies (2005) and DCF is the dose conversion factor for specific age groups (Sv Bq^{-1}) given by the International Atomic Energy Agency (IAEA 2011).

Potential health risk assessment

The permissible limit for uranium in drinking water, as prescribed by various countries and environmental protection organizations, is very different. Therefore, it is better to compare the quality of groundwater on the basis of radiological and chemical toxicity caused by ingestion of uranium containing groundwater in the human body than to assess its suitability on the basis of its absolute concentration (Shin *et al.* 2016; Duggal *et al.* 2017; Sharma *et al.* 2019).

Radiological toxicity risk assessment

Radiological toxicity risk is expressed in terms of excess cancer risk (ECR), which was evaluated by multiplying the uranium activity concentration (U_a) (Bq L^{-1}) and risk factor (RF) (L Bq^{-1}) (USEPA 2000).

$$ECR = U_a \times RF \tag{4}$$

The risk factor (RF) was determined as follows:

$$RF\ (L\ Bq^{-1}) = RC \times IRW \times ED \tag{5}$$

where RC = uranium risk coefficient (Bq^{-1}), IRW = water ingestion rate (2 L day^{-1}) (WHO 2011) and ED = exposure duration (70 years), i.e. 70 × 365 = 25,550 days (WHO 2011; Rani *et al.* 2013). According to the USEPA (1999), the mortality and morbidity cancer risk coefficients of 1.13×10^{-9} Bq^{-1} and 1.73×10^{-9} Bq^{-1}, respectively, have been used for the estimation of cancer mortality risk and cancer morbidity risk of uranium over lifetime consumption of groundwater.

Chemical toxicity risk assessment

The chemical toxicity risk from exposure to uranium is quantified in terms of the lifetime average daily dose (LADD) and hazard quotient (HQ). LADD is defined as the quantity of uranium ingested per kilogram of body weight per day and was evaluated using the following equation (USEPA 2000; WHO 2011; Shin *et al.* 2016; Duggal *et al.* 2017).

$$LADD\ (\mu g kg^{-1} day^{-1}) = \frac{U \times IRW \times EF \times ED}{AT \times BW} \tag{6}$$

where U = uranium concentration in water (μg L^{-1}), IRW = water ingestion rate (2 L day^{-1}) (WHO 2011), EF = exposure frequency (365 days $year^{-1}$) (Ali *et al.* 2019), ED = exposure duration (70 years) (Rani *et al.* 2013), AT = average time (25,550 days) and BW = body weight (70 kg for the Indian standard person) (USEPA 2011; Duggal *et al.* 2017; Duggal & Rani 2018).

The HQ was calculated using the following equation:

$$HQ = \frac{LADD}{RfD} \tag{7}$$

where RfD = reference dose. Its value is 1.0 μg kg^{-1} day^{-1} (WHO 2011). If HQ is found to be less than unity, then no adverse health effects are expected due to the exposure of uranium.

RESULTS AND DISCUSSION

Uranium distribution in groundwater

Table 2 presents the summary statistics of uranium concentrations (μg L^{-1}) in groundwater. The uranium concentrations varied from 1.2 to 274 μg L^{-1} with a mean value of 32.5 μg L^{-1} and a median of 16.5 μg L^{-1}, and approximately 30.9% of the samples exceeded the WHO (2011) provisional guideline level (PGV) of 30 μg L^{-1} (Table S1 in the Supplementary Material). The standard deviation value is higher than the mean and median values, indicating that uranium concentrations are spread out over a wider range (Table 2). When comparing the mean and median values of uranium concentrations, it is observed that the data are not symmetrically distributed. It is a right-skewed distribution, as the mean value is approximately twice the median value. The kurtosis and skewness values are of a positive type. The data are highly skewed, which may be attributed to the variation in the geological formation of the study region. The measured uranium concentrations in groundwater follow a lognormal distribution. According to the literature survey, usually, the environmental samples and radioactive sampled data follow a lognormal distribution (Limpert *et al.* 2001).

The alluvium aquifers of the study region are composed of sand, clay and kankar and these aquifers are semi-confined, similar to the other alluvium aquifers in Northwest India, such as Punjab and Rajasthan (Bonsor *et al.* 2017).

Table 2 | Summary statistics of uranium concentrations (μg L^{-1}) in groundwater

Statistics	Uranium (μg L^{-1})	pH	TDS (mg L^{-1})	EC (μS cm^{-1})
N	68	68	68	68
Arithmetic mean	32.5	8.0	2,309	4,477
Median	16.5	8.0	1,605	2,975
Standard deviation	44.9	0.3	2,212	4,327
Maximum	274	9.2	13,150	26,200
Minimum	1.2	7.4	181	359
Mode	10	8.0	2,770	5,560
Standard error	5.4	0.04	268	525
Sample variance	2,013	0.09	4,893,549	18,721,339
Geometric mean	17.3	8.0	1,475	2,872
GSD	3.12	1.36	2.78	2.74
Skewness	3.2	0.59	2.2	2.31
Kurtosis	13	2.3	7.7	8.6
Sample numbers and proportion above the GV/PGV/AL/PL/MPL	21 (30.9%) samples exceeded the WHO PGV (30 μg L^{-1})	Only one sample exceeded BIS and WHO AL (6.5–8.5)	57 (83.8%) samples exceeded BIS AL (500 mg L^{-1}) and 30 (44%) samples exceeded BIS PL (2,000 mg L^{-1})	57 (83.8%) samples exceeded Water Act MPL (1,000 μS cm^{-1})

TDS, Total dissolved solids; EC, Electrical conductivity; BIS, Bureau of Indian Standards; WHO, World Health Organization; GV, Guideline value; PGV, Provisional guideline value; AL, Acceptable limit; MPL, Maximum permissible limit; PL, Permissible limit.

