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Uranium Content Anomalies in Groundwater of Barnala District of Malwa Belt 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 the ground water samples of Barnala district of Malwa belt of Punjab (India). Out of total 115 habitations covered under this survey, 110 are having uranium content more than 60 ppb (safe limit of AERB for India). The aim of this study is to investigate the uranium content of the ground water in the Malwa belt of Barnala district of Punjab and to assess the radiological and chemical risk due to the uranium present through ingestion. The uranium content of the water samples of the villages under investigation varies from 62.91–290.60 ppb (μgl^{-1}) with an average value of 127.90 ppb (μgl^{-1}). The excess cancer risk varies from 1.78–8.23×$10^{-4}$with average value of 3.62 ×$10^{-4}$and hazard quotient varies from 0.80 to 3.71 with average value of 1.63, respectively. The LADD varies from 3.64–16.81 ($\mu g\ kg^{-1}\ day^{-1}$) with average value of 7.40 ($\mu g\ kg^{-1}\ day^{-1}$).

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 µg l^{-1} as the permissible limit of Uranium in drinking water. The accumulation of the 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 the uranium to the human body is drinking water, which contributes about 85% of ingested uranium and the rest 15% is contributed by the 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 WHO [1].

Punjab is facing a crisis situation due to high levels of Uranium (U) and heavy metals in underground water table of Punjab. More than two dozen reports have been published in The Tribune (www.tribuneindia.com) during the last decade concerning high toxicity of U 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 the groundwater of the Malwa belt of Punjab State and the neighbouring areas in Haryana has been reported by other workers [18–24]. 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 Ministry of Water Resources, Government of India, and website: www.indiawater.gov.in/IMIS reports. The objective of the present investigations is health risk assessment due to natural uranium in drinking water in Barnala district of Punjab, India (Figure 1).

Fig. 1: *District Map of Punjab showing district of Barnala in the Malwa Belt.*

THE STUDY AREA AND GROUNDWATER QUALITY

Location

Barnala district is located in the south-western part of the Punjab state with co-ordinates between 30.00 to 30.52' north latitudes and 75.15' to 75.75' east longitudes. Total geographical area of the district is 1410 sq. km. It has been carved out of the Sangrur district and surrounded by districts of Moga, Ludhiana, Sangrur, Mansa and Bathinda.

Geomorphology and Soil Types [25]

The territory of Barnala district of Punjab forms part of the Indo-Gangetic plain. The whole territory of the block is plain in general. The master slope of the area is towards the south west direction. There is no major drainage system in the area except the Dhaula drain which intersects its territory. This drain carries flood water when heavy rainfall occurs in the catchment area. Abohar branch of Sirhind canal system passes in south eastern part of the district. Soil of the district is loamy sand in general with patches of sandy loam *kaller* land interspersed at a few places.

Ground Water Quality

Central Ground Water Board (CGWB), Ministry of Water Resources, Government of India has carried out studies [25] for chemical quality of ground water in the area. The ground water of the district is alkaline in nature. The EC in the area ranges from 595 to 1260 Micromhos/cm. Nitrate concentration values range between 0.40 to 200 mg/l and fluoride concentration ranges from 0.45 to 5.0 mg/l. At few places high fluoride and nitrate have been observed, thus the ground water in these places is harmful for human consumption.

METHODOLOGY

For collection of samples, 20 ml bottles of superior quality plastic are used. The bottles are washed first with soap solution and then with distilled water. These are rinsed with deionised water and dried. Groundwater from the source is allowed to flow freely before collection in plastic bottles. 10–20 ml of water is 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 poly-atoms.

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. The method measures ions produced by a radiofrequency inductively coupled plasma. 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 the uranium through drinking water results in both the radiological risk (carcinogenic) and chemical risk (non-carcinogenic). The methodology used for the assessment of the 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 [26]:

$$ECR = Ac \times R$$

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

The risk factor R (per Bql^{-1}), linked with ingestion of Uranium from the drinking water may be estimated by the product of the 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 $lday^{-1}$ [27].

$I = 4.05\ lday^{-1} \times 23250$ days

Risk Factor (R) = $r \times I$

Chemical Risk Assessment

The chemical toxicity risk for Uranium is defined in terms of Lifetime Average Daily Dose (LADD) of the 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 [28, 29]:

$$LADD = C \times IR \times ED \times EF / AT \times BW \times 365$$

Where 'C' is the concentration of the 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 the drinking water.

$$HQ = LADD/RfD$$

Where, LADD is Lifetime Average Daily Dose, and RfD is the reference dose = 4.53 μg $kg^{-1}day^{-1}$.

