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# Estimation of Uranium and Related Health Risks Due to Consumption of Groundwater in Lower Himalayas

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Exposure to uranium via ingestion of edible products may lead to serious health hazards when taken in quantities more than recommended limit. Hence, to assess the uranium content in groundwater and concerned health hazards 64 groundwater samples were collected from Hamirpur and Mandi districts of Himachal Pradesh. The samples were collected in pre monsoon season from the handpumps and bowries. The region lies in Lower Himalayan range which is storehouse of various granatic rocks. Presence of uranium deposits in Tileli (Mandi), Rajpura (Una), Lambehra (Hamirpur) makes the area more vulnerable for the study. The groundwater samples were analysed to measure concentration of uranium using LED Fluorimeter developed by Quantalase Private. Limited. The uranium concentration in groundwater samples varied from 0.25 to 17.29 μg $L^{-1}$, with an average value of 1.97. Uranium concentration in none of the samples surpassed the limit of 30μg $L^{-1}$ recommended by WHO(2011), 60μg $L^{-1}$ set by AERB(2004). Health risks were estimated in terms radiological and chemical toxicity for different isotopes of uranium. The calculated average mortality and morbidity risks were lower than the actual prescribed limit. The average Lifetime Average Daily Dose (LADD) was calculated as 0.04 and Hazard Quotient (HQ) below unity. Annual ingestion doses for different age groups were also measured which lies under safe limit. Thus, it is recommended that the groundwater is safe for consumption by public. Using Hair Compartment Model for uranium and mean daily uranium intake of 2.71μg for 60-year exposure period, organ specific doses due to uranium radioisotopes in prime organs/tissues and excretion rates via urine, faeces and hair pathway are estimated.

**Keywords:** Chemical Toxicity; Hair Compartment Model; Hazard Quotient; LED Fluorimeter; Lifetime Average Daily Dose; Radiological toxicity

## 1 Introduction

Uranium is a weakly radioactive naturally occurring element found in many minerals and ores and used in nuclear power stations as fuel, some compounds of uranium are used as catalysts and staining pigments[1]. Acidic magmatic rocks, ores like uraninite, monazite, zircon are some storehouse of uranium[2]. Different agencies, research institutions have reported uranium concentrations in soils, plants, rocks, water, food items etc. from time to time. Apart from natural resources anthropogenic activities, industries, agricultural practices also results in elevation of uranium concentration[3,4]. High uranium concentration has been reported in certain parts of Northern India and in different regions across world[5-16]. Uranium is versatile in terms of chemical properties as it is in its physical properties. It shows reaction with almost all non-metals and their compounds. Since uranium is radioactive metal and unstable, so it undergoes decay releasing alpha particles, beta particles and gamma radiations until a stable product is formed. This property of uranium makes it valuable asset for the development of nuclear arms and weapons. $^{238}U$ is not highly fissile like $^{235}U$ but it undergoes fission by the collision of fast neutrons. Uranium with oxidation state +6 is highly soluble along with carbonate complexes at alkaline level of pH which is also a reason of availability of uranium in ground water. As water is continuously in transition on surface as well as below the surface of earth through the rocks, the water movement through the aquifers and rocks having uranium deposits initiates leaching of uranium. This uranium rich water gets mixed with water table and when ground water without filtering is ingested by people via handpumps, tube-wells or dug wells then they get exposed to uranium. In case of acute exposure headache, vomiting, radiation syndrome occurs. Blood chemistry changes, hair loss, skin burn occur if exposure is long term and quite high dose is received.

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DNA gets damaged when a person is highly exposed. Imbalance in menstrual cycle of women, reduction in sperm activity of men, issues related to growth of child in pregnant women occur. Nephritis is recently observed disease can be caused due to uranium exposure. Assimilation of uranium for a long time after ingestion can result in hazardous impacts in normal functioning of body. In technical terms it is amount of dose received by the body which tells about the severeness of exposure and its results. The decay products when mixes in bloodstream changes the whole chemistry of blood. In estimation of doses received by various body organs/tissues bio kinetic models play crucial role. Bio kinetic models are mathematical tools for comprehensive assessment of retention of various radio elements as well as the calculation of dose received by different tissues and organs. These help researchers to foresee and assess the transfer, distribution, retention and elimination of substances within the body. The kinetics of a substance inside body depends on various factors like age, structure of body etc. Similarly, biokinetics of uranium are affected by various factors such as physical characteristics of the person, age, kind of diet he takes, routine physical workout and exercises, medical history. These models also depends on environmental factors for instance temperature, sweating, moisture, humidity etc.. Over the years many researches have been conducted on animals and humans by taking various specimen of their body fluids, excreta and injecting them with certain doses so that they become exposed to uranium and corresponding health effects can be studied. Ultimately researchers came up with some mathematical formulations and proposed various biokinetic models.[14-20]

