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Uranium Content Anomalies in Groundwater of Patiala District of Punjab (India) for the Assessment of Excess Cancer Risk

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Abstract
Inductively coupled plasma mass spectroscopy (ICPMS) has been used to measure the uranium content of ground water samples of Patiala district of Punjab (India). Out of total 810 habitations covered under this survey, 94 were having uranium content equal or more than 30 ppb (safe limit of the World Health Organization). The aim of this study was to investigate the uranium content of groundwater in the Patiala district of Punjab and to assess the radiological and chemical risk due to the uranium present in water through ingestion. The uranium content of water samples of the 50 villages under investigation varied from 38.00 to 267.00 ppb (μgl^{-1}) with an average value of 68.70 ppb (μgl^{-1}). The excess cancer risk varied from 1.08×10^{-4} to 7.56×10^{-4} with an average value of 1.95×10^{-4} and hazard quotient varied from 0.49 to 3.41 with an average value of 0.88, respectively. The Lifetime Average Daily Dose (LADD) varied from 2.20 to 15.4 5μgkg^{-1} day^{-1} with an average value of 3.97 μgkg^{-1} day^{-1}.

Keywords: *Uranium content, radiological risk, chemical risk, cancer risk*

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INTRODUCTION
The World Health Organization (WHO) [1] recommended a reference level of 30 μgl^{-1} (ppb) as the permissible limit of Uranium in drinking water. The accumulation of uranium inside the human body results in its chemical and radioactive effects for the two important target organs being the kidneys and lungs [2–4]. The major source of supply of uranium to the human body is drinking water, which contributes about 85% of ingested uranium and the rest 15% is contributed by food intake [5]. The transient chemical damage to the kidneys is due to an exposure of about 0.1 mg/kg of body weight of soluble natural uranium [6]. The natural uranium is a radioactive heavy metal; it decays into many other radioactive metals or gases which can further become a health hazard to the public [7]. Uranium itself is a weak radioactive metal but it may be hazardous to human health if its contamination is high in the drinking water. The assessment of health hazards risk is important if the concentration of uranium in water and its extent of getting ingested into human body is higher than the safe limit provided by the WHO [1].

Punjab is facing a crisis situation due to high levels of Uranium (U) and heavy metals in underground water table of Punjab, India. More than two dozen reports have been published in The Tribune (www.tribuneindia.com) during the last decade concerning high toxicity of uranium in the waters of Punjab. The author has reported his findings in The Tribune and other research journals during the last four decades [8–17]. Uranium estimation of groundwater of the Malwa belt of Punjab State and the neighbouring areas in Haryana has been reported by other workers [18–24]. Patnaik *et al.* [25] reported that mobilization of uranium from Siwalik Himalayas is the cause of enhanced geogenic uranium in the Malwa belt of Punjab. The present report is based on the data collected by the Punjab Water Supply and Sanitation Department (PWSSD), Mohali, Punjab, India. It is also available on the Ministry of Water Resources, Government of India, and website: www.indiawater.gov.in/IMIS reports. The objective of the present investigations was health risk assessment due to natural uranium in drinking water in Patiala district of Punjab, India (Figure1).

THE STUDY AREA AND GROUNDWATER QUALITY

Location

Patiala district of Punjab state lies between 29^{o} 49' to 30^{o} 40' north latitudes and 75^{o} 58' to 76^{o} 48' east longitudes. Total geographical area of the district is 3218 sq. km. The Patiala district is divided into five subdivisions (*tehsils*) namely Patiala, Nabha, Ghanaur, Rajpura and Samana comprising eight-community development blocks for the purpose of administration.

Geomorphology and Soil Types [26]

The district area is occupied by Indo-Gangetic alluvial plain and consists of three types of region, through the Upland plain, the Cho-infested Foothill Plain and the floodplain of the Ghaggar river. The elevation of land ranges from 240 to 278 m amsl. Due to arid climate, the soils are light coloured. Tropical arid brown soils exist in major parts of the district. Here soils are deficient in nitrogen, phosphorus and potassium. In Patran and Samana blocks, soils are arid brown. These are calcareous in nature and in most cases *kankar* layers occur.

Ground Water Quality

The district is occupied by Indo-Gangetic alluvial plain of Quaternary age and falls in Ghaggar basin. The ground water occurs in alluvium formations comprising fine to coarse sand, which forms the potential aquifers. In the shallow aquifer (up to 50m) ground water occurs under unconfined/water table conditions, whereas in deeper aquifer, semi-confined/confined conditions exist. The traditional dugwells tapping the shallow aquifer are not in use and most of them have been abandoned; however, this aquifer is being tapped by the hand pumps and shallow tube wells, which are widely used for domestic purposes. The Central Ground Water Board (CGWB) has carried out studies for chemical quality of ground water in the area. The ground water of the district is alkaline in nature. The electrical conductivity (EC) in the area ranges from 687 to 4100 micromhos/cm. Nitrate values range between 0.40–200 mg/l and fluoride concentration ranges from 0.20 to 2.8 mg/l [26], and observed beyond the safe limits suggested by the WHO, thus the ground water is harmful for human consumption at these places.

