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RISK ASSESSMENT OF ^{238}U AND ^{232}Th IN GROUNDWATER IN BATHINDA DISTRICT OF PUNJAB STATE, INDIA

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ABSTRACT

High resolution inductively coupled plasma mass spectrometry (HR-ICP-MS) technique has been used for the microanalysis of ^{238}U and ^{232}Th concentrations in groundwater samples collected from Bathinda district of Punjab State, India. ^{238}U and ^{232}Th concentrations ranged from 4 to 163 µg l^{-1} and 0.41 to 1.79 µg l^{-1} with mean values of 65 µg l^{-1} and 0.93 µg l^{-1}, respectively. ^{238}U concentration in water samples collected from villages exceeded the permissible limit of 30 µg l^{-1} prescribed by World Health Organization and United States Environmental Protection Agency, while the samples collected from towns have ^{238}U concentration well within the permissible limit. The estimated annual effective doses due to intake of ^{238}U and ^{232}Th through drinking water for various age groups ranged from 2 to 201 µSv y^{-1} and 0.4 to 9.8 µSv y^{-1}, respectively. On the radiological aspect, the average cancer mortality and morbidity risks for ^{238}U were found to be 4.6×10^{-5} and 7.1×10^{-5} and for ^{232}Th, 3.6×10^{-7} and 5.3×10^{-7}, respectively, which are lower than the acceptable level. About 80% of the analyzed samples showed hazard quotient greater than unity indicating significant risk due to chemical toxicity of ^{238}U in groundwater. ^{238}U and ^{232}Th concentrations were found to be correlated with the physicochemical properties of the water samples.

Keywords: Annual Effective Dose, Groundwater, HR-ICP-MS, ^{232}Th, ^{238}U.

1. INTRODUCTION

Uranium and thorium are natural lithophilic elements and are contained almost in all natural soils and waters; however, their concentration varies from water to water depending upon their origin. The geochemical processes, geographical location and geological setting influence the concentrations of uranium and thorium in the environment. Uranium and thorium have both radiological and chemical toxicity with two important target organs being the kidneys and lungs [1-3]. Cothern and Lappenbusch have estimated that the drinking water contributes about 85% of ingested uranium while food contributes remaining 15% [4].

In the Malwa region of Punjab, the problem of uranium contamination in groundwater has become very acute. Uranium in drinking water can be extremely dangerous because it becomes part of the entire ecological system of the Malwa region of Punjab [5,6]. Being a prominent agriculture based state; the Malwa region of Punjab is facing an unprecedented crisis of environmental health linked to indiscriminate, excessive, and unsafe use of fertilizers and poor groundwater quality [7]. The Malwa belt has been described as India's "Cancer Capital". The cancer prevalence in Malwa region is indicated to be 1089/million/year, which is much higher than the national average cancer prevalence in India (800/million/year) [8].

This study was undertaken to estimate ^{238}U and ^{232}Th concentrations in groundwater, to compute annual effective doses for different age groups, and to assess radiological and chemical risks to humans due to intake of ^{238}U and ^{232}Th through drinking water from Bathinda district of Punjab. Other water quality parameters viz. pH, total dissolved solids and electrical conductivity have been mentioned in the groundwater.

2. EXPERIMENTAL

2.1 Study Area

Punjab state is located between 29º30´ and 32º32´ North latitudes and 73º55´ and 76º50´ East longitudes in the North India. Fig. 1 shows the geographic location of Bathinda district on the map of Punjab, as well as the location of the sampling sites. Bathinda district is located between 29º33´ and 30º36´ North latitudes and 74º38´ and 75º46´ East longitudes in the south west region of Punjab state. The total human population of the Bathinda district is 1,388,859 (2011 census), and geographical area of 3344 km^2. The normal annual rainfall of the district is 450 mm. The water table elevation in the district varies between 197.5 and 220.6 m above mean sea level. The mean minimum and maximum temperature is 3.9 ºC (January) and 42 ºC (May-June) respectively.