## 2 Materials and Methods

### 2.1 Study area

Topographically, the Himalayan region of has been divided into three categories: Greater Himalayas, Inner Himalayas, Shiwaliks. The study area falls in the Shiwalik region and Inner Himalayas, generally called Lower Himalayan region. Depending on altitude, the climate of the state varies from semi-tropical to semi-arctic. Hamirpur is surrounded by Kangra in North, Mandi in East and Una in West, Bilaspur in South and extends from 76°17’50’’E to 76°43’42’’E longitude and 31º24’48’’N to 31º53’35’’N latitude covering 1,118 sq. km area while Mandi region is centrally located in hilly area of Himachal Pradesh extending from 31°13’26’’ to 32°04’22’’ latitude and 76°36’08’’ to 70°23’36’’E longitude covering 3950 sq. km. area. Jutog group, Shah group, Chail group, Tertiary group of rocks, Graneticrocks, rocks of Dharamshala formation, Sundru formation of Vaikarta group are present in Mandi. Mandirocks belong from Pre-Cambrian to quaternary period. The region includes various thrusts and fault systems which formed due to collision of Indian and Eurasian converging plates. The study area is rich in slates, schists, quartzite. Kullu group is rich in carbon aceous slate, phyllite, quartzite, calcite, schist. Vaikarta groupis rich in dark grey slate, micaceouss and stone, in carbon aceous slate, phylite, quartzite, calcite, schist, shales[21-26].

### 2.2 Sample collection

37 groundwater samples from Mandi and 27 groundwater samples from Hamirpur were collected from handpumps, bowries. Bowries are natural sources of water flowing from inside of mountains in hilly areas. All the samples were collected in pre-monsoon season. Before collecting the samples, the source was made to run water for 4-5 minutes so that fresh groundwater can be obtained. All the water samples were collected in scientific graded Tarson Bottles and were analysed within 10 days of collection. The study area and location of sample sites is shown in Fig. 1.

### 2.3 Measurements

The uranium concentration in collected groundwater samples has been estimated using an LED Fluorimeter. It consists of a pulsed UV LED source. This instrument works on principle of fluorescence of uranyl compounds present in the sample. The instrument has been calibrated using standard solution of uranium oxide (ICP-MS-66N-0.01X-1). Buffer solution of sodium pyrophosphate was used as fluorescent enhancing species. For each measurement 6ml sample along with 10% of buffer solution was used.

## 3. Results and Discussions

### 3.1 Uranium concentration in groundwater

The concentration of uranium in groundwater samples of Mandi and Hamirpur district lied between 0.25 ppb to 17.92 ppb with a mean and median of 1.97 ppb and 1.03 ppb respectively. The calculated value of

