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Health risk assessments due to uranium contamination of drinking water in Bathinda region, Punjab state, India

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ABSTRACT **The mass concentration of uranium in water samples collected from the Bathinda district of Punjab state, India, was estimated using the laser fluorimetric technique. The study region has shown a pronounced number of cancer cases in the recent period. The study aims to calculate the human radiological risk and chemical toxicity associated with uranium consumption through drinking water. The mass concentration of uranium was found to vary from 0.48 to 571.7 µg/l with a mean value of 84.70 µg/l. The radiological risk due to consumption of uranium through contaminated drinking water was observed to be in the range of 1.34×10^{-6} to 1.60×10^{-3} with a mean value of 2.37×10^{-4}. The chemical toxicity was found to vary from 0.04–43.11 $\mu g.kg^{-1}.day^{-1}$ with a mean value of 6.38 $\mu g.kg^{-1}.day^{-1}$.**

1. Introduction

Uranium, a long-lived natural radionuclide present in rocks, soil, natural materials, water, air and food, plays a vital role in delivering the dose to the public through terrestrial exposure, inhalation and ingestion. Uranium enters the human body mainly through water, air and food. So, accurate knowledge of the concentration of uranium in water helps in estimating the dose due to uranium. Uranium exists in oxidation states of +4 (UO_2 and U^{4+}) and +6 (UO_3 and UO_2^{2+}) in its compounds. Uranium bonds with oxygen to form uranyl ion or uranium dioxide, which is soluble in groundwater under aerobic conditions. Uranium dissolves readily in oxygen-rich water, which accounts for its presence in surface water, groundwater and seawater. The hexavalent state of uranium commonly associated with UO_2^{2+} is more significant in water, whereas this state of uranium is insoluble in almost all tetravalent compounds (Sahoo *et al.,* 2009).

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It has been established that concentrations of uranium in drinking water above permissible limits may produce harmful biological effects in humans (WHO, 2008). The health effects of uranium can be divided into carcinogenic and non-carcinogenic effects (Health Canada, 1999; WHO, 1998) and these classifications are based on the radiological risk (carcinogenic) due to radiation of uranium isotopes and the chemical toxicity risk (non-carcinogenic) due to uranium as a heavy metal. The toxic effects of uranium compounds have been extensively studied in the kidney (Kurttio *et al.*, 2002). It was found that uranium is toxic to kidneys. However, in 2006, Kurttio *et al.* found that even at higher exposures uranium was not cytotoxic (Kurttio *et al.*, 2006). The toxic effects of uranium on the bones of laboratory animals were studied, showing that uranium deposition is part of the metabolic process, and showing the uranium concentration in the bones of populations exposed to significant uranium intake (Larivière *et al.*, 2007). It was found that uranium in the range of 0.004 to 9 μg/(kg body weight) may produce interference with kidney functions (Zamora *et al.*, 1998). A study on a Finnish population exposed to well water containing a median uranium concentration of 28 μg/l examined individuals for signs of adverse renal effects (Kurttio *et al.*, 2002). The development of toxicity due to uranium has been studied in mice (Dominigo *et al.*, 1989) and a reduction in maternal weight gain, reduction in daily feed intake and increase in liver weight have been observed.

In the Bathinda region, the authors have already measured high concentrations of uranium in some water samples (Singh *et al.,* 1994). Therefore, it is necessary to study in detail the measurement of the uranium contents in the water and to determine the health risk to the human body. In the present paper, we measured and report the mass concentration of uranium in water samples collected from the different sources of water bodies in the Bathinda region of Punjab state, India. 159 drinking water samples were collected in this region from different locations, different sources and different depths to evaluate the radiological risk and chemical toxicity due to uranium in drinking water.

