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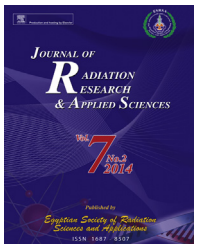

Uranium and other heavy toxic elements distribution in the drinking water samples of SW-Punjab, India

B.S. Bajwa [a,*], Sanjeev Kumar [a], Surinder Singh [a], S.K. Sahoo [b], R.M. Tripathi [b]

[a] Department of Physics, Guru Nanak Dev University, Amritsar, Punjab 143005, India
[b] Health Physics Division, Bhabha Atomic Research Centre, Mumbai 400085, India

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ABSTRACT

In the present investigations, Laser Fluorimetry technique has been used for the micro-analysis of uranium content in drinking water samples collected from different sources like the hand pumps, tube wells of various depths from wide range of locations in the four districts of SW-Punjab, India. The purpose of this study was to investigate the uranium concentration levels of ground water being used for drinking purposes and to determine its health effects, if any, to the local population of this region. Corresponding radiological and chemical risks have also been calculated for the uranium concentrations in ground water samples. Some other heavy elements have also been analysed using the Atomic Absorption Spectrometry. In this region, uranium concentration in 498 drinking water samples has been found to vary between 0.5–579 $\mu gl^{-1}$with an average of 73.5 μgl^{-1}. Data analysis revealed that 338 of 498 samples had uranium concentration higher than recommended safe limit of 30 μgl^{-1} (WHO, 2011) while 216 samples exceeded the threshold of 60 μgl^{-1} recommended by AERB, DAE, India, 2004.

1. Introduction

Chronic exposure of uranium radionuclide in drinking water is a potential health risk (Blantz, Pelayo, Gushwa, Myers, & Evan, 1985). Although ubiquitous in the environment, uranium has no known metabolic function in animals and is currently regarded as non-essential. Uranium accumulated in human results in chemical and radioactive effects. The principal sites of uranium deposition in the body are the kidneys, the liver and the bones. The toxicity of uranium is a function of the route of exposure, particle solubility, contact time, and route of elimination (ATSDR, 1999). The concentrations of radiotoxic elements like uranium in drinking water are hence kept under vigil by different health organizations. The World Health Organization (WHO, 2004) had earlier recommended a reference level 15 μgl^{-1} but now the permissible limit of U in drinking water by WHO is 30 μgl^{-1} (WHO, 2011). The reference level is derived from epidemiological studies, based on the assumption of a 60 kg adult consuming 2 litres of drinking

* *Corresponding author.*
E-mail address: bsbajwa1@gmail.com (B.S. Bajwa).
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water per day and 80% allocation of the Tolerable Daily Intake (TDI) to drinking water. Maximum acceptable level of U in drinking water as per guidelines of India's Atomic Energy Regulatory Board, Department of Atomic Energy, is 60 μgl^{-1} (AERB, 2004).

Physico-chemical parameters (conductivity, pH, salinity, TDS and temperature etc.) of water are important in the sense that these parameters can provide important first hand in-situ information about the suitability of water for drinking purposes apart from being helpful in studying and modelling of speciation of radionuclides and anthropogenic elements in aquatic environment (Kumar et al., 2011). Total dissolved solids (TDS) comprise inorganic salts (principally calcium, magnesium, potassium, sodium, bicarbonates, chlorides and sulphates) and small amounts of organic matter that are dissolved in water. The source of these solids in drinking-water may be a natural one or a sewage, urban run-off and industrial wastewater. Concentrations of TDS in water vary considerably in different geological regions owing to differences in the solubility of minerals. The pH is monitoring parameter to assess aquatic ecosystem health, irrigation sources and discharges, live stock, drinking water sources, industrial discharges and intakes. ORP (Redox Potential) is a measurement of water's ability to oxidize contaminants. Higher ORP simply indicates the greater number of oxidizing agents. In generalized terms for human health, a lower ORP is preferred for consumption due to the high anti-oxidant value. Though, the WHO has not set a standard for ORP in drinking water, anything below −550 mV is considered too strong and not recommended for drinking.