The study region forms a part of the Indo-Gangetic alluvial plains. The study region has both unconfined and confined aquifers. In alluvium thin granular zones exist down to a depth of 450 m. The top aquifer ranges from 40 to 58 m. The depth of the top aquifer in the north is upto 56 m, in the south it is upto 58 m, in the east it is 38 m and in the west it is 40 m. The normal annual rainfall of the district is 450 mm. The general slope of the water table is towards SW from North, NE, East and SE. No important hill exists in the district. The study region is surrounded by Faridkot and Muktsar districts in the north-west, Moga district in the north-east, Sirsa district of Haryana in the south and Sangrur and Mansa districts in the east [9-10].

2.2 Water Sampling and Physico-chemical Analysis

Groundwater samples were collected from 15 sampling sites in the Bathinda district of Punjab from manually operated hand-pumps established by municipal corporations in residential localities or from privately owned hand-pumps. The sampling sites were chosen in such a manner that whole geographical area get covered. Groundwater was pumped for at least 10 minutes before sampling procedures to remove stagnant groundwater from the well. The weather conditions during the sampling period were fairly stable. Prior to collection, the water samples were filtered using 0.45 µm Millipore nitrocellulose filters (disposable not reusable) to remove suspended matter/sediments, acidified to pH < 2 (0.2% v/v) using supra pure nitric acid (HNO_3) for preservation and then stored in pre-cleaned acid-washed high density polyethylene (HDPE) containers until analysis. On-site observations like location, source and depth of the hand-pumps were recorded. Water pH, electrical conductivity (EC), total dissolved solids (TDS) and temperature were analyzed in situ with the help of portable micro-controller water analysis kit (NPC 362D, Naina Solaris Limited, India).

2.3 Analytical Procedure

^{238}U and ^{232}Th concentrations in water samples were determined from acidified, filtered and diluted solutions after adjusting the total dissolved solids to lower levels recommended for high resolution instruments. The clear solutions were analyzed at CSIR-NGRI, Hyderabad, using high resolution inductively coupled plasma mass spectrometer (HR-ICP-MS) (Nu Instruments Attom®, UK) in jump-wiggle mode which permits the analytes of interest (viz., ^{238}U, ^{232}Th) to be measured accurately. The samples introduction consisted of a standard Meinhard® nebulizer with a cyclonic spray chamber housed in Peltier cooling system. All quantitative measurements were performed using the instrument software (Attolab v.1), while the data processing was done using Nu Quant®, which uses knowledge-driven routines in combination with numerical calculations (quantitative analysis) to perform an automated/manual interpretation of the spectrum of interest. Instrumental parameters are given in Table 1. Instrument was optimized using 1 µg l^{-1} tuning solution and the sensitivity of ^{114}In was about 1 million cps. Oxide and oxy-hydroxide ratios were low (< 0.2%) and the double charges ions ratio was < 3%. Mass bias fractionation and several well-known isobaric interferences were addresses by using certified geochemical reference materials. External drift was corrected by repeated analyses of a NIST1640a (NIST, USA). Instrument response was corroborated relative to two analyses of NIST1643e and NIST1640a (NIST, USA). Precision and accuracy are better than RSD 3% for the majority of trace elements. Details of analytical procedure and instrumental operating conditions of HR-ICP-MS are given elsewhere [11].

2.4 Theoretical Calculations

2.4.1 Annual effective dose

Annual radiation dose to human (for different age groups) due to ^{238}U and ^{232}Th consumption was quantified using Eq. (1) [11-12]:

$$D = AC \times DWI \times 365 \times DCF \quad (1)$$

Where

D = uranium effective dose per year for specific age group (µSv y^{-1});

AC = activity concentration of radionuclide (Bq l^{-1});

DWI = daily water intake for specific age group (l day^{-1}); and

DCF = dose conversion factor for the specific age group (Sv Bq^{-1}).

^{238}U and ^{232}Th activity concentrations were calculated by using unit conversion factors of 1 µg l^{-1} = 0.0124 Bq l^{-1} and 1 µg l^{-1} = 0.00406 Bq l^{-1}, respectively [11]. The ingestion dose conversion factors applied are from the

International Atomic Energy Agency [13] and the age-dependent daily water intake is prescribed by Dietary Reference Intakes, Food and Nutrition Board, Institute of Medicine, US National Academy of Science [14].