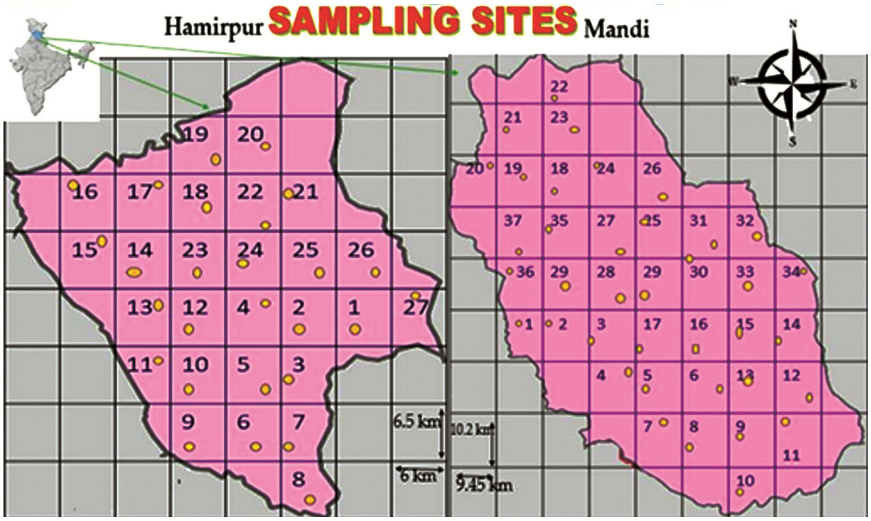


Fig. 1 — Map of Study Area

Table 1 — Statistical analysis of radiological and chemical toxicity in groundwater samples.

| | | Minimum | Maximum | Mean | Standard deviation |
|---|---|---|---|---|---|
| Uranium concentration (μg $L^{-1}$) | | 0.25 | 17.92 | 1.97 | 2.84 |
| Uranium Activity (Bq $L^{-1}$) | | 0.01 | 0.45 | 0.05 | 0.07 |
| Excess Cancer Risk (Mortality) | $^{234}U$ | $6.71 \times 10^{-13}$ | $4.81 \times 10^{-11}$ | $5.30\times 10^{-12}$ | $7.63\times 10^{-12}$ |
| | $^{235}U$ | $9.82 \times 10^{-11}$ | $7.04 \times 10^{-09}$ | $7.75\times 10^{-10}$ | $1.12\times 10^{-09}$ |
| | $^{238}U$ | $1.64 \times 10^{-08}$ | $1.17\times 10^{-06}$ | $1.20 \times 10^{-07}$ | $1.86\times 10^{-07}$ |
| Excess cancer risk (Morbidity) | $^{234}U$ | $1.05 \times 10^{-12}$ | $7.49 \times 10^{-11}$ | $8.25 \times 10^{-12}$ | $1.19 \times 10^{-11}$ |
| | $^{235}U$ | $1.55 \times 10^{-10}$ | $1.11\times 10^{-08}$ | $1.23\times 10^{-09}$ | $1.77\times 10^{-09}$ |
| | $^{238}U$ | $1.64 \times 10^{-08}$ | $1.17 \times 10^{-06}$ | $1.29\times 10^{-07}$ | $1.86\times 10^{-07}$ |
| LADD (μg $kg^{-1}$ $day^{-1}$) | | 0.01 | 0.36 | 0.04 | 0.06 |
| HQ | | 0 | 0.30 | 0.03 | 0.05 |

first and third quartile were 0.51 and 2.46 ppb respectively. 95 percentile and 99 percentiles signified that 95% and 99% percent values of uranium concentration lied below 6.17 ppb and 14.34 ppb respectively. The skewness and kurtosis were measured to be 17.44 and 3.48 respectively. In Mandi district highest uranium concentration (7.06 ppb) was found at Dhalwan while Ladraur recorded highest uranium concentration of 17.92 ppb in Hamirpur district. The concentration of uranium groundwater samples lied below the prescribed limit of 30μg $L^{-1}$ (WHO)[27] and 60μg $L^{-1}$(AERB)[28]. In Mandi 9 samples while in Hamirpur district 1 sample had uranium concentration below detection limit of LED Fluorimeter. Using USEPA 2006 convention that "if less than 15% samples lie below detection limit then those values are replaced by factor obtained after dividing the minimum detection limit of instrument by 2".The BDL values of uranium concentration were replaced by 0.25 ppb.

### 3.2 Uranium activity

It varied in the range 0.01 to 0.45 Bq $L^{-1}$ with mean value of 0.05 Bq $L^{-1}$. Uranium activity was also measured for different isotopes of uranium. The average value of uranium activity for $^{234}U$, $^{235}U$, $^{238}U$ was calculated to be $2.47\times10^{-6}$, $3.55\times10^{-4}$, $4.9\times10^{-2}$Bq $L^{-1}$.