Keeping in view the above mentioned factors and particularly radio-toxicity caused by U, in the present investigations an attempt has been made to evaluate the uranium and other trace elements concentrations in drinking water samples collected from four SW districts (Bathinda, Mansa, Faridkot and Ferozepur) of Punjab, India. The purpose of this study was mainly to investigate the uranium and other trace elements concentration levels in drinking water being utilized by the inhabitants of study regions and its health risk assessment and thus finally to crosscheck their variation with respect to recommendations given by various national/international organizations, from purely the health hazard point of view. Other water quality chemical parameters viz. pH, conductivity, TDS and salinity have also been monitored in the drinking water samples of the study region.

2. Geology of the study region

The geographical location of the study region of SW-Punjab is between latitude 29° 07′ to 30° 57′ N and longitude 74° 05′ to 76° 55′ E at an average elevation of 200 m from the mean sea level. The soil of the study area is loose, sandy, calcareous and alluvial, which is an admixture of gravel, sand, silt and clay in varying proportions. The lands in the study are is used for agriculture all over the year but many industries like thermal power plants, fertilizer factories, chemical factories, cement factories are established in the region. The map of the study region and the geological map of Punjab is given in Figs. 1 & 2 respectively. In the study area, the thickness of the alluvium varies with space and steadily decreases southwards where the basement is at shallow depth. This heterogeneous alluvium ranges in age from the upper pliestocene to recent and is generally classified into older and newer alluvium. These quarternary alluvial deposits consists of clay, gravel, sand, siltysands and silty clays with varying proportions of kankar (Official Website of Punjab, 2011). In the western and southwestern parts, this alluvium is occupied by wind blown, fine grained, buff coloured sand in the form of dunes. Study areas lie on the crest of postulated Aravali-Delhi ridge. The area is also in close vicinity of large evaporates basin widely known as trans-Aravali Vindhyan basin. *The highest part of the Aravali-Delhi ridge which is at about 400 meter depth passes through Sirsa-Mansa and Faridkot area. The basement rocks go down rapidly from Tusam to Bathinda. East of Sirsa, rocks of Malani suite, granites and rhyolites and Delhi quartzite are encountered below the quaternary sediments. At Zira, near Ferozpur, granites rocks are met at a depth of 700 meter below Siwalik sediments.* At Adampur, the basement is encountered at a depth of 2500 meter. The maximum depth to basement in Punjab plains is about 4–5 km and depth increases to some extent under Siwaliks.

3. Experimental techniques

3.1. *Uranium estimation in water samples using fluorimetry*

Laser fluorimeter manufactured by Quantalase Enterprises Pvt. Ltd., Indore, India was used for analyses of water samples for uranium concentrations in this work.

3.1.1. *Analytic procedure*

Analytic procedure begins with taking 10 ml aliquot of filtered water sample in duplicate for wet digestion ($HClO_4$ and HNO_3) on hot plates to destroy organic material in the sample. The residue was then dissolved in Millipore elix-3 water followed by mixing with fluorescence reagent (5% sodium pyrophosphate) to make the total volume 25 ml and to adjust pH to 7.2 levels. The sample solution was then taken into a cuvette for the measurement of U concentration. The instrument was calibrated in the range of 1–100 μgl^{-1} using a stock solution of standard that was prepared by dissolving 1.78 g of uranyl acetate dihydrate $(CH_3COO)_2UO_2 \cdot 2H_2O$) in 1*l* of Millipore elix-3 water containing 1 ml of HNO_3 (70% pure). 5% phosphoric acid in ultra pure water was used as fluorescence reagent. To obtain blank counts, a blank sample containing same amount of fluorescing reagent was measured for U concentration. All the reagents used for experimental work were of ultrapure/analytical grade (Merck, Mumbai, India). Quality assurance was made by analysis of IAEA standard reference materials, spike recovery, replicate analysis, and cross method checking. The concentration of uranium in samples using Laser Flourimeter can be calculated by

$$U\ (\mu g/l) = \frac{D1}{D1 - D2} \times \frac{V1}{V2} C \quad (1)$$

where, D_1 is fluorescence due to sample only, D_2 is fluorescence due to sample and Uranium standard spiked, V_1 is the volume of U standard added, V_2 is the volume of sample taken