2.4.2 Radiological risk

The radiological risk and total radionuclide intake were calculated by using following relations (2 & 3) [15].

$$\text{Lifetime cancer risk} = \text{total radionuclide intake (Bq)} \times \text{risk coefficient } (\text{Bq}^{-1}) \quad (2)$$

$$\text{Total radionuclide intake over a lifetime} = \text{AC} \times \text{IR} \times \text{EF} \times \text{ED} \quad (3)$$

Where AC is activity concentration of radionuclide in drinking water (Bq l^{-1}); ED is the lifetime exposure duration (70 y); EF is the exposure frequency (365.25 days y^{-1}) and IR is the water ingestion rate (2 L day^{-1}) [16-17].

According to USEPA [15], the cancer morbidity and mortality risk coefficients of 2.73×10^{-9} and 1.87×10^{-9} for ^{232}Th and 1.73×10^{-9} and 1.13×10^{-9} for ^{238}U have been used, respectively.

2.4.3 Chemical toxicity risk

The chemical toxicity risk was evaluated in terms of lifetime average daily dose (LADD) using Eq. (4) and compared with the reference dose (RfD) which has been calculated on the basis of the permissible limits of ^{238}U and ^{232}Th in drinking water [18].

$$D = \frac{C \times IR \times EF \times ED}{AT \times BW} \quad (4)$$

Where LADD is lifetime average daily dose (μg kg^{-1} day^{-1}); C is the radionuclide concentration (μg l^{-1}); AT is average exposure time for non-carcinogens (365.25 × 70 days) and BW is body weight (70 kg) [19-20].

The chemical risk has been calculated in terms of hazard quotient (HQ) (Eq. 5).

$$HQ = LADD/RfD \quad (5)$$

If HQ > 1, then the LADD of radionuclide exceeds the RFD, indicating that there is a potential risk associated with radionuclide.

3. RESULTS AND DISCUSSION

3.1 Distribution of ^{238}U and ^{232}Th in Groundwater

^{238}U and ^{232}Th concentrations in groundwater together with depth, location and physicochemical analyses data are presented in Table 2. ^{238}U concentration ranged from 4 to 163 μg l^{-1} with a mean value of 65 μg l^{-1}. The measured ^{238}U concentration in all the water samples collected from villages exceeded the permissible limit of 30 μg l^{-1} recommended by United States Environmental Protection Agency [16] and World Health Organization [2] and 58% of the samples exceeded 60 μg l^{-1} Indian maximum acceptable concentration prescribed by the Atomic Energy Regulatory Board, India [21], while the samples collected from the Bathinda, Rampura Phul and Talwandi Sabo towns have ^{238}U concentration well within the permissible limits. The concentration of ^{232}Th ranged from 0.41 to 1.79 μg l^{-1} with a mean value of 0.93 μg l^{-1}. ^{232}Th concentration in both villages and towns were well within the permissible limit of 137 μg l^{-1} recommended by USEPA [1]. Table 3 presents the summary statistics of ^{238}U and ^{232}Th in groundwater. The negative values of kurtosis indicate that the distributions have lighter tails and a flatter peak than the normal distribution. The skewness values are of positive type. The data is moderately skewed. It can be observed from Table 3 that the arithmetic mean is greater than the standard deviation.

From Table 2, it is evident that at all the sampling sites, the ^{232}Th concentration is much lower than that of ^{238}U, because the ^{232}Th has low mobility under all environmental conditions, mainly due to the high stability of the insoluble ThO_2. Unlike ^{238}U, ^{232}Th cannot be oxidized to a stable cation equivalent to the highly mobile uranyl ion $[UO2]^{2+}$ [22]. The negative correlation between ^{238}U and depth of groundwater (r = -0.47), indicating that drilling deeper hand-pumps can assess groundwater with significantly lower ^{238}U concentration.