METHODOLOGY

For collection of samples, 20 ml bottles of superior quality plastic were used. The bottles were washed first with soap solution and then with distilled water. These bottle were rinsed with deionised water and dried. Groundwater from the source was allowed to flow freely before collection in plastic bottles. About 10–20 ml of water was collected from the running water source. For dissolved metal determinations, samples must be filtered through a 0.45μm capsule filter at the field site. Nitric acid (0.5M HNO_3) solubilization is required before the determination of total recoverable uranium. The preservation and digestion of uranium in acid is used in order to aid breakdown of complexes and to minimize interferences by polyatoms.

The uranium analysis of collected water samples has been done using Model 7700 Agilent Series ICP-MS following standard procedure in the Punjab State laboratory set up in Mohali, India. The method measures ions produced by a radiofrequency inductively coupled plasma (ICP). Analyte species originating in a liquid are nebulized and the resulting aerosol is transported by Argon gas into the plasma torch. The ions produced by high temperatures are entrained in the plasma gas and introduced, by means of an interface, into a mass spectrometer. The ions produced in the plasma are sorted according to their mass-to-charge ratios and quantified with a channel electron multiplier. Interferences must be assessed, and valid corrections applied. Interference correction must include compensation for background ions contributed by the plasma gas, reagents, and constituents of the sample matrix.

A mass spectrometer with inductively coupled plasma (ICP) suitable for multi-element and isotope analysis is required. The spectrometer should be capable of scanning a mass range from 5 m/z (AMU) to 240 m/z (AMU) with a resolution of at least 1 mr/z peak width at 5%

of peak height (mr = relative mass of an atom species; z = charge number). The instrument may be fitted with a conventional or extended dynamic range detection system. Most quadrupole ICP-MS, high-resolution ICP-MS and collision cell ICP-MS instrumentation is fit for this purpose. Data analysis is done automatically by inbuilt system of ICP-MS. In addition to Uranium, data for 40 more trace elements can be retrieved using ICP-MS.

THEORETICAL FORMULATION

Ingestion of uranium through drinking water results in both radiological risk (carcinogenic) and chemical risk (noncarcinogenic). The methodology used for the assessment of radiological and chemical risks due to uranium concentrations in the water samples are described below:

Radiological Risk Assessment

Calculation of Excess Cancer Risk

Excess cancer risk from the ingestion of natural Uranium from the drinking water has been calculated according to the standard method given by the USEPA [27]:

$$ECR = Ac \times R$$

Where, 'ECR' is Excess Cancer Risk; 'Ac' is Activity concentration of Uranium (Bql^{-1}) and 'R' is Risk Factor.

Fig. 1: *District Map of Punjab Showing District of Patiala of Punjab, India.*

The risk factor R (per Bql^{-1}), linked with ingestion of Uranium from drinking water may be estimated by the product of risk coefficient (r) of Uranium (1.19×10^{-9}) for mortality and per capita activity intake I. 'I' for Uranium is calculated as product of life expectancy, assumed to be 63.7 years, i.e. 23250 days and daily consumption of water as 4.05 $lday^{-1}$ [28].
I = 4.05 $lday^{-1}$ × 23250 days
Risk Factor (R) = r× I

Chemical Risk Assessment

The chemical toxicity risk for uranium is defined in terms of Lifetime Average Daily Dose (LADD) of uranium through drinking water intake. LADD is defined as the quantity of the substance ingested per kg of body weight per day and is given by the following equation [29, 30]:

$$\mathrm{LADD} = \frac{\mathrm{C} \times \mathrm{IR} \times \mathrm{ED} \times \mathrm{EF}}{\mathrm{AT} \times \mathrm{BW} \times 365}$$

Where, 'C' is the concentration of uranium (μgl^{-1}), IR is the water consumption rate (4.05 $lday^{-1}$), ED is the lifetime exposure duration (63.7 years), EF is the exposure frequency (365 $daysy^{-1}$), BW is average body weight of the receptor (70 kg), and AT is the averaging time, i.e. life expectancy (63.7 years).

Calculation of Hazard Quotient

Hazard quotient (HQ) is the measure of the extent of harm produced due to the ingestion of uranium from drinking water:
HQ=LADD/RfD,
Where, LADD is Lifetime Average Daily Dose, and RfD is the reference dose = 4.53 $\mu gkg^{-1}day^{-1}$.