### 3.3 Toxicity

Toxicity is measure of how harmful a substance can be for living beings. Hazards due to uranium exposure and ingestion has been calculated in terms of radiological and chemical toxicity. Uranium toxicity depends on environment in which human is living, age, gender, health, body structure, medical history and route of ingestion *etc*. Table 1. shows statistical analysis of radiological and chemical toxicity in groundwater samples.

### 3.4 Radiological Toxicity

The radiological toxicity has been measured in terms of excess cancer risk. This has been measured using method proposed by US Environmental Protection Agency.[29] The radiological toxicity accounts for the harmful impact son the body after exposure to uranium progenies.

#### *3.4.1 Excess Cancer Risk(ECR)*

It was calculated using following formula

$$\boldsymbol{ECR = A \times R}$$

Where A is activity concentration and R is risk factor. Further risk factor has been calculated using

$$\boldsymbol{R = r \times I}$$

Where r is risk co-efficient and I is per capita intake. Following relation has been used to calculate per capita intake

$$\boldsymbol{I = l \times d}$$

Where l is life expectancy and d is daily water intake.

ECR has been calculated for all three isotopes of uranium using different risk co-efficient (Table 2) for these isotopes.

The measured mean values of excess cancer for mortality and morbidity for $^{234}U$, $^{235}U$, $^{238}U$ are $5.30\times10^{-12}$, $7.75\times10^{-10}$, $1.29\times10^{-7}$ and $8.25\times10^{-12}$, $1.25\times10^{-9}$, $1.29\times10^{-7}$ respectively which is well below the prescribed limit of $10^{-3}$.[30].

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### 3.5 Chemical Toxicity

#### *3.5.1 Life time Average Daily Dose*

LADD is measure of intake of dose of a substance averaged over lifetime of human. It is expressed in terms of μg $kg^{-1}day^{-1}$. It has been measured using

$$\boldsymbol{LADD = \frac{EPC \times IR \times IF \times D}{AT \times W}}$$

Where EPC is exposure point concentration, IR is ingestion rate, IF is ingestion frequency, D is daily water intake, AT is averaged time, and W is body weight. LADD has been lying between 0.01 and 0.36 with an average value of 0.04 and lies below there commended limit of WHO 2011[27].

#### *3.5.2 Hazard Quotient*

Hazard quotient is ratio of potential exposure of a substance to the level at which no health hazards are observed. It is ratio of LADD to reference

$$\boldsymbol{HQ = \frac{LADD}{RFD}}$$

Where RFD is reference dose prescribed by WHO (2011)[27]. It is taken as 1.2μg $kg^{-1}$ $day^{-1}$. The average HQ for the study is 0.03.

### 3.6 Annual ingestion doses

Annual ingestion doses for various age groups has been calculated on the basis of their different water intakes using following equation.

$$\boldsymbol{Annual\ Ingestion\ Dose = A \times WI \times DCF}$$

Where A is activity concentration, WI is water intake, DCF is dose conversion factor.[20,31]. These signifies the amount of dose received by individual falling into different age group annually. The dose received by the individual is compared to there commended dose given by various agencies and according to that health hazards are measured. Annual ingestion doses differ for different geographical areas depending upon exposure pathway and amount of uranium and its progenies present at that place. Annual ingestion doses for different age groups are shown in Table 3.

Table2 — Risk coefficients for excess cancer risk.

| Isotope | Mortality ($Bq^{-1}$) | Morbidity ($Bq^{-1}$) |
|---|---|---|
| U-234 | $6.2 \times 10^{-11}$ | $9.5 \times 10^{-11}$ |
| U-235 | $6.32 \times 10^{-11}$ | $9.8 \times 10^{-11}$ |
| U-238 | $7.5 \times 10^{-11}$ | $7.5 \times 10^{-10}$ |

**Physicochemical Parameters and Pearson correlation coefficients**

Various physicochemical parameters of water samples have been measured and an attempt to correlate the dependency of uranium concentration on

Table 3 — Annual ingestion dose for various age-groups (μSv $year^{-1}$).