Many studies (Rani *et al.* 2013; Duggal *et al.* 2017; Mittal *et al.* 2017; Pant *et al.* 2017; Saini *et al.* 2018) have reported uranium concentrations above 30 μg L^{-1} in the states of Punjab and Rajasthan, which are associated primarily with alluvial aquifers. The findings of these studies are consistent with our results, which find high uranium concentrations in groundwater from alluvial aquifers. Mann *et al.* (2018) reported high activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in soil samples collected from the Sirsa, Fatehabad, Hisar and Bhiwani districts of Haryana.

Comparison of uranium concentrations with other studies

The observed concentrations were also compared with the worldwide literature (Table 3). The uranium concentrations in water samples from Canada, Sweden, Saudi Arabia and Northern Rajasthan; India are comparable with the present study. However, the uranium concentrations are higher in the drinking water of SW Punjab; India, NE Portugal and USA, whereas the uranium concentrations in Australia, Switzerland, Bangladesh, Mongolia, Korea, Kosovo, Jammu and Kashmir, Himachal Pradesh, Uttarakhand, Haryana, Uttar Pradesh, Bihar, Jharkhand, West Bengal, Chhattisgarh, Odisha, Maharashtra, Karnataka, Tamilnadu, and Kerala states of India are lower than the concentrations found in the present study.

Correlation between uranium and physicochemical parameters

The physicochemical parameters (pH, TDS and EC) of the groundwater samples are reported in Table 2 and Table S1. The groundwater of the study area is alkaline in nature. The acceptable limit for pH as prescribed by the Bureau of Indian Standards (BIS 2012) and WHO (2011) is 6.5–8.5. Only one water sample had a pH value above 8.5. The WHO (2011) has suggested that water pH has no direct impact on consumers. Activity increases the capacity of water to attack geological materials and leach toxic metals into the water, making it potentially harmful for human consumption. The TDS values in groundwater for whole of the studied area ranged from 181 to 13,150 mg L^{-1} with an average value of 2,309 mg L^{-1}. Approximately 84% of the samples had TDS

Table 3 | Uranium concentrations in groundwater sources worldwide

		Uranium concentration ($\mu g\ L^{-1}$)		
Sr. No.	**Region**	**Range**	**Mean**	**Reference**
1	USA	1.8–7,780	620	Orloff *et al.* (2004)
2	Canada	<1–845	–	Zamora *et al.* (2009)
3	Australia	0.05–160	2.1	Landstetter & Katzlberger (2009)
4	Switzerland	0.05–92.02	–	Stalder *et al.* (2012)
5	Bangladesh	<0.2–10	2.5	Frisbie *et al.* (2009)
6	Mongolia	<0.01–57	4.6	Nriagu *et al.* (2012)
7	Kosovo	0.012–166	5	Berisha & Goessler (2013)
8	Korea	0–3,610	8.0	Shin *et al.* (2016)
9	NE Portugal	8.2–3,483	617.8	Costa *et al.* (2017)
10	Sweden	<0.20–470	–	Seldén *et al.* (2009)
11	Saudi Arabia	<0.8–90.8	32.4	Shabana & Kinsara (2014)
12	Jammu and Kashmir, India	0.18–20.81	4.72	Kumar *et al.* (2016)
13	Himachal Pradesh, India	0.12–19.43	2.57	Kaur & Mehra (2019)
14	Garhwal Himalayan, Uttarakhand, India	0.02–63.7	7	Prasad *et al.* (2019)
15	Southwest Punjab, India	0.13–676	76.27	Saini *et al.* (2018)
16	Northern Rajasthan, India	2.54–133	38.48	Rani *et al.* (2013)
17	Haryana, India	1.07–40.25	17.91	Panghal *et al.* (2017)
18	Eastern Uttar Pradesh, India	11–63.33	–	Meher *et al.* (2015)
19	South Bihar, India	0.1–238.2	12.3	Kumar *et al.* (2018)
20	Jaduguda, Jharkhand, India	0.03–11.6	–	Patra *et al.* (2013)
21	West Bengal, India	<0.01–13.9	1.5	Rahman *et al.* (2015)
22	Balod, Chhattisgarh, India	0.56–78.93	–	Sar *et al.* (2018)
23	Ganjam, Odisha, India	<0.2–13.6	4.3	Mohapatra *et al.* (2015)
24	Mumbai, Maharashtra, India	1.1–10.6	4.8	Sahu *et al.* (2014)
25	Bangalore, Karnataka, India	0.2–770.1	–	Nagaiah *et al.* (2013)
26	Nalgonda, Andhra Pradesh, India	0.2–68	18.5	Brindha *et al.* (2011)
27	Central Tamilnadu, India	0.79–71.93	–	Adithya *et al.* (2019)
28	South Coast districts, Kerala, India	0.31–4.92	–	Byju *et al.* (2012)
29	Hisar district, Haryana	1.2–274	32.5	Present study