2. Description of study region

Bathinda district is situated in the south-western region of Punjab State in Northern India. Bathinda district extends between 29°33´ and 30°36´ north and from 74°38´ to 75°46´ east and lies at an average elevation of 300 m above sea level. The study region is darkened on the map of Punjab state, India, as shown in Figure 1. The study region forms a part of the Indo-Gangetic Alluvial Plains. The temperature and relative humidity in the area lie in the range of 34–49 °C and 18–48% (during summer) and 2–10 °C and 60–73% (during winter), respectively. The soil of the study region is loose and sandy. The soil consists of different layers of clay, sticky clay and fine- to coarse-grained grey micaceous sandstone. The top layer of soil is
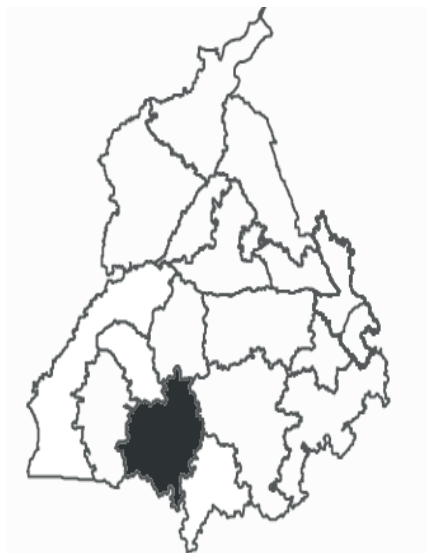
Figure 1 – *Dark area shows the study region on the map of Punjab State, India.*

dark reddish silty soil rich in $CaCO_3$. The hydrogeology of this region is characterised by unconfined and confined aquifers down to 50 m depth (Gill *et al.,* 2005). The land is predominantly used for agricultural purposes. However, Bathinda district has two thermal power plants, a fertiliser factory and an oil refinery.

3. Materials and methods

3.1. Sample preservation and analysis

A laser fluorimeter manufactured by Quantalase Enterprises Pvt. Ltd., Indore, India, was used to measure the concentration of uranium up to ppb level (µg/l). The laser fluorimeter consists of a sealed nitrogen laser tube as an excitation source with a wavelength of 337.1 nm, a sample compartment and a photomultiplier tube (PMT). The detector is placed at a right angle to the radiation source to avoid the background signal. The laser source emits an intense pulsed ultraviolet radiation of 337.1 nm carrying energy of 20 µJ of width 7 ns with a repetition time of 10 pulses per second (Sahoo *et al.,* 2009). Fluorescence interference due to organic matter is removed by using a proper electronic delay, optical filter and gating technology (Veselsky *et al.,* 1988). The fluorescence signals from the PMT are integrated for 4 seconds and then displayed.

Samples were collected from the different sources of drinking water in acid-clean and tightly closed polyethylene 500-ml bottles for uranium analysis. In the study region, the majority of the population uses the groundwater for drinking purposes. Only a very small proportion of the population uses canal and RO-treated water for drinking purposes. In this study, 147 samples of groundwater, 6 samples of canal water and 6 samples of RO-treated water were collected and analysed. RO-treated water was collected from the government-installed ROs in 6 villages of the study region. Each sample was brought to pH range 6.5–7.5 before analysis to overcome the quenching effect due to the presence of high acidic concentration (Veselsky *et al.,* 1988). The presence of a high quantity of other interfering materials (such as iron, manganese, etc.) can cause the quenching effect. So, the standard addition method, explained below, is used for the uranium analyses by laser fluorimeter to avoid the matrix effect.

3.2. Analytical procedure

5 ml of a water sample was placed in the dry and clean quartz cell along with 0.5 ml of 5% sodium pyrophosphate of pH value 7. The instrument was calibrated with standard uranium solution of known concentration. Both micropipettes and an analytical balance were used for the measurements to avoid errors. The concentration of uranium in samples was calculated by:

$$U \text{ (µg/l)} = \frac{D1}{D1 - D2} \frac{V1}{V2} C \tag{1}$$

$D1$ is fluorescence due to the sample only, $D2$ is fluorescence due to the sample and spiked uranium standard, $V1$ is the volume of U standard added, $V2$ is the volume of sample taken and C is the concentration of the U standard solution (Sahoo *et al.,* 2009).

3.3. Risk assessment

In the present investigation, two types of risks associated with uranium were evaluated. One is the radiological risk due to uranium as a radioactive element, and the second is the chemical toxicity due to uranium as a heavy metal.