3.2 Physicochemical Analysis

Water pH ranged between 7.3 and 8.0. The salinity, EC and TDS of the water samples ranged from 0.1 to 0.8 ppt, 141 to 1456 µS cm^{-1} and 69 to 716 mg l^{-1}, respectively. All the samples have pH, TDS and EC within the permissible limits set by the USEPA [16], WHO [2], and Bureau of Indian Standards [23]. ^{238}U and ^{232}Th concentrations were found to be correlated with EC, TDS and salinity. The strong positive correlations were observed between ^{238}U and TDS (r = 0.58) and ^{232}Th and TDS (r = 0.70). The fact that higher the TDS and EC values are related to higher the radioactivity in water holds good in this case [24-25]. The strong correlation of ^{238}U and ^{232}Th with TDS and EC suggest that these parameters influence and control the mobility of ^{238}U and ^{232}Th in the groundwater.

3.3 Age-Dependent Annual Effective Dose

The impact of ^{238}U and ^{232}Th derived from drinking water can be assessed by determining the effective dose to each age group in population. The results are presented in Table 4. The recommended level of annual effective dose to humans from water consumption is 100 µSv y^{-1} [2,26]. This reference dose level represents approximately 4.2% of the average annual effective dose from natural background radiation (2.4 mSv y^{-1}) [2]. The data showed significant variations in the dose rate for different age groups. This is due to different dose coefficient and water consumption rate for different age groups. Even though infants drinking less water than adults, the annual effective doses to infants are significantly higher than that to adults, because of the differences in infants metabolism and smaller organ weights resulting in higher doses for many radionuclides. Females receive higher doses during pregnancy and lactation due to increased water consumption. The mean annual effective dose is higher in the age group of infants 7-12 month old, due to higher annual water intake (292 L) compared with the 0-6 months group (256 L). The estimated annual effective doses due to intake of ^{238}U and ^{232}Th through drinking water for various age groups ranged from 2 to 201 µSv y^{-1} and 0.4 to 9.8 µSv y^{-1} with mean value of 53 and 1.5 µSv y^{-1}, respectively.

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3.4 Radiological Risk

The cancer mortality and morbidity risks for ^{232}Th varied from 1.6×10^{-7} to 6.9×10^{-7} and 2.3×10^{-7} to 1.0×10^{-6} with average values of 3.6×10^{-7} and 5.3×10^{-7}, respectively and for ^{238}U varied from 2.9×10^{-6} to 1.2×10^{-4} and 4.4×10^{-6} to 1.8×10^{-4} with average values of 4.6×10^{-5} and 7.1×10^{-5}, respectively (Table 5). The mean radiological risk values are comparable to those reported from SW-Punjab, India (2.1×10^{-4}) by Bajwa et al. [6], Western Haryana, India (1.10×10^{-4}) by Duggal et al. [27], Northern Rajasthan, India (5.6×10^{-5}) by Duggal et al [20].

3.5 Chemical Toxicity Risk

The LADD of ^{238}U and ^{232}Th due to consumption of groundwater varied from 0.11 to 4.66 μg kg^{-1} d^{-1} and 1.2×10^{-2} to 5.1×10^{-2} μg kg^{-1} d^{-1} with average values of 1.85 μg kg^{-1} d^{-1} and 2.7×10^{-2} μg kg^{-1} d^{-1}, respectively. The HQ was calculated for ^{238}U using reference dose (RfD) recommended by AERB [21] and WHO [2], i.e. 1.714 μg kg^{-1} d^{-1} and 0.857 μg kg^{-1} d^{-1}, respectively. According to AERB and WHO standards, the HQ varied from 0.07 to 2.72 and 0.13 to 5.43, respectively. According to AERB and WHO standards, 47% and 80% samples showed HQ > 1, respectively indicating significant risk due to chemical toxicity of ^{238}U. For ^{232}Th, the HQ was calculated using RfD recommended by USEPA (3.91 μg kg^{-1} d^{-1}) [1] According to USEPA standards, HQ for all samples was less than unity, indicating negligible risk due to chemical toxicity of ^{232}Th.