RESULTS AND DISCUSSION

Groundwater samples were collected from villages falling under the Tehsils (Barnala and Tapa) of Barnala district of Punjab and analysed for Uranium content using calibrated ICP-MS. Uranium content varies from 62.91ppb (Raja village Tubewell) to 290.60 ppb (Tubewell at Ramgarh) with an average value of 127.90 ppb for 110 habitations covered under this survey (Table 1). The safe limit of uranium in groundwater is fixed to be 60 ppb by Atomic Energy Regulatory Board (AERB) [30] in India, while other agencies fix it in much lower limits of 30 ppb (EPA, USA) [26]; 15 ppb (WHO) [11]; 9 ppb (UNSCEAR) [31] and 1.9 ppb (ICRP) [32]. If the observed data of uranium content of water (Table 1) are compared with the guidelines of AERB, all the samples record higher Uranium content than 60 ppb; hence they fail to qualify the safe limit certification of AERB, Government of India.

Radiological Risk

The radiological risk has been calculated due to ingestion of natural uranium in the drinking water of 110 habitations covered in this survey, assuming the consumption rate of 4.05 litre/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 1.78 to $8.23x10^{-4}$with average value of $3.62x10^{-4}$. The value of the excess cancer risk in the surveyed habitations is higher than the maximum acceptable level of 1.67×10^{-4} according to AERB guidelines. If we assume lifetime water consumption rate of 4.05 litre/day with the present uranium content of water, the mean value of excess cancer risk in the surveyed habitations comes out to be 3.62×10^{-4}, which works out to be nearly 4 per 10,000 people. According to 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 reveals that for Barnala district in Malwa belt of Punjab, it has assumed alarming proportions at 362 cancers per million.

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 the uranium, the kidneys are the most important target organ. The chemical toxicity of the uranium dominates over its radiological toxicity on the kidney in general at lower exposure levels [33]. The chemical toxicity has been estimated from the value of lifetime average daily dose (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 μg $kg^{-1}day^{-1}$. The reference level has been calculated for the maximum contamination level of the uranium in water of 60 ppb (μgl^{-1}). The variation in the values of the LADD and Hazard quotients has been observed from 3.64–16.81μg/kg/day and from 0.80 to 3.71, respectively (Table 1).

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

Sr. No.	Location	Source	Uranium Concentration (ppb)	Uranium Concentration (Bq l^{-1})	Excess Cancer risk* $x10^{-4}$	LADD (μg kg^{-1} day^{-1})	Hazard Quotient
1	Khuddi Khurd	Tubewell	70.81	1.79	2.01	4.1	0.9
2	Khuddi Khurd	RO Raw Water	143	3.62	4.05	8.27	1.83
3	Khuddi Kalan	RO Raw Water	144	3.64	4.08	8.33	1.84
4	Khuddi Kalan	RO Raw Water	144	3.64	4.08	8.33	1.84
5	Dhaner	RO Raw Water	164.7	4.16	4.67	9.53	2.1
6	Kattu	RO Raw Water	113	2.86	3.2	6.54	1.44