3.3.1. Radiological risk assessment

The radiological risk was calculated by the USEPA method (USEPA, 2000). The excess cancer risk associated with intake of uranium nuclide was estimated as follows:

$$\text{activity concentration of uranium (Bq/l)} =$$
$$\text{mass concentration of uranium (µg/l)} \times \text{conversion factor (0.025 Bq/µg)}, \tag{2}$$

$$\text{risk factor (per Bq/l)} =$$
$$\text{cancer risk coefficient } (1.19 \times 10^{-9} \text{ Bq}^{-1}) \times \text{water intake}, \qquad (3)$$

$$\text{excess cancer risk} =$$
$$\text{activity concentration of uranium (Bq/l)} \times \text{risk factor (per Bq/l)}. \qquad (4)$$

The average Indian life expectancy for males and females in India is 63.7 years, *i.e.* 23 250 days (HDR, 2009), and daily consumption of water was set as 4.05 l/day (Jain *et al.,* 1994). Thus, the total lifetime water intake turns out to be 94 162.5 liters. The reported value of 1.19×10^{-9} Bq^{-1} of the cancer risk coefficient of uranium was used as given by USEPA in equation (3) (EPA, 1999; UNSCEAR, 2000).

3.3.2. Chemical toxicity risk

The chemical toxicity risk in the drinking water samples due to the uranium concentration was estimated in terms of the *LADD* (lifetime average daily dose, µg.kg^{-1}.day^{-1}) using the following formula (Ye-shin *et al.,* 2004):

$$LADD \text{ (µg.kg}^{-1}.\text{day}^{-1}) = \frac{EPC \times IR \times EF \times ED}{AT \times BW} \qquad (5)$$

where *LADD* = lifetime average daily dose (µg.kg^{-1}.day^{-1}), *EPC* = exposure point concentration (µg/l), *IR* = water ingestion rate (l/day), *EF* = exposure frequency (days/year), *ED* = total exposure duration (years), *AT* = average time (days) and *BW* = body weight.

The water ingestion rate was set as 4.05 l/day due to the highly humid conditions (Jain *et al.,* 1995). The exposure frequency is 350 days. 63.7 years is the total exposure duration, *i.e.* the average Indian life expectancy for both males and females (HDR, 2009). 23 250 days is the average time calculated (63.7 × 365 days) and 51.5 ± 8.5 kg (Dang *et al.*, 1996) was the average body weight taken of an Indian man. The hazard quotient (*HQ*) is given by:

$$HQ = \frac{LADD}{RFD}. \qquad (6)$$

Here, *RFD* is the reference dose calculated on the basis of the AERB permissible limits (60 µg/l) (AERB, DAE, India, 2004) and turned out to be 4.53 µg.kg^{-1}.day^{-1}.

Animal studies have identified the kidney as the most sensitive organ of uranium toxicity (Luessenhop *et al.*, 1958). The Royal Society has also concluded that the acute exposures that lead to concentrations of uranium of 1 µg/g in the kidney have been associated with minor kidney dysfunction and the chronic level

TABLE I
**Safe limits for the uranium concentration in drinking water recommended by different
health organisations.**

Health agency	uranium concentration[1]	excess cancer risk	reference dose[2]	reference
WHO	30	8.4×10^{-5}	2.26	WHO, 2011
Health Canada	20	5.6×10^{-5}	1.51	Ye-shin *et al.,* 2004
USEPA	30	8.4×10^{-5}	2.26	USEPA, 2003
AERB	60	1.68×10^{-4}	4.53	AERB, DAE, India, 2004

[1]Expressed as µg/l; [2]Expressed as µg.kg^{-1}.day^{-1}; *LADD* = lifetime average daily dose; excess cancer risk and reference dose measured according to the above risk assessment methodologies given in equations (4) and (5), respectively.

that leads to minor kidney dysfunction is not well established but likely to be 0.3 µg/g in the kidney (Royal Society, 2002). The nephrotoxic effects of uranium are due to its chemical properties. The threshold concentration of 3 µg/g in the kidney for uranium was proposed (Spoor and Hursh, 1973).