values above the acceptable limit of 500 mg L^{-1} and 44% samples exceeded the permissible limit of 2,000 mg L^{-1} recommended by BIS (2012). High concentration of TDS in the groundwater samples may be attributed to the dissolution or mineralization of organic and inorganic contents in aquifers. Residents of the study area are mostly illiterate farmers, who use this high TDS groundwater for irrigation and for domestic consumption without prior treatment. EC values varied from 359 to 26,200 $\mu S\ cm^{-1}$ with a mean value of 4,477 $\mu S\ cm^{-1}$. Approximately 84% of the samples had EC values above the maximum permissible limit of 1,000 $\mu S\ cm^{-1}$ recommended by the Water Act (1956).

In the present study, no correlation was observed between uranium and pH. A positive correlation was observed between uranium and TDS, indicating that the mobility of uranium in the groundwater was very much influenced and controlled by TDS (Figure 2). Due to high TDS in groundwater, solubility of uranium increases. All the groundwater samples followed

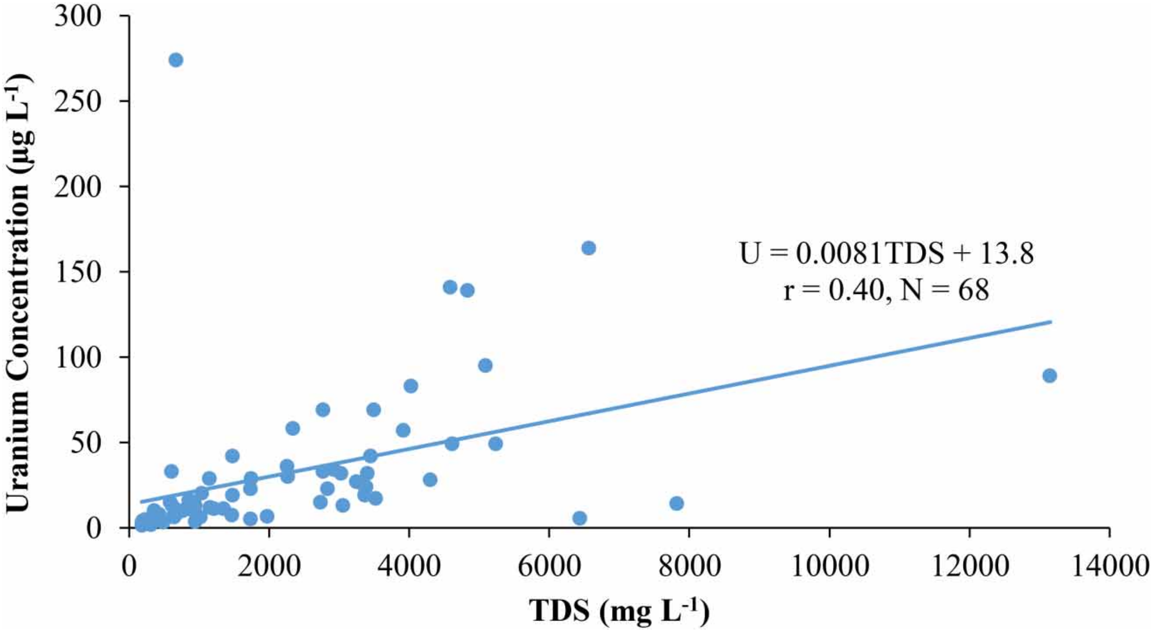

Figure 2 | Dependencies between uranium and TDS for the Hisar district. N denotes the number of correlated pairs and r the correlation coefficient.

the general rule; the higher the TDS (or EC), higher the radioactivity (Ortega *et al.* 1996; Singh *et al.* 2003; Kumar *et al.* 2011; Duggal *et al.* 2017).

Groundwater samples were collected from different depths ranging from 20 to 200 feet. No correlation was observed between uranium concentration and depth of the groundwater.