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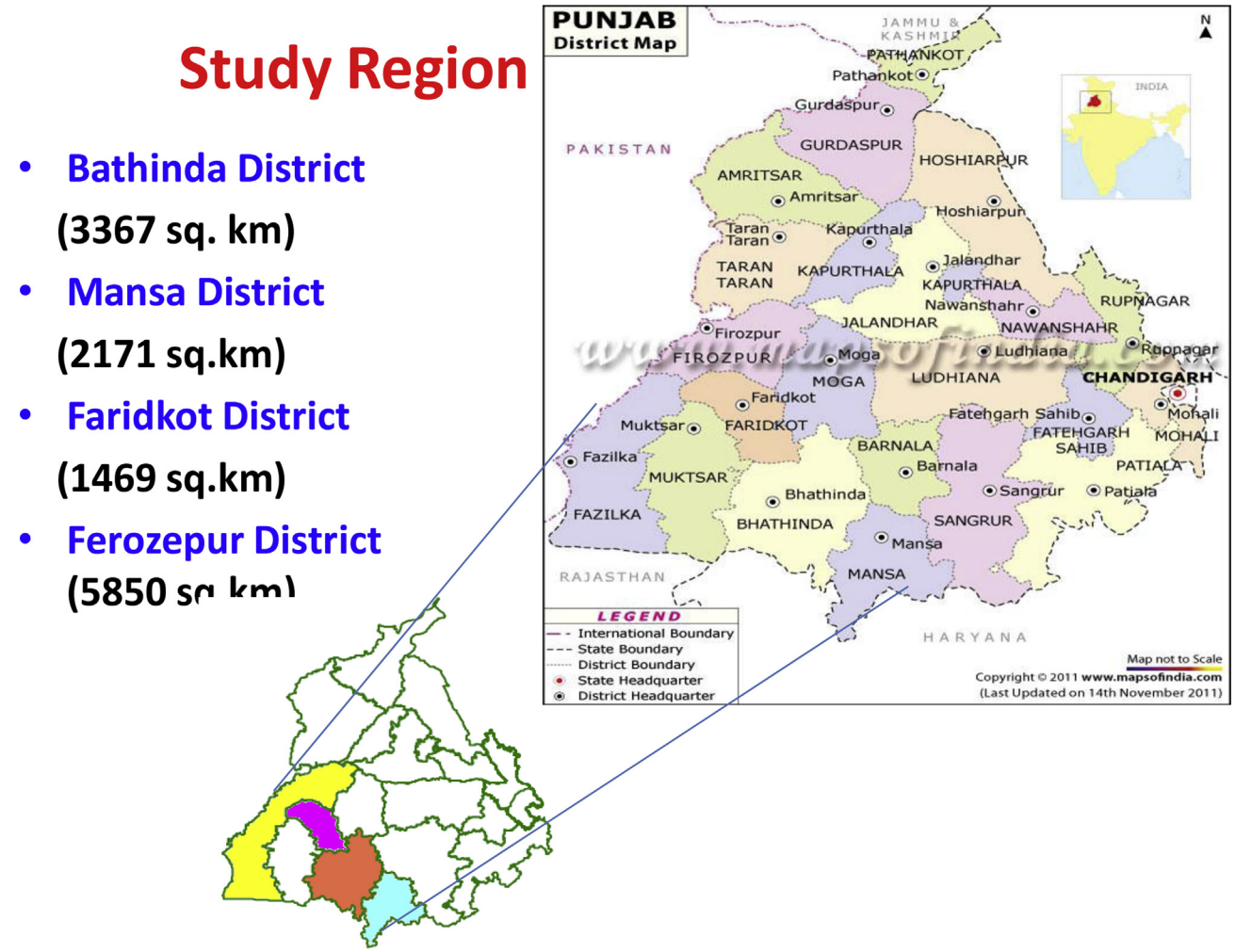

Fig. 1 – Punjab map showing the study region.

and C is the a concentration of U standard solution. The relative standard deviation (RSD) of measurements was calculated to be approximately 10%.