4. CONCLUSIONS

High ^{238}U concentration observed in water samples collected from villages may be due to local natural geology and use of phosphate fertilizers in huge quantity for agriculture purpose and low ^{238}U concentration observed in water samples collected from Bathinda, Rampura Phul and Talwandi Sabo towns indicating that coal-fired power plants (CFPPs) (Guru Nanak Dev Thermal Plant, Bathinda; Guru HarGobind Thermal Plant, Lehra Mohabbat; and Talwandi Sabo Power Limited), national fertilizer limited (NFL), cement factories, chemical factories, Guru Gobind Singh oil refinery, anthropogenic activities and urbanization may not be responsible for ^{238}U contamination. The age-dependent average annual effective dose due to groundwater consumption is lower than the WHO and EU Council recommended level. The most crucial age groups are small babies, below 1 year and lactating females within 14-18 years. It is concluded that there is negligible carcinogenic risk to humans but non-carcinogenic health risks may be due to chemical toxicity of ^{238}U.

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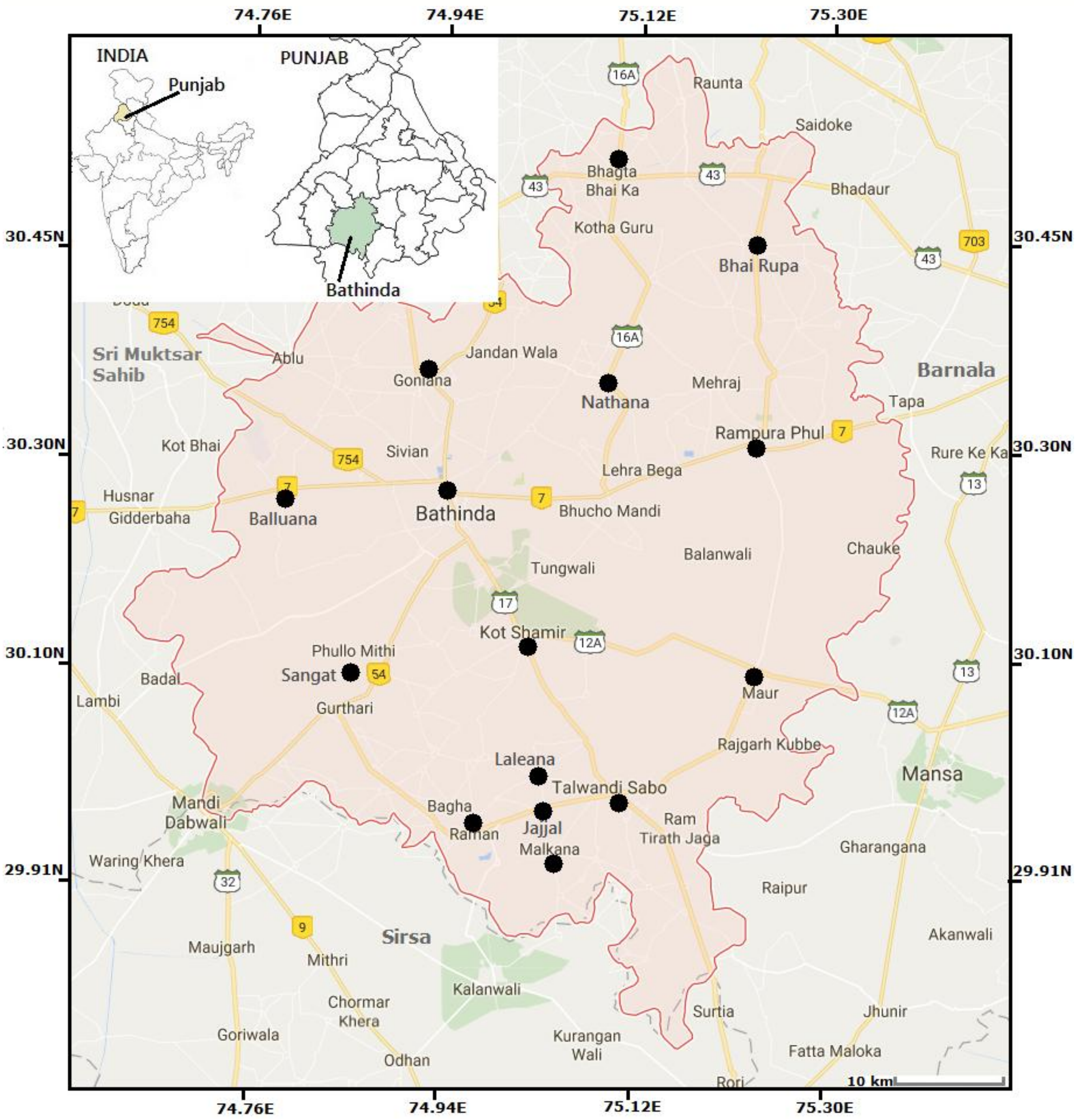

