

Health-related analysis of uranium in Fazilka district, Punjab, India

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Laser fluorimetry technique has been used to estimate uranium concentration in groundwater samples collected from 20 villages of Fazilka district, Punjab, India. The uranium concentration was found to vary from 4.32 to 83.99 $\mu\text{g l}^{-1}$ at different locations with mean concentration of 26.51 $\mu\text{g l}^{-1}$. Also, 24% of the drinking water samples exceeded the safe limits set by WHO, while 9% was above the limit set by AERB. Certain health risk factors like annual effective dose, excess cancer risk and lifetime average daily dose were also evaluated. The study also included uranium estimation in soil samples collected from the same villages using wavelength dispersive X-ray fluorescence technique. All the values were found to be well within the safe limits. Topography of the region seems to be the most likely reason for higher uranium concentration at some locations.

Keywords: Annual effective dose, laser fluorimetry, safe limits, uranium.

URANIUM is a naturally existing, long-lived radioactive element which is present in air, water and soil with wide variation in concentration. It is a material of very high density (18.9 g/cm^3). In fact, it is 65% denser than lead¹. Due to its radioactivity, uranium provides some radiation exposure to the general public, which is inevitable². It diffuses readily in oxygen enriched water which is responsible for its presence in surface water, groundwater and the sea. Weathering of igneous rocks which formed the earth's original crust has accounted for much of the dissolved uranium. Combustion of coals and other fuels, release from the uranium industry and use of phosphate fertilizers have also contributed to uranium in the aquatic environment³. Once it enters the water, uranium does not enter into the atmosphere.

Water intake is an integral part of human diet. This depends upon various factors like sex, age, body weight, metabolic activity, etc. Uranium present in groundwater can be taken up by plants and transferred to the food chain. The element is considered unessential for all organisms as it does not contribute to any metabolic function. However, it can cause adverse health effects in humans due to its radioactive and chemical properties.

Uranium nuclides emit alpha particles of high ionization power and hence may be toxic if inhaled or ingested in high amount. Drinking water contributes about 85% of the ingested uranium, while food about only 15%. Uranium can be severely damaging to the kidneys if taken in large proportions⁴. Thus estimation of uranium content of any region becomes significant for health risk assessment. The US Environmental Protection Agency (EPA) has listed uranium in Group A category of human carcinogens. EPA has also advocated that only zero tolerance is the safe admissible limit for cancerous risk from uranium. The 'minimal risk' level for uranium ingestion proposed by EPA is an oral uptake of $2 \mu\text{g}$ per kg wt/day (ref. 5). Over the last decade, many workers round the globe have estimated uranium content in both water and soil systems⁶⁻²¹.

In the present study, water and soil samples from 20 villages of Fazilka district, Punjab, India have been analysed for uranium concentration. The study also includes computation of various health-related parameters and assessment of groundwater for drinking purposes.

Sampling area

Fazilka district is a part of Malwa region which lies on the southwestern side of Punjab; Bathinda district lies to its east, Ferozepur district to the north, Pakistan to west, Haryana to the south and Sriganaganar district, Rajasthan to the southwest²². Twenty villages of Fazilka district in the vicinity of Abohar city have been selected for the present study (Figure 1). Malwa region in Punjab has become famous for wrong reasons. There has been an unexpected rise in the number of cancer patients in this region over the last decade²³. This demands for examination of the abiotic environmental features of the region. The present study assesses the underground water and soil of this region for uranium concentration, a probable cause for increased carcinogenic activity.

Methodology

Uranium concentration in water

Sixty water samples from 20 villages were examined for uranium concentration using laser-fluorimetry technique.