RESULTS AND DISCUSSION

Groundwater samples were collected from 810 villages falling under the five *tehsils* of Patiala district of Punjab and analysed for uranium content using calibrated ICP-MS. Uranium content varied from 38.00 ppb (Uppli village) to 267.00 ppb (Ahru Kalan village) with an average value of 68.70 ppb for 50 habitations covered under this survey (Table 1). The safe limit of uranium in groundwater is fixed to be 60 ppb by the Atomic Energy Regulatory Board (AERB) [31] in India, while other agencies fix it in much lower limits of 30 ppb (EPA, USA) [26]; 15 ppb (WHO) [11]; 9 ppb (UNSCEAR) [32] and 1.9 ppb (ICRP) [33]. If the observed data of uranium content of water (Table 1) are compared with the guidelines of AERB, 26 samples out of 50 recorded higher uranium content than 60 ppb; hence they fail to qualify the safe limit certification of the AERB, Government of India.

Radiological Risk

The radiological risk has been calculated due to ingestion of natural uranium in the drinking water of 50 habitations covered in this survey, assuming the consumption rate of 4.05 l/day and lifetime expectancy of 63.7 years for both males and females. The excess cancer risk has been observed to be in the range of 1.08–7.56×10^{-4} with average value of 1.95×10^{-4}. The value of excess cancer risk in the surveyed habitations is higher than the maximum acceptable level of 1.67×10^{-4} according to the AERB guidelines. If we assume lifetime water consumption rate of 4.05 l/day with the present uranium content of water, the mean value of excess cancer risk in the surveyed habitations comes out to be 1.95×10^{-4}, which works out to be nearly 2 per 10,000 people. According to the Cancer Registry of Government of India, national average of cancer risk is 80 cancers per million population, for Punjab it is 90 cancers per million but for Malwa belt of Punjab, it is much higher at 136 cancers per million population. Our investigation revealed that for Patiala district of Punjab, it is lower than the Malwa belt of Punjab but 250% higher than the national average with the likelihood of 200 cancers occurring per million of population in Patiala district.

Chemical Toxicity Risk

Uranium is a radioactive heavy metal, so it has health impacts due to its both radioactive and chemical nature. If we take into account chemical toxicity of uranium, the kidneys are the most important target organ. The chemical toxicity of uranium dominates over its radiological toxicity on the kidney in general at lower exposure levels [34]. The chemical toxicity has been estimated from the value of LADD and hazard quotient. Hazard quotient has been estimated by comparing the value of the calculated LADD with the reference dose level of 4.53 $\mu gkg^{-1}day^{-1}$. The reference level has been calculated for the maximum contamination level of uranium in water of 60 ppb (μgl^{-1}). The variation in the values of LADD and hazard quotients have been observed from 2.20–15.45 $\mu g\ kg^{-1}day^{-1}$ and 0.49–3.41, respectively (Table 1).

Table 1: *Uranium Content in Groundwater of Patiala District and Corresponding Risk Factors.*