4. Results and discussion

Different health organisations have recommended different safe limits for the uranium concentration in drinking water, as shown in Table I. The excess cancer risk and reference dose are measured according to these safe limits. The average daily water intake, body weight and life expectancy were taken according to Indian standards, as discussed above in the risk assessment methodologies. However, in the present study, the results were compared with the AERB safe limits.

In the present investigations, 159 samples of drinking water of the Bathinda district were analysed to measure chemical and radiological toxicity. Various statistical parameters, *i.e.* the mean, median, maximum value, minimum value, 5th percentile, 95th percentile, 25th percentile and 75th percentile of various quantities, *i.e.* the mass concentration of uranium, radiological risk, *LADD* and hazard quotient were evaluated and are reported in Table II. The mass concentration of uranium was found to vary in the range of 0.48 to 571.7 µg/l, with a mean concentration of 84.70 µg/l. Out of the analysis of 159 drinking water samples, only 28 (17.6%) were found to be below 15 µg/l; 43 (26%) were found to be below 30 µg/l, the permissible limit recommended by USEPA (2003) and WHO (2011); and 81 (50.9%) samples were found to be below 60 µg/l, the permissible limit recommended by the AERB (AERB, DAE, India, 2004). The pi chart for the number of samples in different ranges of uranium concentration is shown in Figure 2. The excess cancer risk associated with uranium concentration was found

TABLE II
Various statistical parameters calculated from Bathinda district drinking water samples.

parameter	uranium concentration[1]	excess cancer risk	$LADD^2$	HQ
Mean	84.70	2.37×10^{-4}	6.38	1.41
Median	57.00	1.60×10^{-4}	4.30	0.95
Range	0.48–571.7	1.34×10^{-6}–1.60×10^{-3}	0.04–43.11	0.01–9.52
Q_{25}	25.75	7.21×10^{-5}	1.94	0.43
Q_{75}	114.2	3.20×10^{-4}	8.61	1.90
P_5	3.64	1.02×10^{-5}	0.27	0.06
P_{95}	223.5	6.26×10^{-4}	16.86	3.72

[1]Expressed as µg/l; [2]Expressed as µg.kg^{-1}.day^{-1}; Q_{25}= 1st quartile, Q_{75} = 3rd quartile, P_5 = 5th percentile, P_{95} = 95th percentile, $LADD$ = lifetime average daily dose, HQ = hazard quotient.

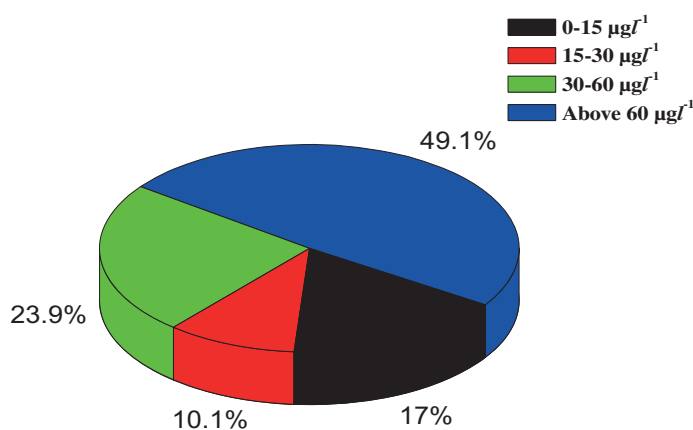**Figure 2** – *Pi chart representation of the number of samples collected in the above ranges of uranium concentration.*

to be in the range 1.34×10^{-6} to 1.60×10^{-3} with a mean value of 2.37×10^{-4}, which is greater than the AERB permissible limit of 1.68×10^{-4} (AERB, DAE, India, 2004). The estimated lifetime average daily dose (*LADD*) was observed to be between 0.04– 43.11 µg.kg^{-1}.day^{-1} with a mean value of 6.38 µg.kg^{-1}.day^{-1}. It was observed that the concentration of uranium in reverse osmosis-(RO) treated and canal water samples was within permissible limits, whereas the groundwater showed quite a high concentration of uranium.