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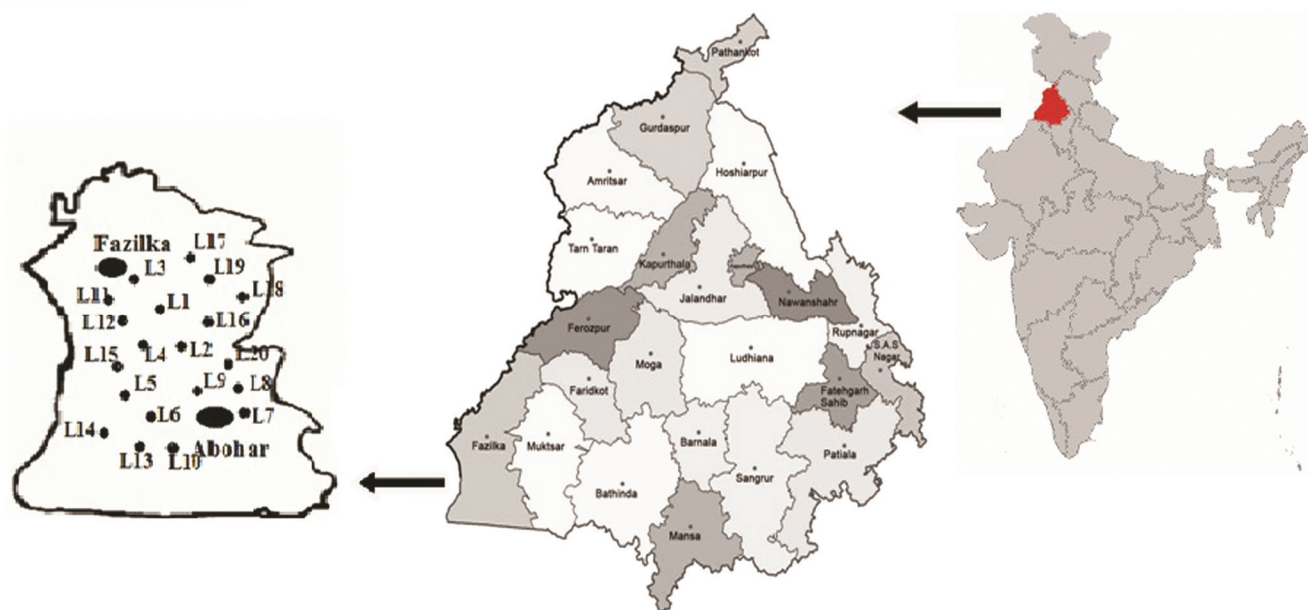


Figure 1. Sampling area of the present study.

The samples were accumulated from groundwater hand pumps. Next 10 ml of the filtered water sample was taken for wet digestion (HClO_4 and HNO_3) to destroy organic substances in the sample. The residue was mixed with fluorescence agent (5% sodium pyrophosphate) and its total volume was increased to 25 ml by adding millipore elix-3 water. The resulting solution was put in a cuvette for uranium concentration measurement²². The device was calibrated in the range 1–100 $\mu\text{g/l}$ using a standard solution made by adding 1.78 g of uranyl acetate dehydrate in 1 litre of millipore elix-3 water containing 1 ml of 70% HNO_3 . To obtain blank counts, a blank sample containing identical amount of fluorescing reagent was used for uranium concentration.

The concentration of uranium in the samples was calculated as²⁴

$$U(\mu\text{g l}^{-1}) = \frac{D_1}{D_1 - D_2} \times \frac{V_1}{V_2} C, \quad (1)$$

where D_1 is the fluorescence due to sample only; D_2 the fluorescence due to sample and uranium standard; V_1 the volume of uranium standard; V_2 the volume of sample and C is the concentration of uranium standard solution.

The methodology is based on luminescence of uranyl ion in solution. The apparatus consists of a molecular nitrogen laser tube as the excitation source, a photomultiplier tube (PMT) and a sample compartment. The laser source emits an intense pulsed UV radiation which is focused on the sample. This excites the uranium complex compounds (which are formed when fluorescence enhancing agent is added). The fluorescence of uranyl ion

is assessed by PMT. Organic matter present in the sample also gives fluorescence when excited by nitrogen laser. However, this fluorescence has much shorter lifetime compared to that of uranyl complex. By measuring the impeded fluorescence signal, fluorescence due to organic substances is removed and only that due to uranium isotopes is measured by PMT. Contribution of organic matter is also nullified using optical filters³.

