

# Chemotoxicity and Radiotoxicity Risk Assessment from Exposure to Uranium in Groundwater from Western Haryana, India

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**Abstract**—In this study, the distribution of uranium in groundwater and health risks were evaluated. The samples were analyzed by using LED fluorimeter. The concentration of uranium ranged from 1.1 to 113  $\mu\text{g l}^{-1}$  with mean value of 22.3  $\mu\text{g l}^{-1}$ . These recorded values were compared with the maximum contamination levels recommended for drinking water by various health and environmental protection agencies. The associated age-dependent annual effective radiation dose is estimated by taking the prescribed water intake values of different age groups. The average cancer mortality and morbidity risks are calculated to be  $3.3 \times 10^{-5}$  and  $5.0 \times 10^{-5}$  respectively, indicate that the probability of carcinogenic risks is negligible. About 20 % of the analyzed samples showed hazard quotient  $> 1.0$ , indicating significant risk due to chemical toxicity of uranium.

**Index Terms**— Drinking water, Uranium, LED fluorimeter, Dose assessment, Carcinogenic risk, Chemical risk.

## I. INTRODUCTION

URANIUM is natural lithophilic element and is contained almost in all natural soils and waters; however, their concentration varies from water to water depending upon their origin. Various factors such as age, sex, body weight and metabolic activity govern the water intake by an individual. The health effects of uranium can be divided into carcinogenic and non-carcinogenic effects, and this classification is based on the radiological risk posed by the radiation due to uranium isotopes and the chemical risk posed by uranium as a heavy metal. The important target of uranium are kidneys and lungs [1], [2]. According to an estimate, food contributes about 15 % of ingested uranium, while drinking water contributes about 85 % [3]. The need of estimation of uranium concentration in water is multifold: it is an important fuel for nuclear power reactors, the hydrogeochemical prospecting for uranium is essential and the assessment of risk of health hazards due to high concentration of uranium in water is most important.

High uranium concentrations in public drinking water around the world have been reported [4], [5], [6], [7] but the variation in dose exposure rate among different life stage

groups has been studied only in a few publications such as Duggal et al. [8], [9], Yadav et al. [10] and Patra et al. [11].

The main objective of this study is to estimate the concentration of uranium in groundwater, to compute of annual effective radiation doses for different life stage groups, and to assess radiological and chemical toxicity risks to humans due to intake of uranium through the ingestion of groundwater from Fatehabad district, Haryana, India.

## II. GEOLOGY OF THE STUDY AREA

The Haryana state is located between  $27^{\circ} 39'$  and  $30^{\circ} 35'$  North latitudes and  $74^{\circ} 28'$  and  $77^{\circ} 36'$  East longitudes in the North India. Figure 1 shows the geographic location of Fatehabad district on the map of Haryana, as well as the location of the sampling sites. The total human population of the Fatehabad district is 9.5 lakh (2011 census), and geographical area of 2520  $\text{km}^2$ . Geography of Fatehabad district comprises part of the alluvial or Ghaggar-Yamuna plain and its southern and western portions mark a gradual transition to Thar Desert in India. The Fatehabad district is sub-divided into three topographic units—sub-recent alluvial plain, late quaternary to sub-recent sand dune areas and plain with sand dunes. The plain locally known as Nali occurs principally as floodplain sediments along the courses of the Ghaggar River. The sand dune areas are classified into conspicuous topographic units having different genetic types of sand dunes such as active, semi-active and fossil or fixed. Hard rock geology of Fatehabad district is concealed under alluvial and aeolian deposits. The region is bounded by Punjab state in north, district Hisar in south, district Jind in east and Rajasthan state and district Sirsa in the west.

## III. MATERIALS AND METHODS

A total of 54 groundwater samples from Fatehabad district, Haryana were collected from manually operated hand-pumps established by municipal corporations in residential localities or from privately owned hand-pumps, tube-wells and electric motors. Prior to collection, the water samples were filtered using 0.45  $\mu\text{m}$  Whatmann filter paper to remove suspended matter/sediments, acidified with supra pure nitric acid ( $\text{HNO}_3$ ) for preservation and then stored in pre-cleaned acid-washed high density polyethylene (HDPE) containers until analysis. Electrical conductivity (EC), pH, and total dissolved solids (TDS) in groundwater were measured *in situ* with the help of a

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micro controller water analysis kit (NPC 362D, Naina Solaris, New Delhi, India) [12].