3.1.2. *Health risk assessment*

Health effects of uranium due to ingestion via drinking water can be divided into two types: carcinogenic (radiological risk) and non-carcinogenic (chemical risk). Below is described methodology adopted to estimate radiological and chemical toxicity due to calculated uranium concentrations in the water samples.

3.1.3. *Radiological risk assessment*

The radiological risk (excess cancer risk) due to ingestion of natural U in drinking water has been evaluated based on the USEPA standard method (USEPA, 2000).

$$\text{Excess Cancer Risk} = \text{Activity Conc. Uranium}\left(\text{Bql}^{-1}\right) \times \text{Risk Factor}\left(\text{per Bql}^{-1}\right) \quad (2)$$

The risk factor R (per Bql^{-1}), associated with intake of Uranium nuclide can be estimated by product of the risk coefficient (r) of Uranium (1.19×10^{-9}) for mortality and per capita activity intake I as

$$R = r \times I \quad (3)$$

Per capita activity intake I of Uranium again can be calculated using 63.7 years i.e.23250 days as life expectancy and daily consumption of water as 4.05 lday^{-1} (HDR, 2009).

$$I = 4.05\ \text{lday}^{-1} \times 23250\ \text{days} \quad (4)$$

3.1.4. *Chemical toxicity risk*

The chemical toxicity risk for a given element is defined in terms of Average Daily Dose (ADD) of the element through drinking water intake. For an observed Exposure Point Concentration (EPC), in μgl^{-1} units, of a given contaminant, average daily dose is the quantity of chemical substance ingested per kilogram of body weight per day is given by following equation (Lee, Chon, & Kim, 2005; Health Canada,1999).

$$\text{ADD} = \frac{C \times IR \times ED \times EF}{BW \times AT \times 365} \quad (5)$$

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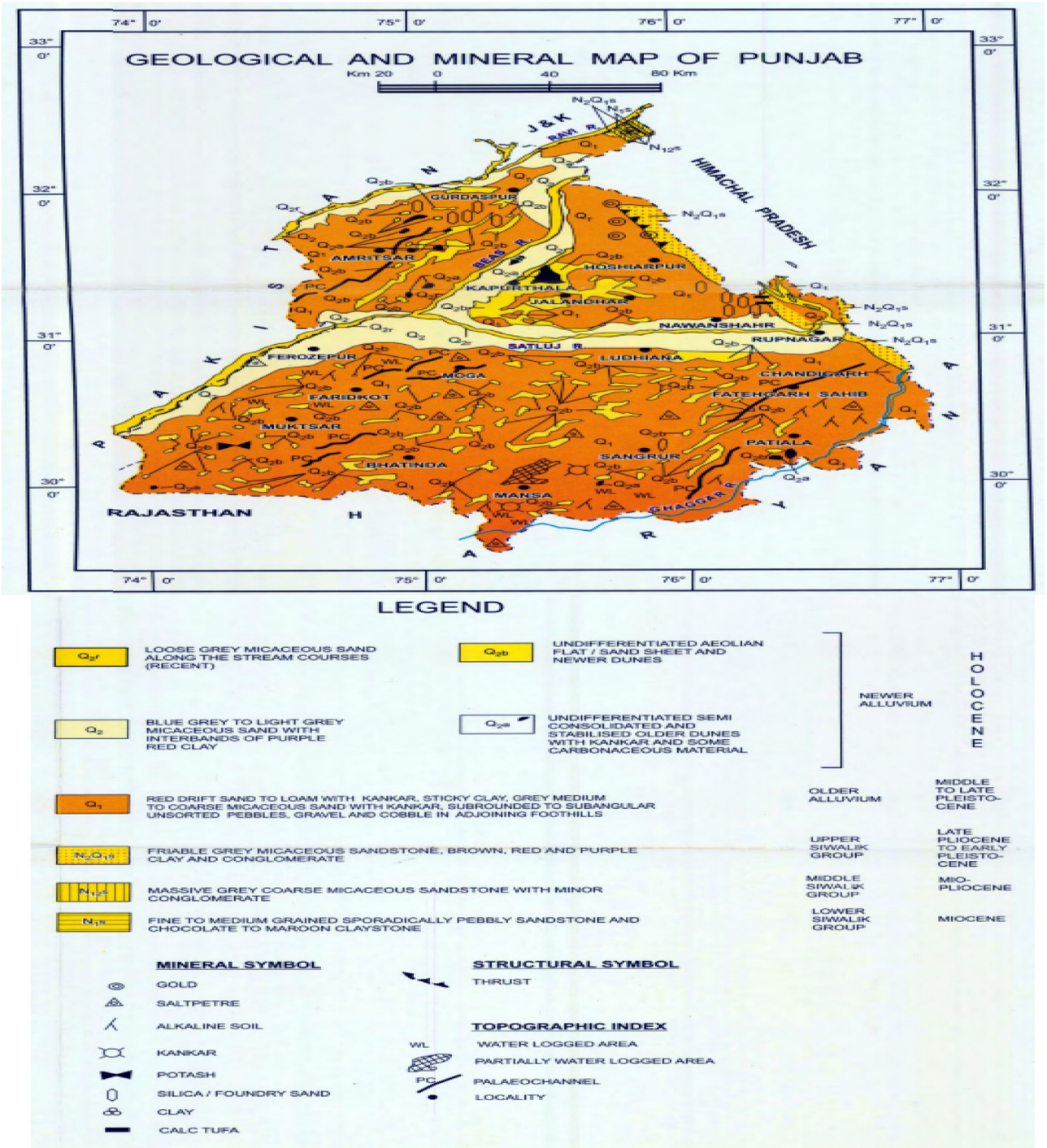