S.No	Location	Source	Depth (m)	U Conc. (ppb)	U Conc. (Bq l^{1})	Excess cancer risk * 10^{-4}	LADD ($\mu g\ kg^{-1}\ day^{-1}$)	Hazard Quotient
1	Ahru Kalan	NULL	NULL	267.00	6.75	7.56	15.45	3.41
2	Ahru Khurd	NULL	NULL	267.00	6.75	7.56	15.45	3.41
3	Daun Kalan	Tubewell	285.00	127.60	3.23	3.61	7.38	1.63
4	Dera Xen Retd.	Tubewell	118.26	87.76	2.22	2.49	5.08	1.12
5	Chunagra	Tubewell	118.26	87.76	2.22	2.49	5.08	1.12
6	Todarwal	NULL	NULL	83.34	2.11	2.36	4.82	1.06
7	Purbia Basti	Tubewell	NULL	82.93	2.10	2.35	4.80	1.06
8	Kalyan	Tubewell	NULL	82.93	2.10	2.35	4.80	1.06
9	DeraBahmnaInderpura	Tubewell	NULL	82.93	2.10	2.35	4.80	1.06
10	Dera Xen Retd.	Tubewell	150.00	76.10	1.92	2.16	4.40	0.97
11	Chunagra	Tubewell	150.00	76.10	1.92	2.16	4.40	0.97
12	Birdhno	NULL	NULL	75.80	1.92	2.15	4.39	0.97
13	DeraBahmnaInderpura	NULL	NULL	73.76	1.86	2.09	4.27	0.94
14	Kalyan	NULL	NULL	73.76	1.86	2.09	4.27	0.94
15	Purbia Basti	NULL	NULL	73.76	1.86	2.09	4.27	0.94
16	AsseMajra	NULL	NULL	73.76	1.86	2.09	4.27	0.94
17	Inderpura	NULL	NULL	73.76	1.86	2.09	4.27	0.94
18	Rathian	NULL	NULL	72.93	1.84	2.07	4.22	0.93
19	Khanora	NULL	NULL	68.81	1.74	1.95	3.98	0.88
20	Dhingi	NULL	NULL	67.04	1.69	1.90	3.88	0.86
21	Seona	NULL	NULL	66.54	1.68	1.88	3.85	0.85
22	Wazidpur	NULL	NULL	62.50	1.58	1.77	3.62	0.80
23	Dera Saini Majra	NULL	NULL	62.50	1.58	1.77	3.62	0.80
24	Paidan	Tubewell	165.00	61.10	1.54	1.73	3.54	0.78
25	Ransihpura	NULL	NULL	61.00	1.54	1.73	3.53	0.78
26	Raipur	Raw Water of RO	42.67	60.63	1.53	1.72	3.51	0.77
27	Rasulpur	Hand Pump	70.00	59.30	1.50	1.68	3.43	0.76
28	Haripur Jhugian	Hand Pump	70.00	59.30	1.50	1.68	3.43	0.76
29	Katlahar	Hand Pump	70.00	59.30	1.50	1.68	3.43	0.76
30	Budanpur	NULL	NULL	51.33	1.30	1.45	2.97	0.66
31	Sarkari Farm	NULL	NULL	51.33	1.30	1.45	2.97	0.66
32	Khaktan Khurd	NULL	NULL	51.33	1.30	1.45	2.97	0.66
33	Bugga Khurd	NULL	NULL	50.75	1.28	1.44	2.94	0.65
34	Hariyou Khurd	Tubewell	NULL	49.50	1.25	1.40	2.86	0.63
35	Ohjhan	Hand Pump	80.00	49.30	1.25	1.40	2.85	0.63
36	Rasauli	Tubewell	NULL	46.86	1.18	1.33	2.71	0.60
37	DeraShingara Singh	Hand Pump	90.00	42.20	1.07	1.20	2.44	0.54
38	Birdhno	Tubewell	90.00	42.00	1.06	1.19	2.43	0.54
39	Uppli	NULL	NULL	41.76	1.06	1.18	2.42	0.53
40	Paror	NULL	NULL	41.76	1.06	1.18	2.42	0.53
41	Kathmathi	Tubewell	NULL	39.94	1.01	1.13	2.31	0.51
42	Gandian	Tubewell	300.00	39.30	0.99	1.11	2.27	0.50
43	Bathonia Kalan	Tubewell	300.00	39.30	0.99	1.11	2.27	0.50
44	Bathonia Khurd	Tubewell	300.00	39.30	0.99	1.11	2.27	0.50
45	Paror	Tubewell	165.00	38.90	0.98	1.10	2.25	0.50
46	Uppli	Tubewell	165.00	38.90	0.98	1.10	2.25	0.50
47	Ghungran	Tubewell	200.00	38.80	0.98	1.10	2.24	0.50
48	Shahpur Raian	Tubewell	200.00	38.80	0.98	1.10	2.24	0.50
49	Balamgarh	NULL	NULL	38.77	0.98	1.10	2.24	0.50
50	Uppli	NULL	NULL	38.00	0.96	1.08	2.20	0.49
			Average	68.70	1.74	1.95	3.97	0.88

**Cancer risk is the likelihood, or chance, of getting cancer. We say "excess cancer risk" because we have a "background risk" of about one in four chances of getting cancer. In other words, in a million people, it is expected that 250,000 individuals would get cancer from a variety of causes. If we say that there is a "one in a million" excess cancer risk from a given exposure to a contaminant, we mean that if one million people are exposed to a carcinogen at a certain concentration over their lifetime, then one cancer above the background chance, or the 250,000th cancer, may appear in those million persons from that particular exposure [35].*

CONCLUSIONS

i. The concentration of uranium in ground water samples collected from the tubewells, hand-pumps or RO raw water of several villages of Patiala district was found to be higher than the safe limit of 30 ppb recommended by the WHO, with 26 samples showing higher values than AERB limit of 60 ppb.
ii. The cancer risk due to presence of uranium in groundwater is found to be lower than the Malwa belt of Punjab but much higher than the average national value for India.
iii. The source of uranium enhancement in Patiala district of Punjab may be attributed to mobilization of uranium by Ghaggar river and its tributaries, for example, Patiala Nadi [25].
iv. It will be of interest to study the epidemiological effects of uranium in groundwater on the inhabitants of Patiala district of Punjab, India.

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