Age-dependent annual effective dose (AED)

The estimated AED due to intake of uranium through drinking water for various age groups varied from 1.3 to 688 μSv y^{-1}, with an average value of 54 μSv y^{-1} (Table 4). The large variations in the annual effective dose are due to the wide range of uranium concentrations in the investigated

Table 4 | Age-dependent annual effective doses (μSv y^{-1}) due to daily consumption of uranium through drinking water

			Annual effective dose (μSv y^{-1})				Number of samples exceeded the WHO & EU Council RDL
Life stage group	**Age group**	**DWI (L day^{-1})**	**Range**	**Mean**	**SD**	**Median**	
Infants	0–6 months	0.7	2.6–602	71	99	36	13
	7–12 months	0.8	3.0–688	82	113	41	15
Children	1–3 y	1.3	1.7–395	47	65	24	7
	4–8 y	1.7	1.5–344	41	56	21	7
Males	9–13 y	2.4	1.8–413	49	68	25	9
	14–18 y	3.3	2.4–559	66	92	34	11
	Adults	3.7	1.8–421	50	69	25	9
Females	9–13 y	2.1	1.6–361	43	59	22	7
	14–18 y	2.3	1.7–390	46	64	23	7
	Adults	2.7	1.3–307	36	50	18	5
Pregnancy	14–18 y	3.0	2.2–509	60	83	31	11
	19–50 y	3.0	1.5–342	41	56	21	7
Lactation	14–18 y	3.8	2.8–644	76	105	39	13
	19–50 y	3.8	1.9–433	51	71	26	9

DWI, Daily water intake; SD, Standard deviation.

groundwater samples. The calculated annual effective dose values for different age groups follow a lognormal distribution. The WHO (2011) guidelines (fourth edition) and the European Union Council Directive (2013) prescribed the measurement of reference dose level (RDL) of the AED received from drinking water ingestion at 100 μSv y^{-1}. This RDL is ˜4.2% of the average AED of 2.4 mSv y^{-1} from natural background radiation (UNSCEAR 2008; WHO 2011). Even though infants consume less water than other age groups, the AED is significantly higher in infants than in other age groups because of the differences in infants' metabolism and smaller organ weights, resulting in higher doses for many radionuclides (Duggal *et al.* 2017). The AED is slightly higher for the 7–12 months age group of infants as compared to 0–6 months due to higher annual water intake. Due to the lower water ingestion rate, all the age groups of females receive lower AEDs as compared to males. The higher AEDs during the lactation and pregnancy periods may be attributed to the need for more water in those periods.

Radiological toxicity risk

The cancer mortality and morbidity risks as evaluated for the people who consume this water for drinking purposes is presented in Table 5. Mortality indicates the incidence of fatal cancers and morbidity indicates the incidence of total cancers (fatal and non-fatal). The cancer mortality and morbidity risks ranged from 0.18×10^{-5} to 40×10^{-5} and 0.27×10^{-5} to 61×10^{-5} with average values of 4.7×10^{-5} and 7.3×10^{-5}, respectively. The estimated mean values of cancer mortality and morbidity risks were lower than the permissible limit of 1.67×10^{-4} as prescribed by AERB (2004). Some studies have reported 10^{-3} as the acceptable level for radiological risk (Tran *et al.* 2000; Kim *et al.* 2004). The cancer mortality risk from four locations (Dhani Raju H-6, Hansi H-17, Agroha H-53 and Balak H-67) exceeded the recommended permissible limit (Table S2 in the Supplementary Material). On the basis of maximum uranium concentrations, it can be concluded that due to continuous exposure to uranium through consumption of groundwater, there could be the occurrence of 40 cancer cases per one lakh of population. The values of cancer mortality risk are comparable to those reported in Northern Rajasthan, India (3.7×10^{-6} to 2.5×10^{-4}) by Duggal *et al.* (2017), Uttarakhand State, India (5.04×10^{-8} to 1.79×10^{-4}) by Prasad *et al.* (2019), Punjab State, India (1.34×10^{-6} to 1.80×10^{-3}) by Saini *et al.* (2016), Sonipat district, Haryana (2.60×10^{-5} to 4.39×10^{-4}) and Panipat district, Haryana (4.2×10^{-5} to 3.49×10^{-4}) by Daulta *et al.* (2018).

Table 5 | Cancer mortality risk, cancer morbidity risk, LADD and other statistical parameters for groundwater samples

Statistical parameters	Cancer mortality risk (10^{-5})	Cancer morbidity risk (10^{-5})	LADD (μg kg^{-1} day^{-1})
Mean	4.7	7.3	0.93
Median	2.4	3.7	0.47
Minimum	0.18	0.27	0.03
Maximum	40	61	7.83
SD	6.6	10	1.28
Q_{25}	1.2	1.8	0.23
Q_{75}	4.9	7.4	0.95
P_{10}	0.64	0.98	0.13
P_{90}	10.7	16.4	2.09

SD, Standard deviation; Q25, 1st quartile; Q75, 3rd quartile; P10, 10th percentile; P90, 90th percentile; LADD, lifetime average daily dose.

Chemical toxicity risk

The lifetime average daily dose (LADD) of uranium due to ingestion of groundwater varied from 0.03 to 7.83 μg kg^{-1} day^{-1} and the HQ accordingly has the same numerical values because RfD is unity (WHO 2011) (Table 5). Approximately 23.5% of the samples showed a hazard quotient greater than unity (Table S2 in the Supplementary Material), indicating a significant risk due to the chemical toxicity of uranium. Therefore, periodic monitoring and management are needed for these areas. The LADD observed in the present study is comparable to those reported in Northern Rajasthan, India (0.07 to 4.89 μg kg^{-1} day^{-1}) by Duggal *et al.* (2017), Uttarakhand State, India (0.001 to 3.69 μg kg^{-1} day^{-1}) by Prasad *et al.* (2019), Jammu district, Jammu & Kashmir, India (0.01 to 1.52 μg kg^{-1} day^{-1}) by Kumar *et al.* (2016), Sonipat district, Haryana (0.616 to 10.383 μg kg^{-1} day^{-1}) and Panipat district, Haryana (0.998 to 8.259 μg kg^{-1} day^{-1}) by Daulta *et al.* (2018).