Fig. 2 – The geological and minerals map of Punjab.

where C is the concentration of the contaminant in the environmental media (mg kg^{-1} or mg l^{-1}), IR is the ingestion rate per unit time (mg day^{-1} or $lday^{-1}$), ED is the exposure duration (y), EF is the exposure frequency (days y^{-1}), BW is the body weight of the receptor (kg), and AT is the Averaging time (years), equal to the life expectancy, and 365 is the conversion factor from year to days. Toxic risks refer to the non-carcinogenic harms incurred due to the exposures. The extent of the harm is indicated in terms of a hazard quotient (HQ):

$$HD = \frac{ADD}{RfD} \quad (6)$$

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Table 1 – District wise summary of analysis of uranium in ground water samples of SW-Punjab.

District	Area covered (Sq. Km)	No of samples analyzed	Mean uranium concentration (μgl^{-1})	Maximum concentration (μgl^{-1})	Minimum concentration (μgl^{-1})
Bathinda	3344	185	80.7	571.7	0.5
Mansa	2192	117	80.3	579.0	1.3
Faridkot	1468	66	68.4	476.0	2.4
Firozepur	5850	130	64.6	467.5	2.8
Entire region	**11854**	**498**	**73.5**	**579.0**	**0.5**

Table 2 – Statistical distribution of water samples for uranium concentration above WHO and AERB permissible limits.

	No of samples	Range	Mean	Median	Above WHO safe limit	Above AERB safe limit
Bathinda	185	0.5–571.7	81.1	56.4	131 (71%)	87 (47%)
Mansa	117	1.3–579.0	80.3	50.5	75 (64%)	48 (41%)
Faridkot	66	2.4–476.0	68.4	49.6	45 (68%)	27 (41%)
Ferozepur	130	2.7–467.5	64.6	45.0	87 (67%)	54 (42%)

where, R_fD is the reference dose. The reference dose is the daily dosage that enables the exposed individual to sustain this level of exposure over a prolonged time period without experiencing any harmful effect.

3.2. *Elemental analysis in water using atomic absorption spectrophotometry*

Few of collected water samples have also been analysed for trace elemental analysis using atomic absorption spectrophotometry.