| | Age Group | Minimum | Maximum | Average | Standard Deviation |
|---|---|---|---|---|---|
| Infants | 0-6 months | 0.54 | 38.98 | 4.29 | 6.18 |
| | 6-12 months | 0.62 | 44.48 | 4.90 | 7.06 |
| Children | 1-3 years | 0.36 | 25.51 | 2.81 | 4.05 |
| | 4-8 years | 0.31 | 22.24 | 2.45 | 3.53 |
| Males | 9-13 years | 0.37 | 26.69 | 2.94 | 4.23 |
| | 14-18 years | 0.50 | 36.15 | 3.98 | 5.74 |
| | >18 years | 0.11 | 7.87 | 0.87 | 1.25 |
| Females | 9-13 years | 0.33 | 23.35 | 2.57 | 3.71 |
| | 14-18 years | 0.35 | 25.20 | 2.78 | 4.00 |
| | >18 years | 0.08 | 5.74 | 0.63 | 0.91 |
| Pregnancy | | 0.09 | 6.38 | 0.70 | 1.01 |
| Lactation | | 0.11 | 8.08 | 0.89 | 1.28 |

Table 4 — Pearson correlation matrix between Uranium and various Physicochemical parameters.

| | pH | EC | TDS | ORP | Salinity | DO | U |
|---|---|---|---|---|---|---|---|
| pH | 1.00 | | | | | | |
| EC | -0.01 | 1.00 | | | | | |
| TDS | -0.10 | 0.59 | 1.00 | | | | |
| ORP | -0.82 | 0.15 | 0.14 | 1.00 | | | |
| Salinity | -0.03 | 0.94 | 0.51 | 0.13 | 1.00 | | |
| DO | 0.21 | 0.14 | 0.20 | -0.15 | 0.14 | 1.00 | |
| U | -0.12 | 0.21 | 0.38 | 0.14 | 0.20 | 0.27 | 1 |

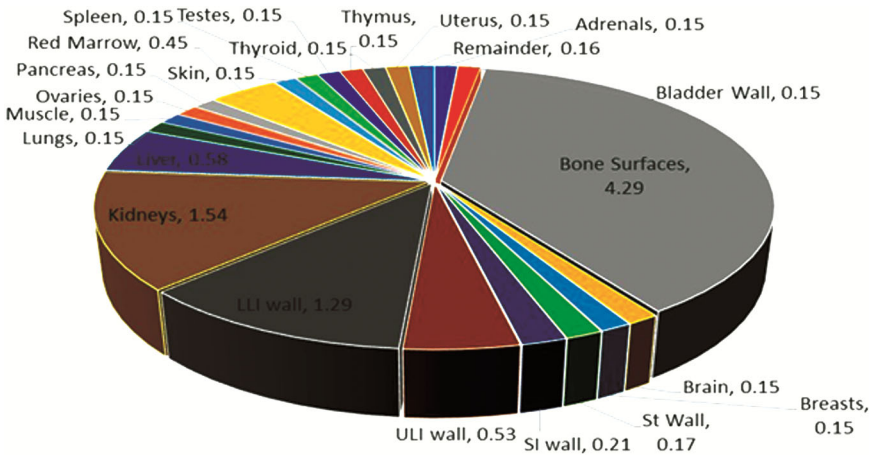


Fig. 2 — Pie chart showing organ specific doses received by various tissues/organ

these physicochemical parameters has been made. Table 4 shows the pearson correlation matrix with different physicochemical parameters. No strong correlation among uranium concentration and any of physicochemical parameters been found in the study region.

### 3.7 Organ specific doses

Organ specific doses has been calculated using Hair compartmentmodel[8]. Doses received by different organs/tissues have been shown in Fig. 2. Bones are most vulnerable sites receiving highest amount of dose of 4.29μSv. After bone surfaces kidneys and lower large intestine receives high doses of uranium making them more prone toil effects due to accumulation and de composition of uranium.

## 4 Conclusions

Uranium concentration in all the samples lied below there commended value of 30μg $L^{-1}$given by USEPA and WHO, 2011 and 60μg $L^{-1}$ (AERB 2004). The calculated radiological toxicity, chemical toxicity, annual ingestion doses are all within prescribed limits. All doses are below the permissible limit 100μ $Svy^{-1}$ recommended by WHO, 2004. Organ-specific doses for different organs/tissues showed that bone surfaces are highest dose receiving organs followed by kidneys and LLI wall and are more prone to hazards caused by uranium.

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