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By H.S. Virk
SGGS World University

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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 range of uranium content in water in these areas is reported to vary from 0.2 μgl^{-1} to 74.98 μgl^{-1}.The objective of present investigations is health risk assessment due to natural uranium in drinking water in Bathinda district of South Punjab.

Author: Visiting Professor, SGGS World University, Fatehgarh Sahib 416 406, India . e-mail: hardevsingh.virk@gmail.com

II. The Study Area

a) Location

Bathinda district is situated in the southern part of Punjab State. It lies between 29°33´and 30°36´North latitude and 74°38´and 75°46´Eastlongitude.It covers an area of 3367 Sq. Km. The district is surrounded by Sirsa and Fatehabad districts of Haryana State in the south, Sangrur and Mansa districts in the East, Moga in the North-East and Faridkot and Muktsar districts in the North-West.

b) Geomorphology and Soil types

The district area is occupied by Indo-Gangetic alluvium. The soil in the district is mostly sandy. Being sandy, plain region is dotted with scattered sand dunes which have a tendency to shift towards eastern side. But with the development of latest technology and machinery the topography is under vast change with respect to various aspects connected with green revolution. The district has two types of soils, the arid brown soils and siezoram soils. The arid brown soils are calcareous in nature; these soils are imperfectly to moderately drained. Salinity and alkalinity are the principal problems of these soils. In siezoram soils, the

accumulation of calcium carbonate is in amorphous or concretionary form (*kankar*). Presence of high amount of calcium carbonate and poor fertility is the main problem of this soil. The arid brown soils are found in mostly eastern parts of the district and siezoram soils are found in the western part of the district [16].

III. Methodology

a) Pre-processing of the sampling bottles

Washed the container and cap with a dilute solution of detergent and tap water (kept overnight). Rinsed thoroughly with tap water and then rinsed with aqueous 10% nitric acid solution.Drained and completely filled with an aqueous 10% nitric acid solution. After that capped it and stored for at least 24 hours. Then empty the container rinsed with distilled water and kept the carboy in closed condition.

b) Sampling

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.

c) 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 photomultiplier 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.

d) 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.

e) 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.

f) Analytical Procedure

A water sample of quantity 6ml is used to find its uranium content. The water sample is takenin 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:

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

Concentration of uranium in water sample = CF x (Fluorescence from sample – 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.

IV. 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:

a) 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}), 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 as 63.7 years, i.e. 23250 days and daily consumption of water as 4.05 $lday^{-1}$ [18].

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

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

b) 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 substanceingested per kg of body weight per day andis 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(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 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).

c) 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 $kg^{-1}day^{-1}$.

V. Results and Discussion

Groundwater samples were collected from villages around Talwandi Sabo area of Bathinda district of Punjab (India) and analysed for Uranium content using calibrated LED Flourimeter (Quantalase Make). Uranium content varies from 9.72 ppb (Canal water) to 186.61 ppb (Hand Pump). 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]. The proposed guideline of concentration of the uranium in water by EPA from radiochemical toxicity is 30 μgL^{-1}, which indicates that uranium content of natural water is usually not high enough to cause a health risk. In comparison with WHO, UNSCEAR and ICRP safe limits, the values obtained for U in groundwater are significantly higher than the safe limits suggested by these agencies? If the observed data of uranium content of water is compared with the guideline of AERB, 9 water samples exceed the proposed concentration level of 60 ppb.During our survey of March 2012, we reported high uranium content in the same area in the range 69.57 to 205.62 ppb using Laser Flourimetery. Table 1 reports a comparison of both surveys. As sample sites are not exactly the same in the two surveys, some large scale fluctuations in data are observed along with concordance in reported data for Jajjal, Bhagi Vandar and Jagga Ram Tirth.

a) Radiological risk

In the present investigation, uranium content of the ground water samples of the Bathinda district of 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 $0.28\times10^{-4} - 5.29\times10^{-4}$. The value of the excess cancer risk in the surveyed district is approximately higher 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 district comes out to be about 2 per 10,000 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 0.56 μg/kg/day –10.80 μg/kg/day and from 0.12 – 2.38, respectively.

The concentration of the uranium is found to be high in ground water samples collected from the hand pumps or other ground water sources of several villages of Bathinda district. The high uranium concentrations found in this investigation support the earlier reported measured values[13,15]. What is the source of high U content in groundwater? It is as yet an open question begging for answers. However, some authors have

offered plausible solutions as follows: (i) the presence of high uranium concentration in the ground water samples of the southwest Punjab has been attributed to the high uranium content in granitic rocks of Tosham hills in the neighbouring Haryana state [25]; (ii)the use of phosphate fertilizers in the fields of selected villages of Bathinda district is linked to the enhanced uranium concentration in the ground water [26]. The water samples containing the high carbonate or phosphate concentration enhances the dissolved uranium content of the water, as it has been established that uranium forms complexes with the carbonate and phosphate ions. Uranium concentration would be lower in the water samples having low carbonate or phosphate levels[27].

Our investigation reveals that the mean value of excess cancer risk in the surveyed district comes out to be about 2 per 10,000 people. The maximum value of excess cancer risk for residents of Kot Shamir is estimated to be 5 per 10,000 persons. However, this area is reported to be in grip of cancer disease at an alarming rate, especially villages falling in Talwandi Sabo block of Bathinda district. Our study shows that U in groundwater is not the sole cause of cancer induction; one has to look for other causes, e.g., use of pesticides and an overdose of fertilizers, by taking recourse to epidemiological investigations.

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Table 1: Total Uranium content in the water samples of Bathinda district and corresponding risk factors

S.No	Location	Water Source	Year 2012	Year 2016	Uranium Concentration (Bq l^{-1})	Excess Cancer risk * 10^{-4}	LADD ($\mu g\ kg^{-1}\ day^{-1}$)	Hazard Quotient
			Uranium Concentration (ppb)					
1	Jajjal	Hand Pump (HP)	76.37	61.62	1.56	1.75	3.57	0.79
2	Jajjal	Motor Driven Pump (MP)	96.10	24.98	0.63	0.71	1.45	0.32
3	Malkana	HP	109.07	35.85	0.91	1.02	2.07	0.46
4	Giana	HP	-	69.57	1.76	1.97	4.03	0.89
5	Giana	Canal Water	-	9.72	0.25	0.28	0.56	0.12
6	Giana	Tubewell	205.62	92.96	2.35	2.63	5.38	1.19
7	Takhatmal	HP	-	96.90	2.45	2.74	5.61	1.24
8	Sangat	HP	-	140.10	3.54	3.97	8.11	1.79
9	Kot Shamir	HP	86.27	186.61	4.72	5.29	10.80	2.38
10	Bhagi Vandar	HP	129.45	139.86	3.54	3.96	8.09	1.79
11	Talwandi Sabo	HP	117.82	24.24	0.61	0.69	1.40	0.31
12	Jagga Ram Tirth	HP	82.85	77.55	1.96	2.20	4.49	0.99
13	Jiwan Singh Wala	HP	120.07	67.18	1.70	1.90	3.89	0.86
14	Raman Village	Tubewell	-	55.64	1.41	1.58	3.22	0.71
15	Raman Village	MP	-	34.51	0.87	0.98	2.00	0.44
16	Akal University Talwandi Sabo	MP	-	44.09	1.11	1.25	2.55	0.56

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