CONCLUSIONS

- The results show that 30.9% of the samples exceeded the WHO PGV.
- Due to high TDS in groundwater of the study region, solubility of uranium increases.
- The mean AEDs for all age groups were well below the recommended RDL of $100\ \mu\mathrm{Sv\ y}^{-1}$. The infants have received relatively high mean AEDs compared to other age groups.
- The mean values of cancer mortality risk and cancer morbidity risk were well below the AERB's permissible limit.
- Approximately 23.5% of the samples showed $HQ > 1$, indicating chemical toxicity due to the presence of uranium in groundwater, therefore, unsuitable for drinking.
- Remedial action is required at sampling sites with a high concentration of uranium.

CONFLICTS OF INTEREST

The authors declare that they do not have any conflict of interest.

FUNDING

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

ACKNOWLEDGEMENTS

The authors are thankful to the residents of the study area for their cooperation during the field work and Department of Physics, Guru Nanak Dev University, Amritsar for providing state-of-the-art laboratory facilities.

DATA AVAILABILITY STATEMENT

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

REFERENCES

Adithya, V. S., Chidambaram, S., Keesari, T., Mohokar, H. V. & Prasanna, M. V. 2019 Occurrence of uranium in groundwater along the lithological contacts in Central Tamilnadu, India: an isotope hydrogeochemical perspective. *Exposure and Health* **11**, 277–290.

AERB 2004 *Drinking Water Specifications in India*. Atomic Energy Regulatory Board, Department of Atomic Energy, Mumbai, India.

Ali, W., Aslam, M. W., Feng, C., Junaid, M., Ali, K., Li, S., Chen, Z., Yu, Z., Rasool, A. & Zhang, H. 2019 Unraveling prevalence and public health risks of arsenic, uranium and co-occurring trace metals in groundwater along riverine ecosystem in Sindh and Punjab, Pakistan. *Environmental Geochemistry and Health* **41**, 2223–2238. https://doi.org/10.1007/s10653-019-00278-7.

ATSDR 2013 *Toxicological Profile for Uranium*. U.S. Department of Health and Human Services, Agency for Toxic Substances and Disease Registry, Atlanta, GA, USA, pp. 1–526.

Bajwa, B. S., Kumar, S., Singh, S., Sahoo, S. & Tripathi, R. M. 2017 Uranium and other heavy toxic elements distribution in the drinking water samples of SW-Punjab, India. *Journal of Radiation Research and Applied Sciences* **10**, 13–19.

Berisha, F. & Goessler, W. 2013 Uranium in Kosovo's drinking water. *Chemosphere* **93**, 2165–2170.

Bonsor, H. C., MacDonald, A. M., Ahmed, K. M., Burgess, W. G., Basharat, M., Calow, R. C., Dixit, A., Foster, S. S. D., Gopal, K., Lapworth, D. J., Moench, M., Mukherjee, A., Rao, M. S., Shamsudduha, M., Smith, L., Taylor, R. G., Tucker, J., Steenbergen, F. V., Yadav, S. K. & Zahid, A. 2017 Hydrogeological typologies of the Indo-Gangetic basin alluvial aquifer, South Asia. *Hydrogeology Journal* **25**, 1377–1406.

Brindha, K., Elango, L. & Nair, R. N. 2011 Spatial and temporal variation of uranium in a shallow weathered and rock aquifer in southern India. *Journal of Earth System Science* **120**, 911–920.

Bronzovic, M. & Marovic, G. 2005 Age-dependent dose assessment of ^{226}ra from bottled water intake. *Health Physics* **88**, 480–485.

Bundesministerium für Gesundheit 2011 *Erste Verordnung zur Änderung der Trinkwasserverordnung, Bundesgesetzblatt Teil I, 748-774.*

Bureau of Indian Standards 2012 *Indian Standard Drinking Water – Specification (Second Revision)*. IS 10500:2012, New Delhi, India.

Byju, S. B., Sunil, A., Reeba, M. J., Christa, E. P., Vaidyan, V. K., Prasad, R. & Jojo, P. J. 2012 Uranium in drinking water from the south coast districts of Kerala, India. *Iranian Journal of Radiation Research* **10**, 31–36.

Central Ground Water Board (CGWB) 2017 *Report on Aquifer Mapping and Management Plan, Hisar District, Haryana.* Ministry of Water Resources, River Development and Ganga Rejuvenation, Government of India.

Costa, M. R., Pereira, A. J. S. C., Neves, L. J. P. F. & Ferreira, A. 2017 Potential human health impact of groundwater in non-exploited uranium ores: the case of Horta da Vilariça (NE Portugal). *Journal of Geochemical Exploration* **183**, 191–196.