Health effects of uranium due to intake of groundwater can be estimated through calculation of certain radiological and chemical risk parameters²⁵ which are listed below²⁵.

Annual effective dose (AED)

$$\text{AED} = U_a \times W_{\text{in}} \times D_{\text{coeff}}, \quad (2)$$

where U_a is the uranium concentration (Bq l^{-1}), W_{in} the annual consumption of water (4.05 l day^{-1}) and D_{coeff} is the radioactivity dose conversion factor ($4.5 \times 10^{-8} \text{ SvBq}^{-1}$).

Excess cancer risk (ECR)

$$\text{ECR} = U_a \times \text{RF}, \quad (3)$$

where U_a is the uranium concentration (Bq l^{-1}) and RF is the risk factor (per Bq l^{-1}) which is calculated as

$$\text{RF} = R_{\text{coeff}} \times \text{IR} \times \text{LE}, \quad (4)$$

where R_{coeff} is the risk coefficient for ingestion ($1.19 \times 10^{-9} \text{ Bq}^{-1}$), IR the ingestion rate of water (4.05 l day^{-1}) and LE is the life expectancy (63.7 years).

Table 1. Mean uranium concentration in groundwater and soil samples at different locations

Location and code	Mean uranium concentration in soil (mg kg ⁻¹)	Mean uranium concentration in groundwater (µg l ⁻¹)	SD	Annual effective dose (µSv year ⁻¹)	Excess cancer risk (× 10 ⁻⁴)	Lifetime average daily dose (µg kg ⁻¹ d ⁻¹)
Nihalkhera (L-1)	5.8	15.54	2.9	25.97	0.44	1.17
Dangerkhera (L-2)	2.0	20.32	3.1	33.96	0.57	1.53
Khuikhera (L-3)	1.8	83.99	0.8	141.19	2.37	6.33
Churiwala (L-4)	2.2	65.73	3.6	110.55	1.86	4.96
Panjkoski (L-5)	1.2	59.07	1.2	99.23	1.67	4.45
Killianwali (L-6)	2.8	13.17	1.4	21.97	0.37	0.99
Gobindgarh (L-7)	2.2	12.13	1.4	20.64	0.35	0.91
Kundal (L-8)	3.0	11.19	4.3	18.64	0.31	0.84
Mamukhera (L-9)	1.8	14.87	2.1	24.64	0.41	1.12
Bajitpura (L-10)	3.6	41.21	1.6	69.26	1.17	3.11
Ghallu (L-11)	3.0	24.02	0.2	40.62	0.68	1.81
Jhumianwali (L-12)	3.0	10.20	2.4	17.31	0.29	0.77
Roharianwali (L-13)	2.0	20.67	1.4	34.63	0.58	1.56
Khipanwali (L-14)	1.8	19.16	1.6	31.96	0.54	1.44
BurjMahar (L-15)	1.8	27.88	1.0	46.62	0.78	2.10
Katehra (L-16)	2.6	18.44	3.2	31.30	0.53	1.39
Kamalwala (L-17)	2.8	20.96	2.7	35.29	0.59	1.58
Bodiwala (L-18)	3.2	4.32	1.8	7.32	0.12	0.33
Patrewala (L-19)	2.0	31.25	1.6	52.61	0.88	2.35
Danewala (L-20)	1.6	16.15	0.9	27.30	0.46	1.22

Table 2. Statistical evaluation of mean uranium concentration, annual effective dose and other parameters

Statistical parameter	Mean uranium concentration in soil (mg kg ⁻¹)	Uranium concentration in water (µg l ⁻¹)	Annual effective dose (µSv year ⁻¹)	Excess cancer risk (× 10 ⁻⁴)	Lifetime average daily dose (µg kg ⁻¹ d ⁻¹)	HQ
Minimum	1.2	4.32	7.32	0.12	0.33	0.275
Maximum	5.8	83.99	141.19	2.37	6.33	5.275
Average	2.51	26.51	44.55	0.75	1.99	1.66
SD	0.97	20.14	33.86	0.57	1.52	1.26
GM	2.36	20.92	35.15	0.59	1.58	1.31
GSD	1.40	1.97	1.97	1.98	1.97	1.97
Skewness	1.93	1.71	1.71	1.71	1.71	1.71
Kurtosis	5.49	2.38	2.38	2.36	2.38	2.38
First quartile	1.8	14.44	23.97	0.4	1.09	0.91
Third quartile	3	28.72	48.12	0.80	2.16	1.80

