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Natural Uranium Content in Ground Waters of Mohaliand Fatehgarh Districts of North Punjab (India) for the Assessment of Excess Cancer Risk

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Hardev Singh Virk [α], Rajan Jakhu [σ] & Pargin Bangotra [ρ]

Abstract- LED Fluorimeter has been used to measure the uranium content of the ground water samples of Mohali and Fatehgarh districts of North Punjab (India).33 locations have been selected for the present investigation. The aim of this study is to investigate the uranium content of the ground water in Northern districts of Punjab for sake of comparison with its occurrence in Southern districts of Punjab; and to assess the radiological and chemical risk due to the uranium present through ingestion. The uranium concentration of the water samples of the studied villages varies from 0.63 to 57.82 μgl^{-1} with an average value of 16.93 μgl^{-1}.Theuranium content of all the samples in groundwater lies within the safe limit of 60 μgl^{-1}(ppb) of uranium proposed by AERB, India.

I. Introduction

The presence of natural Uranium in rocks, soils, plants and even in sea water makes its transportation easy in the environment. The rocks of the particular area are the prime source of the uranium to the environment. The solubility of the uranium in water in hexavalent (U^{6+}) form and to precipitate as a discrete mineral in tetravalent (U^{4+}) form, the uranium got deposited in the earth's surface provided to the favorable geological or environmental conditions. Surface water and especially ground water plays a vital role in the migration and redistribution of the nuclides in the earth's crust. Uranium present in water is transferred to plants and hence it enters the food chain and it becomes a source of health hazard to the humans. The World Health Organization recommended a reference level of the permissible limit of Uranium in drinking water 30 $\mu g\ l^{-1}$ (WHO) [1]. The accumulation of the uranium inside the human body results in its chemical and radioactive effects for two important target organs being the kidneys and lungs [2 - 4]. Uranium and radium have the bone seeking properties hence the kidneys, liver and the bones become the principle sites of deposition. The toxicity of uranium depends upon many factors like the route of exposure, particle solubility, contact time, and route of elimination [5]. Drinking water is the major source of the uranium to the human body. Drinking water contributes about 85% and food contributes about 15% of ingested uranium [6]. An exposure of about 0.1 mg/kg of body weight of soluble natural uranium results in transient chemical damage to the kidneys [7]. Uranium is a radioactive heavy metal, it decays into many other radioactive metals or gases which can further become a health hazard [8]. Though Uranium is a weak radioactive metal, if uranium content of the drinking water is high it may be hazardous. Due to high concentration of uranium in water and its extent of getting ingested into human body, the assessment of risk of health hazards are important. Uranium estimation of water systems of the Punjab State and the neighboring areas has been reported by some workers [9 - 15]. The objective of present investigations is health risk assessment due to natural uranium in drinking water in Mohai and Fatehgarh districts of North Punjab.

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II. The Study Area

a) Location

S.A.S Nagar (Mohali) district is located in the eastern part of the Punjab state and lies between North latitudes of 30°21´00" and 30°56´00" and East longitudes of 76°30´00" and 76°55´00" covering a geographic ambience of 1189 sq.km. The district is bounded by Patiala and Fatehgrah Sahib districts in the south-west, Ropar district in the northwest, Chandigarh and Panchkula in the east and Ambala district of Haryana state in the south. Fatehgarh Sahib district is located in southeastern part of Punjab state and lies between 300 25' 00" to 300 45' 45" north latitude & 760 04' 30" to 760 35' 00" east longitude covering an area 1147 sq. km.

b) Geomorphology and Soil types

The area can be broadly grouped into two depending upon its geomorphic features as alluvial fan and alluvial plains. Alluvial fans are deposited by hill torrents with a wavy plain rather than a steep slope. Adjacent to the alluvial fan are the alluvial plains which forms a part of large Indo-Gangetic Quaternary basin

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comprises of thick sand and silty sand layers inter-bedded with silt and clay beds. The alluvial plains are of vital economic value as it supports the dense population of the district. The soils are mainly developed on alluvium under the dominant influence of climate followed by topography and time. The major soil type of the district is weakly solonized tropical arid brown soils. In Fatehgarh Sahib district,the soils are loamy sand at the surface and calcareous sandy loam in subsurface layers. Sand constitutes 80% in the soil profile, silt constitutes 11%, and clay 9% in the soils.