Sr. No.	Location	Source	Uranium Concentration (ppb)	Uranium Concentration (Bq l^{-1})	Excess Cancer risk* $x10^{-4}$	LADD (µg kg^{-1} day^{-1})	Hazard Quotient
7	Bhadalwad	RO Raw Water	84.18	2.13	2.38	4.87	1.08
8	Lohgarh	Tubewell	90.9	2.3	2.57	5.26	1.16
9	Maur Nabha	Tubewell	65.8	1.66	1.86	3.81	0.84
10	Maur Nabha	Tubewell	65.8	1.66	1.86	3.81	0.84
11	Maur Patiala	Tubewell	65.8	1.66	1.86	3.81	0.84
12	Ramgarh	Tubewell	83.5	2.11	2.37	4.83	1.07
13	Attar Singh Wala	Tubewell	85.8	2.17	2.43	4.96	1.1
14	Badbar	Tubewell	117.89	2.98	3.34	6.82	1.51
15	Badra	Tubewell	89.66	2.27	2.54	5.19	1.15
16	Bhaini Mehraj	Tubewell	83.22	2.1	2.36	4.81	1.06
17	Bhathlan	Tubewell	72.23	1.83	2.05	4.18	0.92
18	Patti Sohlan	Tubewell	99.9	2.53	2.83	5.78	1.28
19	Cheema	Tubewell	99.9	2.53	2.83	5.78	1.28
20	Dangarh	Tubewell	92.5	2.34	2.62	5.35	1.18
21	Dhaunala Khurd	Tubewell	68.2	1.72	1.93	3.95	0.87
22	Dhaunala Khurd	Tubewell	68.2	1.72	1.93	3.95	0.87
23	Dhola 1	Tubewell	83.56	2.11	2.37	4.83	1.07
24	Dhola 2	Tubewell	73.7	1.86	2.09	4.26	0.94
25	Fatehgarh Chhanna	Tubewell	105.5	2.67	2.99	6.1	1.35
26	Jhaloor	Tubewell	101.6	2.57	2.88	5.88	1.3
27	Karmgarh	Tubewell	87.05	2.2	2.47	5.04	1.11
28	Karmgarh	Tubewell	87.05	2.2	2.47	5.04	1.11
29	Khuddi Kalan	Tubewell	70.42	1.78	1.99	4.07	0.9
30	Khuddi Kalan	Tubewell	70.42	1.78	1.99	4.07	0.9
31	Khuddi Khurd	Tubewell	138.82	3.51	3.93	8.03	1.77
32	Pakho Kalan	Tubewell	84.64	2.14	2.4	4.9	1.08
33	Pakho Kalan	Tubewell	84.64	2.14	2.4	4.9	1.08
34	Patti Sekhwa	Tubewell	122.2	3.09	3.46	7.07	1.56
35	Pharwahi	Tubewell	218.79	5.53	6.2	12.66	2.79
36	Rajia	Tubewell	62.91	1.59	1.78	3.64	0.8
37	Sekha	Tubewell	101.8	2.57	2.88	5.89	1.3
38	Sekha	Tubewell	101.8	2.57	2.88	5.89	1.3
39	Uppli	Tubewell	163	4.12	4.62	9.43	2.08
40	Bhadalwad	Tubewell	218.2	5.52	6.18	12.62	2.79
41	Bhadalwad	Tubewell	218.2	5.52	6.18	12.62	2.79
42	Bihla	Tubewell	157.7	3.99	4.47	9.12	2.01
43	Chananwal	Tubewell	173.1	4.38	4.9	10.02	2.21
44	Chhappa	Tubewell	168.2	4.25	4.76	9.73	2.15
45	Hardaspura	Tubewell	168.2	4.25	4.76	9.73	2.15
46	Chiniwal Kalan	Tubewell	137.3	3.47	3.89	7.94	1.75
47	Chhiniwal Khurd	Tubewell	131.8	3.33	3.73	7.63	1.68
48	Chohanke Kalan	Tubewell	76.5	1.93	2.17	4.43	0.98
49	Chohanke Khurd	Tube well	159.7	4.04	4.52	9.24	2.04
50	Chohanke Khurd	Tubewell	159.7	4.04	4.52	9.24	2.04
51	Sadowal	Tubewell	160.7	4.06	4.55	9.3	2.05
52	Gagewal	Tubewell	160.7	4.06	4.55	9.3	2.05
53	Gangohar	Tubewell	158.7	4.01	4.5	9.18	2.03
54	Gehal	Tubewell	233.7	5.91	6.62	13.52	2.98
55	Kalal Majra	Tubewell	190.4	4.81	5.39	11.02	2.43
56	Kalal Majra	Tubewell	190.4	4.81	5.39	11.02	2.43
57	Kalala	Tubewell	165.3	4.18	4.68	9.56	2.11
58	Khiali	Tubewell	193.6	4.89	5.48	11.2	2.47
59	Kirpal S wala	Tubewell	185.5	4.69	5.25	10.73	2.37
60	Kirpal S Wala	Tubewell	185.5	4.69	5.25	10.73	2.37
61	Kurar	Tubewell	170.7	4.32	4.84	9.88	2.18
62	Bhamanian	Tubewell	206.7	5.23	5.86	11.96	2.64