3.2.1. *Analytic procedure*

100 ml aliquot of filtered water sample was taken in duplicate for wet digestion (HNO_3) on hot plates to destroy organic material in the sample. 25 ml of the prepared sample was injected in the nebulizer-spray chamber-burner system of the Atomic Absorption Spectrophotometer. Air-acetylene compressed gas has been used as oxidant and fuel. Atomic Absorption Spectrophotometer was standardized with standard element concentrations in prior.

4. Results and discussion

Results of uranium concentration in 498 drinking water samples from the four districts namely Bathinda, Mansa, Faridkot and Ferozepur of the SW-Punjab, analyzed using the Laser Flourimetric technique are tabulated in Table 1. Overall, uranium concentration in the drinking water samples of the study region have been found to be varying between 0.5–579 μgl^{-1} with an average of 73.5 μgl^{-1}. By comparing the observed data with the permissible limits of 30 μgl^{-1} and 60 μgl^{-1} prescribed by the WHO and AERB respectively, a statistical distribution has also been reported in Table 2. It has been observed that around 68% of the 498 collected water samples were found to exceed the permissible limit of the WHO and 43% samples exceed the limit prescribed by the AERB. Summary of the uranium concentration, corresponding radiological (excess cancer risk) and chemical risk (HQ) for all the collected drinking water samples from this region has been reported in Table 3.

The heavy metal concentration variations observed in drinking water samples of the study region is reported in Table 4. It can be inferred from the table that the concentration of As, Pb, Ni, Zn and Cr have been observed to be above their respective safe limits given by WHO in considerable number of samples. In the present analysis, concentration of As in drinking water samples has been found to be varying from 1.0–59.6 μgl^{-1} with an average of 32.0 μgl^{-1}. Although it is exceeding the safe limit 10 μgl^{-1} only in six samples, but the average concentration of the As is below the recommended safe limit. The concentration of Pb in all the collected samples ranged between MDL (Minimum Detection Limit) – 444, with an average of 46.2 μgl^{-1}. Similary, Ni, Zn and Cr were among heavy metals which were observed above the permissible limits and their concentrations vary from MDL-308, MDL-2365 and MDL-228 μgl^{-1} respectively; while elements like Cu, Mn, Co, Fe, Na and K were not observed above safe limits in the

Table 3 – Summary of uranium concentration, corresponding radiological (excess cancer risk) and chemical risk (HQ) for all drinking water samples of SW-Punjab.

Parameter	Uranium concentration (μgl^{-1})	Excess cancer risk	LADD ($\mu g\ kg^{-1}\ day^{-1}$)	HQ
Mean	73.5	2.06×10^{-4}	5.54	1.22
Range	0.5 ± 0.2–579 ± 3.8	1.40×10^{-6}–1.62×10^{-3}	0.04–43.66	0.01–9.64

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Table 4 – Variation of heavy metal concentration in the water samples of SW-Punjab.

Element (No of samples analyzed)	As (μg/L) (60)	Pb (μg/L) (110)	Cu (μg/L) (170)	Mn (μg/L) (110)	Co (μg/L) (110)	Ni (μg/L) (170)	Fe (μg/L) (110)	Zn (μg/L) (170)	Cr (μg/L) (170)	Na (μg/L) (110)	K (μg/L) (110)
Min	1.00	<0.01	<0.05	<0.5	<0.2	<0.2	10	<0.05	<0.5	<0.01	<0.05
Max	59.6	444	15	508	481	308	3424	2365	228	855	300
Mean	32.0	46.2	145.0	21.5	6.8	34.6	830.7	833.8	28.3	503.68	69.2
Permissible Limit (WHO)	10	10	2000	Guideline not Estd	Guideline not Estd	70	Guideline not Estd	10	50	Guideline not Estd	Guideline not Estd
Sample above WHO limit	54	142	nil			17		163	24		

study region. It has also been observed that Zn with average value of 833.8 μgl^{-1} has been found well above the permissible limit 10 μgl^{-1} given by the WHO. This corroborate the pervious study carried out by Kumar, Singh, and Mahajan (2006), where it has been reported to be observed in the range 3–1044 μgl^{-1}.