Cothern, C. R. & Lappenbusch, W. L. 1983 Occurrence of uranium in drinking water in the US. *Health Physics* **45**, 89–99.

Daulta, R., Singh, B., Kataria, N. & Garg, V. K. 2018 Assessment of uranium concentration in the drinking water and associated health risks in Eastern Haryana, India. *Human and Ecological Risk Assessment: An International Journal* **24**, 1115–1126.

Duggal, V. & Rani, A. 2018 Carcinogenic and non-carcinogenic risk assessment of metals in groundwater via ingestion and dermal absorption pathways for children and adults in Malwa Region of Punjab. *Journal of the Geological Society of India* **92**, 187–194.

Duggal, V., Rani, A., Mehra, R., Saini, K. & Bajwa, B. S. 2017 Assessment of age-dependent radiation dose and toxicity risk due to intake of uranium through the ingestion of groundwater from Northern Rajasthan, India. *Toxicological & Environmental Chemistry* **99**, 516–524.

Duggal, V., Sharma, S. & Mehra, R. 2020 Risk assessment of radon in drinking water in Khetri Copper Belt of Rajasthan, India. *Chemosphere* **239**, 124782. https://doi.org/10.1016/j.chemosphere.2019.124782.

Environmental Public Health (Quality of Piped Drinking-water) Regulations, Singapore 2008.

European Food Safety Authority 2009 *Uranium in Foodstuffs, in Particular Mineral Water*. Scientific opinion of the Panel on Contaminants in the Food Chain. The EFSA Journal 1018, 1–59.

European Union Council Directive 2013 *Council Directive 2013/51/EURATOM of 22 October 2013 Laying Down Requirements for the Protection of the Health of the General Public with Regard to Radioactive Substances in Water Intended for Human Consumption*. Official Journal of the European Union, L 296/12.

Frisbie, S. H., Mitchell, E. J., Mastera, L. J., Maynard, D. M., Yusuf, A. Z., Siddiq, M. Y., Ortega, R., Dunn, R. K., Westerman, D. S., Bacquart, T. & Sarkar, B. 2009 Public health strategies for Western Bangladesh that address arsenic, manganese, uranium, and other toxic elements in drinking water. *Environmental Health Perspectives* **117**, 410–416.

Health Canada 2019 *Guidelines for Canadian Drinking Water Quality-Summary Table*. Water and Air Quality Bureau, Healthy Environments and Consumer Safety Branch, Health Canada, Ottawa, ON, Canada.

IAEA 2011 *Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards*. Interim Edition, General Safety Requirements Part 3, No. GSR Part 3 (Interim). International Atomic Energy Agency, Vienna, Austria.

Institute of Medicine of the National Academies 2005 *Dietary Reference Intakes for Water, Potassium, Sodium, Chloride, and Sulphate. Panel on Dietary Reference Intakes for Electrolytes and Water. Standing Committee on the Scientific Evaluation of Dietary References Intake*. Food and Nutrition Board. Institute of Medicine of the National Academies. The National Academies Press, Washington, DC, USA.

Japan Ministerial Ordinance Concerning Drinking-water Quality Standards 2010.

Kaur, S. & Mehra, R. 2019 Toxicological risk assessment of protracted ingestion of uranium in groundwater. *Environmental Geochemistry and Health* **41**, 681–698.

Kim, Y. S., Park, H. S., Kim, J. Y., Park, S. K., Cho, B. W., Sung, I. H. & Shin, D. C. 2004 Health risk assessment for uranium in Korean groundwater. *Journal of Environmental Radioactivity* **77**, 77–85.

Kumar, A., Rout, S., Narayanan, U., Mishra, M. K., Tripathi, R. M., Singh, J., Kumar, S. & Kushwaha, H. S. 2011 Geochemical modelling of uranium speciation in the subsurface aquatic environment of Punjab State in India. *Journal of Geology and Mining Research* **3**, 137–146.

Kumar, A., Kaur, M., Mehra, R., Sharma, S., Mishra, R., Singh, K. P. & Bajwa, B. S. 2016 Quantification and assessment of health risk due to ingestion of uranium in groundwater of Jammu district, Jammu & Kashmir, India. *Journal of Radioanalytical and Nuclear Chemistry* **310**, 793–804.

Kumar, D., Singh, A., Jha, R. K., Sahoo, S. K. & Jha, V. 2018 Using spatial statistics to identify the uranium hotspot in groundwater in the mid-eastern Gangetic plain, India. *Environmental Earth Sciences* **77**, 702. https://doi.org/10.1007/s12665-018-7889-1.

Landstetter, C. & Katzlberger, C. 2009 Determination of ^{3}H, ^{226}ra, ^{222}rn and ^{238}u in Austrian ground- and drinking water. *Journal of Radioanalytical and Nuclear Chemistry* **282**, 467–471.

Limpert, E., Stahel, W. A. & Abbt, M. 2001 Log-normal distribution across the sciences: keys and clues. *BioScience* **51**, 341–352.