Uranium estimation in groundwater has been carried out in the Environmental Radiation Research Laboratory of Guru Nanak Dev University, Amritsar, India using LED fluorimeter LF-2a (Quantalase Enterprises Private Limited, Indore, India). 0.5 mL of 5 % sodium pyrophosphate solution was added to 5 mL of the water sample and a pH value of 7.0 is adjusted by drop-wise addition of orthophosphoric acid. This solution acts as fluorescence enhancing reagent [11]. The upper limit of detection of uranium in groundwater was 1000  $\mu\text{g L}^{-1}$  and the lower limit of detection was 0.5  $\mu\text{g L}^{-1}$ . The laser fluorimeter was calibrated with standard solution of uranium and reagent blanks were run with water samples to ensure the accuracy of the results. Details of instrument calibration, analytical procedure, quality assurance, and quality control are given elsewhere [13], [14], [15].

#### IV. RESULTS AND DISCUSSION

##### A. Distribution of Uranium in Groundwater

Uranium concentrations in the groundwater together with location, source, depth, and physico-chemical analysis data are presented in Table 1. The concentration of uranium varied from 1.1 to 113  $\mu\text{g l}^{-1}$  with a mean value of 22.3  $\mu\text{g l}^{-1}$ . Uranium content in groundwater varied greatly in different villages. The measured uranium content in 20 % of the analyzed samples exceeded the World Health Organization [1] and United States Environmental Protection Agency [16] drinking water guidelines of 30  $\mu\text{g l}^{-1}$  and 9 % of the samples exceeded the 60  $\mu\text{g l}^{-1}$  Indian maximum acceptable concentration recommended by the Atomic Energy Regulatory Board, India [17]. Hence it is important to assess its possible impact on humans who may consume this water. No correlation was observed between uranium and depth of groundwater.

The depth from the ground surface to the groundwater level ranged from 40 to 550 ft. Water pH ranged between 7.4 and 8.5. All the water samples had pH values within permissible limits recommended by the Bureau of Indian Standards [18]. TDS and EC of the drinking water samples ranged from 182 to 5870  $\text{mg l}^{-1}$  and 362 to 8300  $\mu\text{S cm}^{-1}$  with average values of 1391  $\text{mg l}^{-1}$  and 2551  $\mu\text{S cm}^{-1}$ , respectively. For TDS and EC, only 24 % and 37 % of the water samples had values within the permissible limits of 600  $\text{mg l}^{-1}$  [1] and 1500  $\mu\text{S cm}^{-1}$  [19], respectively. Uranium concentrations were not found to be correlated with pH, TDS and EC.

##### B. Radiation Dose

Annual radiation dose to human (for different age groups) due to uranium consumption was quantified using Equation 1 [9], [20].

$$D (\mu\text{Sv y}^{-1}) = C \times DWI \times 365.25 \times DCF \quad (1)$$

where D annual radiation dose ( $\mu\text{Sv y}^{-1}$ ), C activity concentration of uranium ( $\text{Bq l}^{-1}$ ); DWI age-dependent daily

water intake prescribed by Dietary Reference Intakes, Food and Nutrition Board, Institute of Medicine, US National Academy of Science [21], DCF dose conversion factor for specific age group ( $\text{Sv Bq}^{-1}$ ) given by International Atomic Energy Agency [22].