III. Methodology

a) Sampling

Sample collection was done in both the districts in a contiguous area starting from Mohali tehsil, then entering Fatehgarh tehsil and winding up in Mohali in a circular loop. Before collecting the sample, we run the hand-pump or motor for few minutes and then collected the samples in the pre-processed bottles after rinsing twice with the water to be collected. Samples were filtered with 0.45 micron filter paper. The samples were analyzed within a week.

b) LED Fluorimeter

Quantalase has developed Fluorimeters which use banks of pulsed LEDs to excite fluorescence in sample under study. The wavelength, pulse duration and peak power of the LED output can be set to match the excitation requirements of the sample. The fluorescence is detected by a pulsed photomultiplier. Suitable filters after the LEDs and before the photomultiplier tube prevent LED light from reaching the photomultiplier tube directly. The filters can be broadband coloured glass filters or multilayer narrow band filters. The instrument is controlled by a microcontroller which pulses the LEDs and photo-multiplier tube. The microcontroller also controls the ADC which convert the fluorescence signal from photomultiplier to digital form for further processing. A single board computer averages the photomultiplier output over 2000 pulses and carries out any calculations necessary. A touch screen display permits the operator to set necessary parameters and also display the fluorescence measurement.

c) Calibration of Fluorimeter

Standard solution of Uranium is used to calibrate LED Fluorimeter. The instrument was calibrated in the range of 1-100 ppb using a stock solution of standard which was prepared by dissolving 1.78g uranyl acetate dehydrate ($(CH_3\ COO)_2\ UO_2.2H_2O$) in 1L of Millipore elix-3 water containing 1ml of HNO_3. The blank sample containing the same amount of fluorescing reagent was also measured for the uranium concentration. 5% phosphoric acid in ultra-pure water was used as fluorescence reagent. All reagents used for experimental work were of analytical grade.

d) Preparation of FLUREN (Buffer Solution)

Weigh 5gms of Sodium Pyrophosphate powder and add it to a flask/plastic bottle. Add 100ml. of double distilled water and shake well to dissolve the Sodium Pyrophosphate powder. Add Ortho-phosphoric acid drop by drop while monitoring the pH of solution until a pH of 7 is reached. This is the desired buffer solution, also called FLUREN.

Adding buffer solution to a uranium sample increases the fluorescence yield by orders of magnitude. It is recommended that 1 part of buffer solution be added to 10 parts of uranium sample solution and this mixture be used for measurements.

e) Analytical Procedure

A water sample of quantity 6ml is used to find its uranium content. The water sample is taken in the clean and dry quartz cuvette made up of ultrapure fused silica. The instrument was calibrated with the standard uranium solution of known activity. The water sample of quantity 6 ml is mixed with 10% of the buffer solution. Buffer solution is made from sodium pyrophosphate and orthophosphoric acid of pH 7. Buffer solution is used to have the same fluorescence yield of all the uranium complexes present in the water.

The concentration of the uranium in the water sample is calculated as follows:

$$\text{Calibration factor CF} = \frac{\text{Concentration of Uranium in standard solution}}{\text{Fluorescence of standard} - \text{Fluorescence of water}}$$

$$\text{Concentration of uranium in water sample} = \text{CF x (Fluorescence from sample} - \text{Fluorescence from water)}$$

All these calculations are done by the instrument itself. The instrument averages the fluorescence for 256 pulses and displays the average value of U concentration in the sample.

f) Theoretical Formulation

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

g) 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 [17].

$$ECR = Ac \times R$$

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

The risk factor R (per Bq l^{-1}), linkedwith 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 as 63.7 years, i.e. 23250 days and daily consumption of water as 4.05 $lday^{-1}$ [18].

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

$$\text{Risk Factor (R)} = r\times I$$

h) 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 [19, 20].

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

Where 'C' is the concentration of the uranium($\mu g l^{-1}$), 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 days y^{-1}), BW is average body weight of the receptor (70kg), and AT is the Averaging time i.e. life expectancy (63.7 years).

i) 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.

$$HQ = \frac{LADD}{RfD}$$

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

IV. Results and Discussion

Groundwater samples were collected from villages falling under Mohali and Fatehgarh Tehsils of both these districts of Punjab (India) and analysed for Uranium content using calibrated LED Flourimeter (Quantalase Make). Uranium content varies from 0.63 ppb (RO filtered water) to 24.20 ppb (Motor Driven Pump) in Mohali district. In Fatehgarh district, the U content varies from 2.14 ppb (RO System in Reona) to 57.82 ppb for a deep bore Tubewell in Banda Bahadur Engg. College Campus. In Badali Mai Ki village, U content in water of hand pump is 17.22 ppb while it is below detection limit (BDL) in RO filtered water being supplied to the village. It clearly proves that RO System is highly efficacious for getting rid of Uranium from groundwater in Punjab. The safe limit of uranium in groundwater is fixed to be 60 ppb by AERB [21] in India, while other agencies fix it in much lower limits of 30 ppb (EPA, USA)[17]; 15 ppb (WHO)[1]; 9 ppb (UNSCEAR)[22] and 1.9 ppb (ICRP) [23]. If the observed data of uranium content of water (Table 1) is compared with the guideline of AERB, none of the samples record higher than 60 ppb, hence qualify the safe limit certification of AERB, Government of India.

a) Radiological risk

In the present investigation, uranium content of the ground water samples of the Mohali and Fatehgarh districts of North Punjab has been measured and further analysis has been carried out for the excess cancer risk assessment.