Mann, N., Kumar, A., Kumar, S. & Chauhan, R. P. 2018 Measurement of radium, thorium and potassium and associated hazard indices from the soil samples collected from Northern India. *Indoor and Built Environment* **27**, 1149–1156.

Meher, P. K., Sharma, P., Khare, A. & Mishra, K. P. 2015 Uranium in ground water of Eastern Uttar Pradesh, India: a preliminary study. *International Research Journal of Environment Sciences* **4**, 70–74.

Ministerio de Salud, Lima-Perú 2011 *Reglamento de la Calidad del Agua Para Consumo Humano, DS N° 031-2010-SA*. Dirección General de Salud Ambiental, Lima, Perú.

Ministry of Health Malaysia 2004 *National Standard for Drinking Water Quality*. Engineering Services Division, Ministry of Health Malaysia.

Ministry of Health, New Zealand 2008 *Drinking-water Standards for New Zealand 2005 (Revised 2008)*. Wellington, New Zealand.

Mittal, S., Rani, A., Mehra, R., Balaram, V., Satyanarayanan, M. & Sawant, S. S. 2017 Assessment of uranium in correlation with physico-chemical properties of drinking water of Northern Rajasthan. *Journal of the Geological Society of India* **90**, 233–238.

Mohapatra, S., Sahoo, S. K., Dubey, J. S., Patra, A. C., Thakur, V. K., Tripathy, S. K., Sagar, D. V., Godbole, S. V., Ravi, P. M. & Tripathi, R. M. 2015 Characterization of uranium and its progenies in drinking water and assessment of dose to public around a NHBRA, Odisha, India. *Journal of Radioanalytical and Nuclear Chemistry* **303**, 601–613.

Nagaiah, N., Mathews, G., Balakrishna, K., Rajanna, A. M. & Naregundi, K. 2013 Influence of physico-chemical parameters on the distribution of uranium in the ground water of Bangalore, India. *Radiation Protection and Environment* **36**, 175–180.

NHMRC, NRMMC 2011 *Australian Drinking Water Guidelines Paper 6 National Water Quality Management Strategy*. National Health and Medical Research Council, National Resource Management Ministerial Council, Commonwealth of Australia, Canberra, Australia.

Nriagu, J., Nam, D. H., Ayanwola, T. A., Dinh, H., Erdenechimeg, E., Ochir, C. & Bolormaa, T. A. 2012 High levels of uranium in groundwater of Ulaanbaatar, Mongolia. *Science of the Total Environment* **414**, 722–726.

Omani Standard 2012 *OS 8/2012, Un Bottled Drinking-Water*.

Orloff, K. G., Mistry, K., Charp, P., Metcalf, S., Marino, R., Shelly, T., Melaro, E., Donohoe, A. M. & Jones, R. L. 2004 Human exposure to uranium in groundwater. *Environmental Research* **94**, 319–326.

Ortega, X., Valles, I. & Serrano, I. 1996 Natural radioactivity in drinking water in Catalonia (Spain). *Environment International* **22**, 347–354.

Panghal, A., Kumar, A., Kumar, S., Singh, J., Sharma, S., Singh, P., Mehra, R. & Bajwa, B. S. 2017 Radiation dose-dependent risk on individuals due to ingestion of uranium and radon concentration in drinking water samples of four districts of Haryana, India. *Radiation Effects and Defects in Solids* **172**, 441–455.

Pant, D., Keesari, T., Sharma, D., Rishi, M., Singh, G., Jaryal, A., Sinha, U. K. & Tripathi, R. M. 2017 Study on uranium contamination in groundwater of Faridkot and Muktsar districts of Punjab using stable isotopes of water. *Journal of Radioanalytical and Nuclear Chemistry* **313**, 635–639.

Patra, A. C., Mohapatra, S., Sahoo, S. K., Lenka, P., Dubey, J. S., Tripathi, R. M. & Puranik, V. D. 2013 Age-dependent dose and health risk due to intake of uranium in drinking water from Jaduguda, India. *Radiation Protection Dosimetry* **155**, 210–216.

Peraturan Menteri Kesehatan Republik Indonesia 2010 *NOMOR 492/MENKES/PER/IV/2010*.

Prasad, M., Kumar, G. A., Sahoo, S. K. & Ramola, R. C. 2019 Health risks associated with exposure to uranium and heavy metals through potable groundwater in Uttarakhand state of India. *Journal of Radioanalytical and Nuclear Chemistry* **319**, 13–21.

Rahman, M. M., Dong, Z. & Naidu, R. 2015 Concentrations of arsenic and other elements in groundwater of Bangladesh and West Bengal, India: potential cancer risk. *Chemosphere* **139**, 54–64.

Rani, A., Mehra, R. & Duggal, V. 2013 Analysis of uranium concentration in drinking water samples using ICPMS. *Health Physics* **104**, 251–255.

Sahoo, S. K., Mohapatra, S., Chakrabarty, A., Sumesh, C. G., Jha, V. N., Tripathi, R. M. & Puranik, V. D. 2010 Determination of uranium at ultra trace level in packaged drinking water by laser fluorimeter and consequent ingestion dose. *Radioprotection* **45**, 55–66.