4.1. *Water quality parameters*

Physico-chemical parameters (pH, ORP, Conductivity, Salinity and TDS) for the water samples are reported in the Table 5. Physico-chemical parameters of water 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 concentrations of the drinking water samples for the present study, varied from 55–2020 mgl^{-1} with an average of 599 mgl^{-1}. Since the safe limit of TDS in the drinking water samples is considered to be 600 mgl^{-1} (WHO, 2011), thus although the average of the observed TDS is just below the desirable limit, but the highest TDS has been observed in quite few samples with high content of uranium. The pH in the water samples has been observed to be in range of 6.63–8.21 with average 7.47, which are within the safe limits of 6.5–8.5 recommended by the Bureau of Indian Standards (BIS limits IS10500, 2012). The ranges of other water quality parameters i.e. ORP, conductivity and salinity for the collected water sample in the study region have been observed as −108–(−13) mV, 0.16–4.05 mS/cm and 0.00–2.80 ppt respectively.

Overall, the quite higher concentrations observed in ground water samples of the SW-Punjab region may be due to local natural geology, industrial activities in the region or use of phosphate fertilizers in the region in huge quantity for agricultural purpose as the region is well known for it or due to any other human activities (Tripathi et al., 2013). The present case may be quite similar to the study observed in Central Valley, California, USA (Jurgens, Fram, Belitz, Burow, Landon, 2009). As it has been indicated in the geology of this region stated above, since the granites and acid volcanic of Malani age form the basement in the study region, so the interaction of ground water with soils formed from the weathering of granites. Thus, the interaction of ground water with the soils formed from weathering of Malani granites (which are outcropping in Tosham ring complex) and the basement rocks (Delhi quartzite) encountered in the study area might have been one of the cause of high uranium observed in this region. Secondly, the ground water in this region is found to be rich in biocarbonates, nitrate and chlorides anions (Kumar et al., 2011) and soil is calcareous one. As the region is well known for its agricultural activities, plant root respiration and microbial oxidation of organic matter in soil produce carbon dioxide, resulting in CO_2 partial pressure in the soil zone that are greater than the atmospheric pressure. Water percolating through the soil equilibrates with the soil atmosphere by dissolving CO_2 (g) to form carbonic acid. The carbonic acid reacts with the calcium carbonates (calcareous soil) to form bicarbonate which is a well-known efficient leaching agent for uranium from soils and sediments. Formation of bicarbonate while water is percolating through soil enhances its leaching efficiency. This may be one of the mechanisms which explain the high

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Table 5 – Physico-chemical parameters for the drinking water samples of SW-Punjab.

	pH	ORP (mV)	Conductivity (mS/cm)	TDS (mg/ml)	Salinity (ppt)
Average	7.47	−64.90	1.207	599	0.66
Range	6.63–8.21	−108–(−13)	0.16–4.05	55–2020	0.00–2.80
BIS limits (IS10500,2012)	**6.5–8.5**	–	–	**500**	–

Table 6 – Uranium concentration in drinking water samples in different states of India.

Sl. No.	States/cities of India	Basic source	Uranium concentration range ($\mu g.l^{-1}$)	References
1	Himachal Pradesh, Bilaspur	Spring water	0.1–4.6	Singh, Singh, Sandhu, and Singh (1999)
2	Himachal Pradesh, Kulu	Ground water	0.3–2.5	Singh, Malhotra, Kumar, Singh, and Singh (2001)
3	Himalayas (Kumaun, Siwallik)	–	1.1–35.8	Ramola, Singh, and Virk (1988).
4	Hyderabad	Ground water	0.6–82.0	Balbudhe et al. (2011)
5	Punjab, Amritsar	Ground water	3.2–45.6	Singh, Rani, Mahajan, and Singh Walia (2003)
6	Punjab, Bhatinda	Ground water	11.7–113.7	Singh, Singh, and Singh (1995)
7	Kolar, Karnataka	Ground water	0.3–1442.9	Babu et al. (2008)
8	Himachal Pradesh, Chamba	–	0.3–6.8	Singh, Singh, Singh, and Virk (1984)
9	Punjab, Malwa	Ground water	5.4–43.4	Mehra, Singh, and Singh (2007)
10	Jhansi	Ground water	0.9–6.4	Singh, Rana, Azam, Naqvi, and Srivastava (1996)
11	Kanpur	Ground water	3.3–9.1	
12	Allahabad	River stream	0.9–2.3	
13	West Bengal	Tap water	1.3–13.2	Bansal, Tyagi, and Prasad (1988)
14	Uttar Pradesh	Ground water	1.4–19.2	
15	Ghaziabad	Ground water	4.2–11.4	Singh, Rana, Azam, Naqvi, and Srivastava (1996)
16	Rajasthan	Tap water	0.9–3.0	Bansal et al. (1988)
17	Delhi	Hand pump	2.2–8.8	
18	Maharashtra	River water	0.03–7.8	Rao and Shah (1976)
	Present Study	**Ground Water**	**0.5–579**	