Sr. No.	Location	Source	Uranium Concentration (ppb)	Uranium Concentration (Bq l^{-1})	Excess Cancer risk* x10^{-4}	LADD (µg kg^{-1} day^{-1})	Hazard Quotient
63	Kutba	Tubewell	206.7	5.23	5.86	11.96	2.64
64	Lohgarh	Tubewell	194.9	4.93	5.52	11.28	2.49
65	Mehal Kalan Sodhe	Tubewell	149.6	3.78	4.24	8.66	1.91
66	Mehal Kalan	Tubewell	149.6	3.78	4.24	8.66	1.91
67	Mehal Kalan	Tubewell	149.6	3.78	4.24	8.66	1.91
68	Mehal Khurd	Tubewell	143.6	3.63	4.07	8.31	1.83
69	Mehal Khurd	Tubewell	143.6	3.63	4.07	8.31	1.83
70	Moom	Tubewell	152.9	3.87	4.33	8.85	1.95
71	Nariangar hSohian	Tubewell	156	3.94	4.42	9.03	1.99
72	Nihaluwal	Tubewell	191.4	4.84	5.42	11.07	2.44
73	Pandori	Tubewell	159.4	4.03	4.52	9.22	2.04
74	Sahour	Tubewell	117.5	2.97	3.33	6.8	1.5
75	Wazid Kekalan	Tubewell	73.5	1.86	2.08	4.25	0.94
76	Wazid Ke Khurd	Tubewell	167.5	4.23	4.74	9.69	2.14
77	Alkara	Tubewell	189.6	4.79	5.37	10.97	2.42
78	Mallian	Tubewell	67.3	1.7	1.91	3.89	0.86
79	Bakhatgarh	Tubewell	67.3	1.7	1.91	3.89	0.86
80	Bhotna	Tubewell	163.3	4.13	4.63	9.45	2.09
81	Bhotna	Tubewell	163.3	4.13	4.63	9.45	2.09
82	Burj Fatehgarh	Tubewell	156.6	3.96	4.44	9.06	2
83	Patti Daraka	Tubewell	156.6	3.96	4.44	9.06	2
84	Cheema	Tubewell	99.9	2.53	2.83	5.78	1.28
85	Patti Sohlan	Tubewell	99.9	2.53	2.83	5.78	1.28
86	Chhana Gulab Singh Wala	Tubewell	92.5	2.34	2.62	5.35	1.18
87	Chung	Tubewell	141.3	3.57	4	8.18	1.8
88	Daraj	Tubewell	112.3	2.84	3.18	6.5	1.43
89	Daraka	Tubewell	112.3	2.84	3.18	6.5	1.43
90	Deepgarh	Tubewell	152.7	3.86	4.33	8.83	1.95
91	Majhu ke	Tubewell	152.7	3.86	4.33	8.83	1.95
92	Dhilwan Patiala	Tubewell	146.6	3.71	4.15	8.48	1.87
93	Dharmpura	Tubewell	83.5	2.11	2.37	4.83	1.07
94	Jandsar	Tubewell	83.5	2.11	2.37	4.83	1.07
95	Maksudan	Tubewell	83.5	2.11	2.37	4.83	1.07
96	Jangiana	Tubewell	163.7	4.14	4.64	9.47	2.09
97	Maur Nabha	Tubewell	156.6	3.96	4.44	9.06	2
98	Maur Nabha	Tubewell	156.6	3.96	4.44	9.06	2
99	Maur Patiala	Tubewell	156.6	3.96	4.44	9.06	2
100	Nainewal	Tubewell	175.3	4.43	4.97	10.14	2.24
101	Patti Jaimal Singh	Tubewell	74.6	1.89	2.11	4.32	0.95
102	Ramgarh	Tubewell	290.6	7.35	8.23	16.81	3.71
103	Sandhu Kalan	Tubewell	97.1	2.45	2.75	5.62	1.24
104	Sukhpura	Tubewell	68.0	1.72	1.93	3.93	0.87
105	Kothe Nim wale	Tubewell	68.0	1.72	1.93	3.93	0.87
106	Kothe Taran Taran	Tubewell	68.0	1.72	1.93	3.93	0.87
107	Santpura	Tubewell	66.7	1.69	1.89	3.86	0.85
108	Ugo ke	Tubewell	66.7	1.69	1.89	3.86	0.85
109	Ugo ke	Tubewell	66.7	1.69	1.89	3.86	0.85
110	Jawanda Pindi	Handpump	72.5	1.83	2.05	4.19	0.93

**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 [34].*

CONCLUSIONS

(i) The concentration of the uranium in ground water samples collected from the Tubewells or RO raw water of several villages of Barnala district is found to be higher than the safe limit of 60 ppb recommended by AERB, India.
(ii) The cancer risk due to presence of U in groundwater is found to be among the highest for any district in India.
(iii) Our study establishes that uranium content in the Malwa belt is higher than Majha or Doaba belts of Punjab. If agricultural practices are similar in all districts of Punjab, e.g., use of fertilizers and crop pattern etc., then what is the source of U enhancement in Barnala district of Punjab? This needs to be investigated further.
(iv) It will be of interest to study the epidemiological effects of U in groundwater on the inhabitants of Barnala district of Punjab, India.

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