Sahu, S. K., Maity, S., Bhangare, R. C., Pandit, G. G. & Sharma, D. N. 2014 *Determination of Uranium in Ground Water Using Different Analytical Techniques*. Government of India Atomic Energy Commission, Bhabha Atomic Research Centre, Mumbai, BARC/2014/E/011, pp. 1–30.

Saini, K., Singh, P. & Bajwa, B. S. 2016 Comparative statistical analysis of carcinogenic and non-carcinogenic effects of uranium in groundwater sample from different regions of Punjab, India. *Applied Radiation and Isotopes* **118**, 196–202.

Saini, K., Duggal, V. & Bajwa, B. S. 2018 Assessment of radiation dose due to intake of uranium through groundwater and its carcinogenic and non-carcinogenic risks in southwest and northeast Punjab, India. *Indoor and Built Environment* **27**, 983–991.

Sar, S. K., Diwan, V., Biswas, S., Singh, S., Sahu, M., Jindal, M. K. & Arora, A. 2018 Study of uranium level in groundwater of Balod district of Chhattisgarh State, India and assessment of health risk. *Human and Ecological Risk Assessment: An International Journal* **24**, 691–698.

Seldén, A. I., Lundholm, C., Edlund, B., Hogdahl, C., Ek, B. M., Bergstrom, B. E. & Ohlson, C. G. 2009 Nephrotoxicity of uranium in drinking water from private drilled wells. *Environmental Research* **109**, 486–494.

Shabana, E. I. & Kinsara, A. A. 2014 Radioactivity in the groundwater of a high background radiation area. *Journal of Environmental Radioactivity* **137**, 181–189.

Sharma, S., Duggal, V., Srivastava, A. K. & Mehra, R. 2017 Assessment of radiation dose from exposure to radon in drinking water from Western Haryana, India. *International Journal of Environmental Research* **11**, 141–147.

Sharma, T., Sharma, A., Kaur, I., Mahajan, R. K., Litoria, P. K., Sahoo, S. K. & Bajwa, B. S. 2019 Uranium distribution in groundwater and assessment of age dependent radiation dose in Amritsar, Gurdaspur and Pathankot districts of Punjab, India. *Chemosphere* **219**, 607–616.

Shin, W., Oh, J., Choung, S., Cho, B. W., Lee, K. S., Yun, U., Woo, N. C. & Kim, H. K. 2016 Distribution and potential health risk of groundwater uranium in Korea. *Chemosphere* **163**, 108–115.

Singh, S., Rani, A., Mahajan, R. K. & Walia, T. P. S. 2003 Analysis of uranium and its correlation with some physico-chemical properties of drinking water samples from Amritsar, Punjab. *Journal of Environmental Monitoring* **5**, 917–921.

South African National Standard 2011 *SANS241:2011 Drinking-Water*.

Stalder, E., Blanc, A., Haldimann, M. & Dudler, V. 2012 Occurrence of uranium in Swiss drinking water. *Chemosphere* **86**, 672–679.

The Russian Federation 2003 *State Sanitary and Epidemiological Rules and Regulations, the Most Acceptable Concentrations (MAC) Chemicals in Water Aquatic Facilities Casting and Cultural Household Water use Hygiene Standards*. Ministry of Health of Russia, Moscow, Russia.

Tran, N. L., Locke, P. A. & Burke, T. A. 2000 Chemical and radiation environmental risk management: differences, commonalities, and challenges. *Risk Analysis* **20**, 163–172.

Uganda Standard 2008 *US 201:2008 Drinking (Potable) Water – Specification*.

UNSCEAR 2008 *Report to General Assembly with Scientific Annexes*. United Nations Scientific Committee on the Effects of Atomic Radiations. United Nations Sales Publications, United Nations, New York, NY, USA.

USEPA 1999 *Cancer Risk Coefficients for Environmental Exposure to Radionuclides*. Federal Guidance Report No. 13. United States Environmental Protection Agency, EPA 402-R-99-001, Washington, DC, USA.

USEPA 2000 *National Primary Drinking Water Regulations; Radionuclides*. Final Rule, 40 CFR Parts 9, 141 and 142, Washington, DC, USA.

USEPA 2011 *Edition of the Drinking Water Standards and Health Advisories*. United States Environmental Protection Agency, EPA 820-R-11-002, Office of Water, Washington, DC, USA.

Water Act 1956 The Water Act (Act 54 of 1956) and its requirements in terms of water supplies for drinking water and for waste water treatment and discharge into the environment.

WHO 2011 *Guidelines for Drinking-Water Quality*, 4th edn. World Health Organization, Geneva, Switzerland, pp. 1–564.

Zamora, M. L., Zielinski, J. M., Moodie, G. B., Falcomer, R. A., Hunt, W. C. & Capello, K. 2009 Uranium in drinking water: renal effects of long-term ingestion by an aboriginal community. *Archives of Environmental & Occupational Health* **64**, 228–241.

First received 30 July 2020; accepted in revised form 30 October 2020. Available online 11 November 2020