Figure 1: Map showing the sample locations in Bathinda district of Punjab

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Table 1: Instrument operating conditions of HR-ICP-MS (Nu Attom)

Plasma control parameters	
Coolant gas flow rate	13 l/min
Auxillary gas flow rate	1.05 l/min
Nebulizer gas pressure	33.3 psi
Forward RF power	1300 W
Peristaltic rate	15 rpm
Peltier cooling temperature	5 °C
Spray chamber	Glass-Cyclonic
Sample uptake rate	0.2 ml/min
Detector	Ion counter and faraday
Sensitivity	1.1×10^6 counts for ^{115}In 2.1×10^6 counts for ^{238}U
Scan type	Magnet jumping with electric scan over a small mass range
Ion lens setting	Optimized for sensitivity and resolution peaks
Data acquisition parameters	
Dwell time per peak	3 ms
Switch delay per peak	200 µs
Number of sweeps	50
Number of cycles	3
Instrument resolution	300
Internal standard	^{103}Rh

Table 2: Concentrations of ^{238}U, ^{232}Th and physico-chemical parameters in groundwater

Sr. No.	Location	Latitude (N)	Longitude (E)	Depth (ft.)	Temp. (°C)	^{238}U (µg l^{-1})	^{232}Th (µg l^{-1})	pH	TDS (mg l^{-1})	EC (µS cm^{-1})	Salinity (ppt)
1	Malkana	29°56′7″	75°2′9″	90	18	39	0.41	7.8	323	656	0.2
2	Rama Mandi	29°57′23″	74°58′6″	75	16	73	1.66	7.6	371	754	0.3
3	Jajal	29°57′57″	75°1′45″	65	18	156	1.17	7.6	465	982	0.4
4	Talwandi Sabo	29°58′54″	75°4′58″	70	19	6	0.77	8.0	100	199	0.1
5	Laleana	29°59′44″	75°1′21″	60	19	143	1.71	7.5	716	1456	0.8
6	Maur	30°3′59″	75°13′23″	75	15	64	0.66	7.3	556	1161	0.6
7	Sangat	30°4′54″	74°50′23″	40	13	43	0.70	7.9	238	486	0.1
8	Kot Shamir	30°6′42″	75°0′27″	75	16	37	1.79	7.6	621	1242	0.7
9	Bathinda city	30°12′19″	74°55′34″	120	15	4	0.49	7.4	69	141	0.1
10	Balluana	30°13′16″	74°46′56″	35	12	88	1.28	8.0	438	885	0.4
11	Rampura Phul	30°16′28″	75°14′8″	80	15	5	0.48	7.6	72	144	0.1
12	Nathana	30°18′43″	75°5′31″	70	18	65	0.55	7.6	334	672	0.3
13	Goniana	30°19′19″	74°54′47″	60	18	42	0.89	7.5	192	378	0.1
14	Bhai Rupa	30°25′48″	75°13′13″	130	18	41	0.84	7.8	289	583	0.2
15	Bhagta	30°28′59″	75°5′53″	50	14	163	0.52	7.3	272	554	0.2

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Table 3: Summary statistics of ^{238}U and ^{232}Th in groundwater

Statistics	^{238}U	^{232}Th
Arithmetic Mean	64.6	0.93
Standard error	13.5	0.12
Median	43	0.77
Standard deviation	52.4	0.48
Sample variance	2784	0.23
Geometric mean	40.4	0.82
GSD	3.3	1.64
Skewness	0.86	0.83
Kurtosis	-0.25	-0.75
Minimum	4	0.41
Maximum	163	1.79
N	15	15

Table 4: Age-dependent annual effective dose (μSv y^{-1}) due to intake of ^{238}U and ^{232}Th through the ingestion of groundwater