HQ, hazard quotient; SD, standard deviation; GM, geometric mean; GSD, geometric standard deviation.

Lifetime daily dose (LADD)

$$LADD = \frac{EPC}{AT} \times \frac{IR}{BW} \times ET \times LE, \quad (5)$$

where EPC is the exposure point concentration of uranium (µg l⁻¹), ET the exposure frequency (350 days year⁻¹), AT the average time and BW is the average body weight.

Hazard quotient (HQ)

The extent of non-carcinogenic harms due to exposure is indicated in terms of hazard quotient. It is the ratio of LADD and reference dose (RD) which is taken to be 1.2 µg kg⁻¹ day⁻¹

$$HQ = LADD/RD.$$

Uranium concentration in soil

Forty soil samples collected from 20 villages were assessed for uranium concentration using wavelength dispersive X-ray fluorescence (WDXRF) technique. This is the most popular method for elemental analysis having the ability to detect elements from Be to U. This technique is much faster than γ-spectrometric measurements. To start with, all the samples were dried in an oven at 105°C for 10 h. Next 9 g of each sample was mixed with 2.7 g of binder and the resulting mixture ground to form a homogenized fine powder (particle size ~50 µm). Later 10 g of the above mixture was subjected to 15 tonnes of pressure for 10 sec using a hydraulic press to form a pellet of diameter 34 mm and thickness 4 mm. This pellet was analysed for elemental composition of uranium using a WDXRF spectrometer (model: S8 TIGER, make: Bruker, Germany).

In WDXRF, X-rays produced by a source irradiate the sample (pellet). Uranium present in the sample will emit fluorescent X-radiation with discrete energy which is typical for U. By measuring the intensity of the emitted energy, the amount of uranium in the sample is determined²⁶. The detection system includes a diffraction crystal, a set of collimators and a detector. The crystal diffracts the X-rays with different wavelengths in different angles. By placing the detector (scintillation counter) in a certain direction, the intensity of X-rays with a certain wavelength is estimated. For this purpose, a detector mounted on a goniometer can also be used, which can be moved through an angular range to measure intensities of many wavelengths. The recorded spectra are evaluated by fundamental parameters method using the software linked to the equipment²⁷.

Results and discussion

Table 1 shows results of uranium concentration measured in 60 drinking water samples from the 20 villages studied. Uranium concentration varied from 4.32 to 83.99 $\mu\text{g l}^{-1}$ at different locations. The World Health Organization (WHO) has recommended 30 $\mu\text{g l}^{-1}$ as the permissible limit for uranium in drinking water²⁸ while

the Atomic Energy Regulatory Board (AERB) has suggested 60 $\mu\text{g l}^{-1}$ as the safe limit²⁹. In this study, 24% of the drinking water samples exceeded the safe limits set by WHO, while 9% exceeded the limit set by AERB (Figure 2). Certain health risk factors like annual effective dose, excess cancer risk and lifetime average daily dose were also evaluated. The annual effective dose varied from 7.32 to 141.19 $\mu\text{Sv year}^{-1}$ at different locations with an average value of 44.55 $\mu\text{Sv year}^{-1}$. All the values were within the permissible limit of 100 $\mu\text{Sv year}^{-1}$ as suggested by International Commission on Radiological Protection except for the villages of Khuikhera, Churiwala and Panjkosi, where the values were higher (Figure 3). For assessment of carcinogenic risk due to intake of uranium, excess cancer risk was calculated (Table 1). The values ranged from 0.12×10^{-4} to 2.37×10^{-4} at different locations with an average of 0.75×10^{-4} . Ten per cent of the values was above the safe limit of 1.67×10^{-4} set by AERB. For assessment of non-carcinogenic risk due to uranium intake, lifetime daily dose was calculated (Table 1). The values were between 0.33 and 6.33 $\mu\text{g kg}^{-1} \text{day}^{-1}$ with an average of 1.99 $\mu\text{g kg}^{-1} \text{day}^{-1}$ (Figure 4). AERB had suggested a safe limit of 4.53 $\mu\text{g kg}^{-1} \text{day}^{-1}$ for lifetime daily dose, and 10% of the values were above this limit.