In the same study region, Kumar *et al.* have carried out similar investigations (Kumar *et al.*, 2011). In their study, they investigated 235 water samples from four

TABLE III
RO-treated water samples (number of samples = 6).

parameter	uranium concentration[1]	excess cancer risk	LADD[2]	HQ
mean	3.21	8.98×10^{-6}	0.24	0.05
median	2.66	7.44×10^{-6}	0.20	0.04
range	0.48–8.30	1.34×10^{-6}–5.78×10^{-6}	0.04–0.63	0.01–0.14
Q_{25}	2.06	5.78×10^{-6}	0.16	0.03
Q_{75}	3.13	8.76×10^{-6}	0.24	0.05
P_5	0.86	2.41×10^{-6}	0.06	0.01
P_{95}	7.03	1.97×10^{-5}	0.53	0.12

[1]Expressed as µg/l; [2]expressed as µg.kg^{-1}.day^{-1}; Q_{25} = 1st quartile, Q_{75} = 3rd quartile, P_5 = 5th percentile, P_{95} = 95th percentile, *LADD* = lifetime average daily dose, *HQ* = hazard quotient.

districts (Bathinda, Mansa, Ferozepur and Faridkot) of Punjab. Of these 235 samples, 80 samples were from the present study region. The uranium concentration in these 80 water samples ranged from 2-644 ppb with a mean of 80.83 ppb. In the present work, in the 159 water samples analysed, the average of 84.70 ppb of uranium concentration did not differ significantly. It confirms that the uranium content in the subsurface water of the study region is above the permissible limits.

In RO-treated water samples, the concentration of uranium was found to vary from 0.48–8.30 µg/l with a mean value of 3.21 µg/l, and is reported in Table III. All samples have a concentration of uranium within the permissible limits recommended by the AERB. A comparison of RO-treated water samples with groundwater samples shows that the RO system was effective in lowering the uranium concentration of the water by up to 90%. This was also supported by an earlier published report (WHO, 2011). In canal water samples, the concentration of uranium was found to vary from 3.00–8.20 µg/l with a mean value of 5.18 µg/l, and is reported in Table IV. It was further observed that the concentration of uranium in canal water was also within the permissible limits. The maximum value of the radiological risk was found to be 5.78×10^{-6} and 2.30×10^{-5} in the case of RO-treated water samples and canal water samples, respectively, which is less than the AERB permissible limit (1.68×10^{-4}) (AERB, DAE, India, 2004). The RO-treated and canal water samples were found to be safe for drinking from a radiological and chemical toxicity risk point of view.

The statistical parameters for mass concentration of uranium, radiological risk and chemical toxicity of groundwater samples are presented in Table V. It was observed that the mean concentration of uranium and mean radiological risk were 91.91 µg/l and 2.57×10^{-4}, respectively, which is higher (53.2%) than the AERB

TABLE IV
Canal water samples (number of samples = 6).

parameter	uranium concentration[1]	excess cancer risk	LADD[2]	HQ
mean	5.18	1.45×10^{-5}	0.39	0.09
median	5.08	1.42×10^{-5}	0.38	0.08
range	3.00–8.20	8.40×10^{-6}–2.30×10^{-5}	0.23–0.62	0.05–0.14
Q_{25}	3.89	1.09×10^{-5}	0.29	0.06
Q_{75}	5.96	1.67×10^{-5}	0.45	0.10
P_5	3.15	8.82×10^{-6}	0.24	0.05
P_{95}	7.69	2.15×10^{-5}	0.58	0.13

[1]Expressed as µg.l^{-1}; [2]expressed as µg.kg^{-1}.day^{-1}; Q_{25} = 1st quartile, Q_{75} = 3rd quartile, P_5 = 5th percentile, P_{95} = 95th percentile, *LADD* = lifetime average daily dose, *HQ* = hazard quotient.

TABLE V
Groundwater samples (number of samples = 147).