Life stage group	Age group	Total daily water intake (DWI) (L day^{-1})	Annual effective dose (μSv y^{-1})							
			^{238}U				^{232}Th			
			AM	SD	Max	Min	AM	SD	Max	Min
Infants	0–6 months	0.7	70	55	176	4	4.4	2.2	8.5	2.0
	7–12 months	0.8	80	62	201	5	5.1	2.5	9.8	2.2
Children	1 – 3 y	1.3	46	36	115	3	0.8	0.4	1.6	0.4
	4 – 8 y	1.7	40	31	100	3	0.8	0.4	1.6	0.4
Males	9 – 13 y	2.4	48	37	120	3	1.0	0.5	1.9	0.4
	14 – 18 y	3.3	65	51	163	4	1.1	0.6	2.2	0.5
	Adults	3.7	49	38	123	3	1.2	0.6	2.3	0.5
Females	9 – 13 y	2.1	42	33	105	3	0.8	0.4	1.6	0.4
	14 – 18 y	2.3	45	35	114	3	0.8	0.4	1.5	0.4
	Adults	2.7	36	28	90	2	0.9	0.4	1.7	0.4
Pregnancy	14 – 18 y	3.0	59	46	148	4	1.0	0.5	2.0	0.5
	19 – 50 y	3.0	40	31	100	2	1.0	0.5	1.8	0.4
Lactation	14 – 18 y	3.8	74	58	188	5	1.3	0.7	2.5	0.6
	19 – 50 y	3.8	50	39	126	3	1.2	0.6	2.3	0.5

Table 5: Radiological and chemical toxicity risks due to intake of ^{238}U and ^{232}Th through the ingestion of groundwater

Sr. No.	Sample Location	Lifetime cancer risk				Chemical toxicity risk				
		^{238}U		^{232}Th		^{238}U			^{232}Th	
		Mortality risk	Morbidity risk	Mortality risk	Morbidity risk	LADD	HQ according to		LADD	HQ according to USEPA
							WHO and USEPA	AERB		
1	Malkana	2.8E-05	4.3E-05	1.6E-07	2.3E-07	1.11	1.30	0.65	1.2E-02	3.0E-03
2	Rama Mandi	5.2E-05	8.0E-05	6.4E-07	9.4E-07	2.09	2.43	1.22	4.7E-02	1.2E-02
3	Jajal	1.1E-04	1.7E-04	4.5E-07	6.6E-07	4.46	5.20	2.60	3.3E-02	8.5E-03
4	Talwandi Sabo	4.3E-06	6.6E-06	3.0E-07	4.4E-07	0.17	0.20	0.10	2.2E-02	5.6E-03
5	Laleana	1.0E-04	1.6E-04	6.6E-07	9.7E-07	4.09	4.77	2.38	4.9E-02	1.3E-02
6	Maur	4.6E-05	7.0E-05	2.6E-07	3.7E-07	1.83	2.13	1.07	1.9E-02	4.8E-03
7	Sangat	3.1E-05	4.7E-05	2.7E-07	4.0E-07	1.23	1.43	0.72	2.0E-02	5.1E-03
8	Kot Shamir	2.7E-05	4.1E-05	6.9E-07	1.0E-06	1.06	1.23	0.62	5.1E-02	1.3E-02
9	Bathinda city	2.9E-06	4.4E-06	1.9E-07	2.8E-07	0.11	0.13	0.07	1.4E-02	3.6E-03
10	Balluana	6.3E-05	9.7E-05	5.0E-07	7.3E-07	2.51	2.93	1.47	3.7E-02	9.3E-03
11	Rampura Phul	3.6E-06	5.5E-06	1.9E-07	2.7E-07	0.14	0.17	0.08	1.4E-02	3.5E-03
12	Nathana	4.7E-05	7.1E-05	2.1E-07	3.1E-07	1.86	2.17	1.08	1.6E-02	4.0E-03
13	Goniana	3.0E-05	4.6E-05	3.5E-07	5.0E-07	1.20	1.40	0.70	2.5E-02	6.5E-03
14	Bhai Rupa	2.9E-05	4.5E-05	3.3E-07	4.8E-07	1.17	1.37	0.68	2.4E-02	6.1E-03
15	Bhagta	1.2E-04	1.8E-04	2.0E-07	3.0E-07	4.66	5.43	2.72	1.5E-02	3.8E-03