Table 1 also shows the measured uranium concentration in 40 soil samples from the same 20 villages (Figure 5).

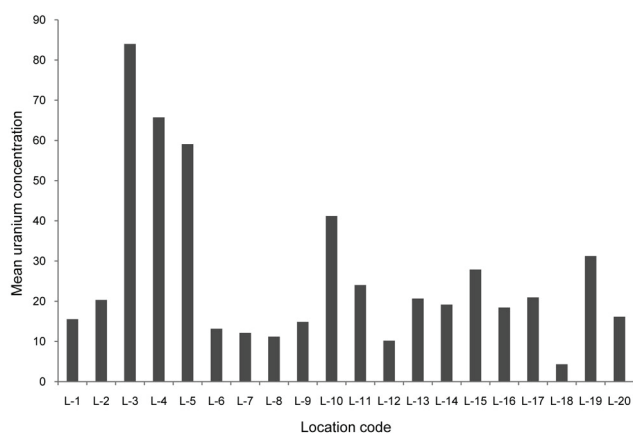


Figure 2. Mean uranium concentration ($\mu\text{g l}^{-1}$) in groundwater samples at different locations.

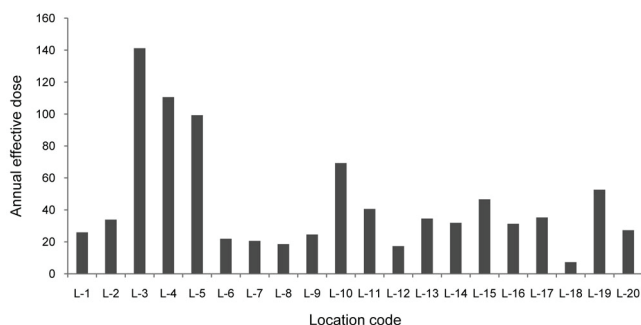


Figure 3. Annual effective dose ($\mu\text{Sv year}^{-1}$) in groundwater samples at different locations.

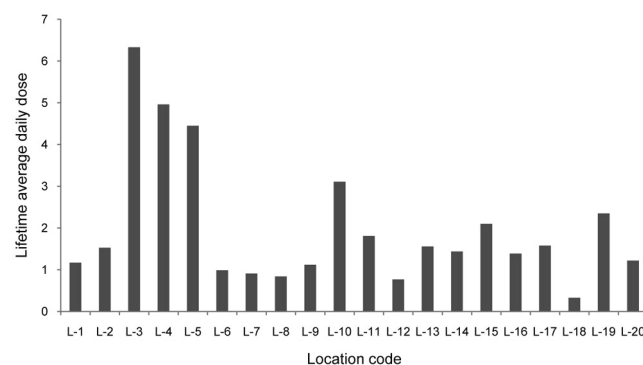


Figure 4. Lifetime average daily dose ($\mu\text{g kg}^{-1} \text{d}^{-1}$) in groundwater samples at different locations.

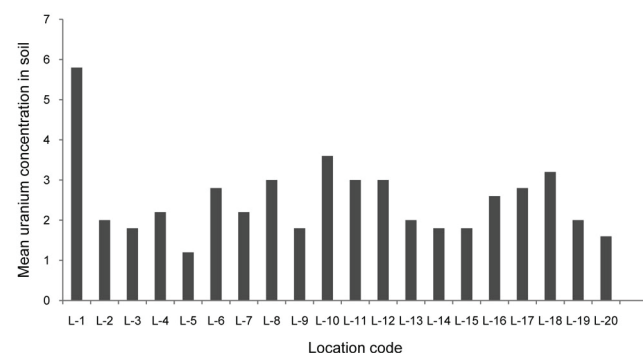


Figure 5. Mean uranium concentration (mg kg^{-1}) in soil at different locations.