Parameter	Uranium Concentration[1]	Excess Cancer Risk	LADD[2]	HQ
mean	91.91	2.57×10^{-4}	6.93	1.53
median	65.20	1.83×10^{-4}	4.91	1.09
range	3.10–571.7	8.68×10^{-6}–1.60×10^{-3}	0.23–43.11	0.05–9.52
Q_{25}	39.4	1.10×10^{-4}	2.97	0.65
Q_{75}	124.5	3.49×10^{-4}	9.38	2.07
P_5	10.77	3.02×10^{-5}	0.81	0.18
P_{95}	229.5	6.43×10^{-4}	17.31	3.82

[1]Expressed as µg/l; [2]expressed as µg.kg^{-1}.day^{-1}; Q_{25} = 1st quartile, Q_{75} = 3rd quartile, P_5 = 5th percentile, P_{95} = 95th percentile, *LADD* = lifetime average daily dose, *HQ* = hazard quotient.

permissible limit of 1.68×10^{-4} (AERB, DAE, India, 2004). It means that 2–3 persons out of 10 000 may be prone to cancer. The 95th percentile was considered to be the worst case and the worst case of radiological risk was calculated as 6.43×10^{-4}, which means that 6–7 persons out of 10 000 may be cancer patients. The mean value of the hazard quotient was calculated as 1.53 and the hazard quotient value for the worst case was 3.82.

Groundwater showed higher levels of uranium than canal and RO-treated water samples. The box whisker plot of uranium concentration in water samples at different depth ranges, canal water samples and RO-treated water samples is shown in Figure 3. The depth interval of 200–300 feet was found to have the highest mean concentration of 121 µg/l in the groundwater samples. No correlation of uranium concentration was found with depth. Therefore, a possible reason for
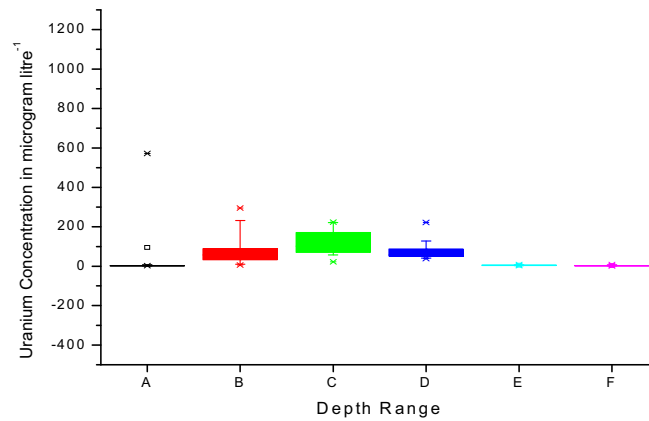
Figure 3 – *Box whisker plot of uranium concentration in groundwater with different depth ranges, canal water and RO-treated water samples. A in the graph stands for the groundwater sample depth in the range 0–100 feet, B for 100–200 feet, C for 200–300 feet, D above 300 feet. E and F represent canal water and RO-treated samples, respectively.*

the high concentration of uranium in groundwater samples may be due to leaching of uranium through the soil to groundwater by the heavy use of phosphate fertilisers in the studied area. It has been established that the use of phosphate fertilisers may enrich the soil with uranium (Schnug and Haneklaus, 2008). The soil of the study region is rich in $CaCO_3$ (Gill *et al.,* 2005) and the leaching of uranium in the carbonate medium has already been confirmed by some authors (Mason *et al.*, 1997).

5. Conclusion

The present results reveal that negligible radiological risk and chemical toxicity were found among the drinking water samples of the study region at the 5th percentile, but the water samples of the study region were found to have a high value of the excess cancer risk, 6.26×10^{-4}, which means about 6–7 persons out of 10 000 people may be prone to cancer, and a high value of the hazard quotient; 3.72. This means that the uranium contamination in the drinking water samples was found to be 3.72 times the AERB safe limit. The mean hazard quotient was found to be 1.41, which is also greater than 1. The drinking water samples collected had mean radiological risk and chemical toxicity greater than the permissible limits of the AERB. It was concluded that the RO-treated water samples and canal water samples have a uranium concentration within the permissible limits recommended by different organisations and are considered to be safe for drinking, but the groundwater samples of the study region have a

uranium concentration higher than the safe limits recommended by the different organisations in Table I and are considered to be unfit for drinking without any treatment.

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