The radiological risk has been calculated due to ingestion of natural uranium in the drinking water, 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 $0.02\times10^{-4} - 1.64\times10^{-4}$. The value of the excess cancer risk in the surveyed districts is lower than the maximum acceptable level of 1.67×10^{-4} according to AERB, DAE 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 districts comes out to be 0.48×10^{-4}, which works out to be approximately 1 per 20,000 people.

b) 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 [24]. 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 μg/L. The variations in the values of the LADD and Hazard quotient are observed from 0.04 μg/kg/day – 3.35 μg/kg/day and from 0.01 – 0.74, respectively.

V. Conclusions

- The concentration of the uranium in ground water samples collected from the hand pumps or other ground water sources of several villages of Mohali and Fatehgarh districts is found to be within the safe limit of 60 ppb recommended by AERB, India.
- The cancer risk due to presence of U in groundwater is almost negligible.

- Our investigations establish that uranium content in North Punjab districts is much lower than South Punjab [13, 15].
- If agricultural practices are similar in North and South Punjab, e.g., use of fertilizers and crop pattern etc., then what is the source of Uranium enhancement in Southern districts of Punjab?
- It will be of interest to study nature of aquifers in North and South Punjab based on geological, morphological and hydrogeological investigations.

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Table 1 : Uranium content in the water samples of Mohali and Fatehgarh Sahib Districts and corresponding risk factors

S.No	Location	Water Source	Uranium Concentration (ppb)	Uranium Concentration (Bq l^{-1})	Excess Cancer risk * 10^{-4}	LADD (µg kg^{-1} day^{-1})	Hazard Quotient
	District Mohali						
1	CGC Jhanjheri	Tubewell (T.W.)	10.29	0.26	0.29	0.60	0.13
2	Jhanjheri	Hand Pump (H.P.)	14.39	0.36	0.41	0.83	0.18
3	Landran Gurudwara	T.W./M.P.	24.20	0.61	0.69	1.40	0.31
4	Kargil Park, Sector 71, Mohali	T.W.	12.40	0.31	0.35	0.72	0.16
5	Majat	H.P.	14.82	0.37	0.42	0.86	0.19
6	Bharatpur	T.W.	4.92	0.12	0.14	0.28	0.06
7	Chudiala	H.P.	3.74	0.09	0.11	0.22	0.05
8	Chudiala Sudan	H.P.	7.06	0.18	0.20	0.41	0.09
9	Pattran	M.P.	10.36	0.26	0.29	0.60	0.13
10	Maujpur	H.P.	3.63	0.09	0.10	0.21	0.05
11	Mohali Water Supply	Canal Water	3.26	0.08	0.09	0.19	0.04
12	HS Virk House	RO	0.63	0.02	0.02	0.04	0.01
	District Fatehgarh						
1	SGGS WU Fategarh	T.W.	55.12	1.39	1.56	3.19	0.70
2	BBEC Fatehgarh	Borewell (B.W.)	57.82	1.46	1.64	3.35	0.74
3	Atewali Gurudwara	H.P.	30.57	0.77	0.87	1.77	0.39
4	Kotla Bijwara	T.W.	24.00	0.61	0.68	1.39	0.31
5	Raipur Gujran	T.W.	25.39	0.64	0.72	1.47	0.32
6	Badali Ala Singh	Motor Driven Pump (M.P.)	24.62	0.62	0.70	1.42	0.31
7	Akal Akademi Chuni	T.W.	17.04	0.43	0.48	0.99	0.22
8	Biromajri	H.P.	2.81	0.07	0.08	0.16	0.04
9	Bhagrana	H.P.	6.98	0.18	0.20	0.40	0.09
10	Badali Mai Ki	H.P.	17.22	0.44	0.49	1.00	0.22
11	Badali Mai Ki	RO	BDL	BDL	BDL	BDL	BDL
12	Slaimpur	HP on Canal	20.29	0.51	0.57	1.17	0.26
13	Pola 1	H.P.	18.99	0.48	0.54	1.10	0.24
14	Pola 2	H.P.	15.3	0.39	0.43	0.89	0.20
15	Rajindergarh	H.P.	26.24	0.66	0.74	1.52	0.34
16	Sadugarh	H.P.	6.18	0.16	0.18	0.36	0.08
17	Hansali	H.P.	22.18	0.56	0.63	1.28	0.28
18	Dageri	H.P.	24.26	0.61	0.69	1.40	0.31
19	Hindupur	H.P.	16.76	0.42	0.47	0.97	0.21
20	Panjola	H.P.	18.14	0.46	0.51	1.05	0.23
21	Reona Neevan	RO	2.14	0.05	0.06	0.12	0.03