Table 3. Comparison of mean uranium concentration in water samples from different areas of Northern India

Area studied	Mean uranium concentration ($\mu\text{g l}^{-1}$)	Reference
Northern Rajasthan	38.48	32
Western Haryana	19.14	33
Northwest Punjab	13.22	34
Western Himachal Pradesh	2.17	35
Jammu, J&K	4.72	1
Udhampur, J&K	11.01	36
Western Punjab	26.51	Present study

All the values were well within the safe limit of 23 mg/kg (ref. 30). Table 2 provides a statistical evaluation of the results summarized in Table 1. Various statistical parameters like geometric mean, geometric standard deviation, skewness, kurtosis, 1st quartile and 3rd quartile were evaluated. Positive values of skewness for uranium concentration in water and soil and for various health-related parameters indicate that the frequency distribution of all of the above has a long right tail, or the distribution is concentrated on the left of the mean. Positive values of kurtosis depict the longish tails or flat peaks of distribution of the above discussed parameters. Table 2 also shows the values of first and third quartiles of distribution of all the parameters. Twenty-five per cent of the uranium concentration values in water samples was below $14.44 \mu\text{g l}^{-1}$ while 75% was below $28.72 \mu\text{g l}^{-1}$, which is roughly the average value of the region. Thus uranium concentration was not very high, barring a few locations.

Elevated values of uranium in drinking water samples seem to be due to natural geology of the region, industrial activities in the region or due to excessive use of phosphate-based fertilizers. Geology of the region indicates that uranium-rich granites form the basement of this region. Interaction of groundwater with these granite rocks might have resulted in high values of uranium measured in the region. Secondly, the sediments of this region have been derived from Shivalik Himalaya, which is known for uranium mineralization³¹. Finally, soil of this region is calcareous in nature²⁴. Thus, water is found to be rich in bicarbonates which are known to be an efficient leaching agent for uranium from soils and sediments. However, no direct correlation was found between uranium concentration in water and soil. This indicates that uranium contamination in groundwater is not due to its concentration in top few layers of the soil of the region. It is the inner core structure of the region well below the top layers which is probably the cause of high uranium concentration in groundwater at some locations. So geology of the region seems to be the most likely reason for higher uranium content, but other possibilities cannot be ruled out.

The uranium concentration estimated in the present study was also compared with those obtained from other

parts of northern India (Table 3). The average uranium concentration from drinking water samples in northern Rajasthan³², western Haryana³³, northwest Punjab³⁴, western Himachal Pradesh³⁵, Jammu region of Jammu and Kashmir¹, and Udhampur region of Jammu and Kashmir³⁶ has been reported to be 38.48, 19.14, 13.22, 2.17, 4.72 and $11.01 \mu\text{g l}^{-1}$ respectively, while it was $26.51 \mu\text{g l}^{-1}$ in the present study. Except from northern Rajasthan, the value estimated in the present study is generally higher than those obtained from other areas of northern India.

Conclusion

The uranium concentration in water samples collected from the studied area varied from 4.32 to $83.99 \mu\text{g l}^{-1}$ with an average value of $26.51 \mu\text{g l}^{-1}$. Nearly one-fourth of the drinking water samples exceeded the safe limits set by WHO. Less than 10% values are above the limit set by AERB. The annual effective dose calculated was found to be high at some locations. Uranium content in soil samples collected from the studied area was well within the permissible limits. The key factor for the increased uranium content at some places seems to be geology of the region (uranium-rich granites which form the base of the region and bicarbonate-rich soil), but other human-related factors cannot be ruled out. The study suggests further research to look for a possible link between increased carcinogenic activity and uranium content in the region.

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