uranium in ground water in the regions but other possibilities can't be ruled out. Thus, since the anthropogenic activities and urbanization is also responsible for increase in the TDS, salinity of the region and which certainly can raise the concentration of bicarbonate, nitrogen etc., so might be another cause of high uranium observed in the study region. Even, since this study region falls in the major cotton belt of Punjab (MALWA region of Punjab) and as there has been a wide spread use of pesticides/fertilizers extensively by the farmers from the last many decades, like diammonium phosphate, cyhalothrein & even fertilizers such as urea, super phosphate and NPK, which might have also contributed to certain extent towards the high concentration of uranium observed in ground water of this region, as well as the heavy toxic elements present in the region. For comparison purposes, the uranium concentration variations in the drinking water samples in different states all over Indian and in the world over are depicted in Tables 6 and 7 respectively.

Table 7 – Reported uranium concentrations in drinking water worldwide.

Sl. No.	Country	Uranium concentration range ($\mu g.l^{-1}$)	References
1	Ontario, Canada	0.05–4.2	OMEE (1996)
2	New York, USA	0.03–0.1	Fisenne and Welford (1986)
3	Argentina	0.04–11	Bomben, Equillor, and Oliveira (1996)
4	Jordan	0.04–1400	Gedeon, Smith, Amro, and Jawadeh (1994)
5	Kuwait	0.02–2.5	Bou-Rabee (1995).
6	United States	1.1652	Cothern and Lappenbusch (1983)
7	South Greenland	0.5–1.0	Brown, Steenfelt, and Kunzzenorf (1983)
8	Turkey	0.2–17.6	Kumru (1995)
9	Germany	2.2–24.0	UNSCEAR (2000)
10	Finland, Europe	0.02–6000	
11	China, Asia	0.004–28	
12	Iran	1.0–10.9	Alirezazadeh and Garshasbi (2003)
13	Brazil	0.5–2.3	Geraldo, Cesar, Mafra, and Tanaka (1979)
14	Norway	<0.02–170	Banks, Royset, Strand, and Skarphagen (1995)
15	Sweden	<0.2–470	Selden et al. (2009)

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5. Conclusions

- A large variation (0.5–579 μgl^{-1}) in the uranium concentrations has been observed in the analysed drinking water samples of the SW-Punjab.
- In this region, uranium concentration in 498 drinking water samples has been found to vary between 0.5–579 μgl^{-1} with an average of 73.5 μgl^{-1}. Data analysis reveals that, 68% of the collected samples have uranium concentration higher than safe limit of 30 μgl^{-1} (WHO, 2011) while 43% samples exceed the threshold of 60 μgl^{-1} recommended by AERB, DAE, India, 2004.
- Higher concentrations observed in ground water samples of SW-Punjab might be due to leaching of uranium from adjoining/basement granite rich rock formations. The anthropogenic activities, urbanization and wide spread use of pesticides/fertilizers which is responsible for increase in the TDS/salinity of the region might be another cause. But, overall it seems that the plausible source of high uranium observed in this region may be of geogenic in nature.
- Although some elements viz. As, Pb, Ni, Zn and Cr have also been observed to be above their respective safe limits in the considerable number of water samples, but none of the elements including even uranium have been observed in abnormally high amounts in the collected soil samples from this region.

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