The results for annual effective radiation dose due to intake of uranium through the drinking water for different age groups are presented in Table 2. The uranium activity concentration was calculated by using unit conversion factor ( $1 \mu\text{g L}^{-1} = 0.02528 \text{ Bq L}^{-1}$ ) [8], [23]. The recommended level of annual effective dose to human from water consumption is 100  $\mu\text{Sv y}^{-1}$  [1]. This reference dose level represents approximately 4.2 % of the average annual effective dose from natural background radiation ( $2.4 \text{ mSv y}^{-1}$ ) [1]. The mean annual effective dose is lower in the age group of infants 0–6 months old, due to lower annual water intake (256 L) compared with the 7–12 months group (292 L). Even though infants drink less water than adults, the annual effective dose to infants are significantly higher than that to adults, because of the differences in infant's metabolism and smaller organ weights resulting in higher doses for many radionuclides. Females receive higher radiation dose during lactation and pregnancy due to increased water consumption. The estimated annual effective dose due to the intake of uranium through drinking water for various groups ranged from 1.4 to 284  $\mu\text{Sv y}^{-1}$  with an average value of 37.3  $\mu\text{Sv y}^{-1}$ . The mean annual effective dose in all the age groups was found to be lower than the WHO recommended level of 100  $\mu\text{Sv y}^{-1}$ .

##### C. Radiological Risk

The radiological risk and intake were estimated as proposed by U.S. Environmental Protection Agency [24] (Equations 2 and 3).

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

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

Where C is the activity concentration of uranium in drinking water ( $\text{Bq l}^{-1}$ ), IR the water ingestion rate ( $2 \text{ L day}^{-1}$ ) [25], EF the exposure frequency ( $365.25 \text{ days y}^{-1}$ ), and ED the lifetime exposure duration (70 y) [16].

According to the USEPA [24], the mortality and morbidity cancer risk coefficients of  $1.13 \times 10^{-9}$  and  $1.73 \times 10^{-9}$ , respectively, have been used for the estimation of cancer mortality and morbidity risks of uranium over lifetime consumption of groundwater (Table 3). The cancer mortality and morbidity risks ranged from  $1.6 \times 10^{-6}$  to  $1.7 \times 10^{-4}$  and  $2.5 \times 10^{-6}$  to  $2.5 \times 10^{-4}$  with the average values of  $3.3 \times 10^{-5}$  and  $5.0 \times 10^{-5}$ , respectively. The calculated radiological risk was comparatively lower than those reported for SW-Punjab, India ( $2.06 \times 10^{-4}$ ) by Bajwa et al. [26], and for Northern Rajasthan, India ( $5.6 \times 10^{-5}$ ) by Duggal et al. [8], but higher than those reported for Jaduguda, India ( $4.8 \times 10^{-6}$ ) by Patra et al. [11].

#### D. Chemical Toxicity Risk

The chemical/non-carcinogenic risk has been calculated as hazard quotient (HQ) as the ratio of the lifetime average daily dose (LADD) to its reference dose [27] Equations (4) and (5).

$$\text{Hazard Quotient (HQ)} = \text{LADD/RfD} \quad (4)$$

$$\text{LADD } (\mu\text{g kg}^{-1} \text{d}^{-1}) = (\text{C}_U \times \text{IR} \times \text{EF} \times \text{ED}) / (\text{AT} \times \text{BW}) \quad (5)$$

where  $C_U$  is the concentration of uranium in drinking water ( $\mu\text{g l}^{-1}$ ), AT average exposure time for non-carcinogens ( $365.25 \times 70$  days), and BW the body weight (70 kg) [28].

The LADD of uranium due to consumption of groundwater ranged from 0.03 to  $3.23 \mu\text{g kg}^{-1} \text{d}^{-1}$  with a mean value of  $0.64 \mu\text{g kg}^{-1} \text{d}^{-1}$ . HQ was calculated for uranium using reference dose of  $0.857 \mu\text{g kg}^{-1} \text{d}^{-1}$  recommended by WHO [1]. According to WHO standard, the HQ ranged from 0.04 to 3.77. About 20 % of the analyzed samples showed  $\text{HQ} > 1.0$ , indicating significant risk due to chemical toxicity of uranium.

#### V. CONCLUSION

After evaluating data, it is clear that the level of uranium at some sampling sites is higher than the permissible limits recommended by WHO, USEPA and Indian Standard (AERB) for drinking water. High uranium concentrations observed in groundwater may be due to local natural geology, anthropogenic activities, urbanization, and use of phosphate fertilizers in huge quantity for agriculture purpose. But overall it seems that the plausible source of high uranium observed may be geogenic in nature. The results showed that the probability of carcinogenic risk is negligible but non-carcinogenic health risk may be due to chemical toxicity of uranium.

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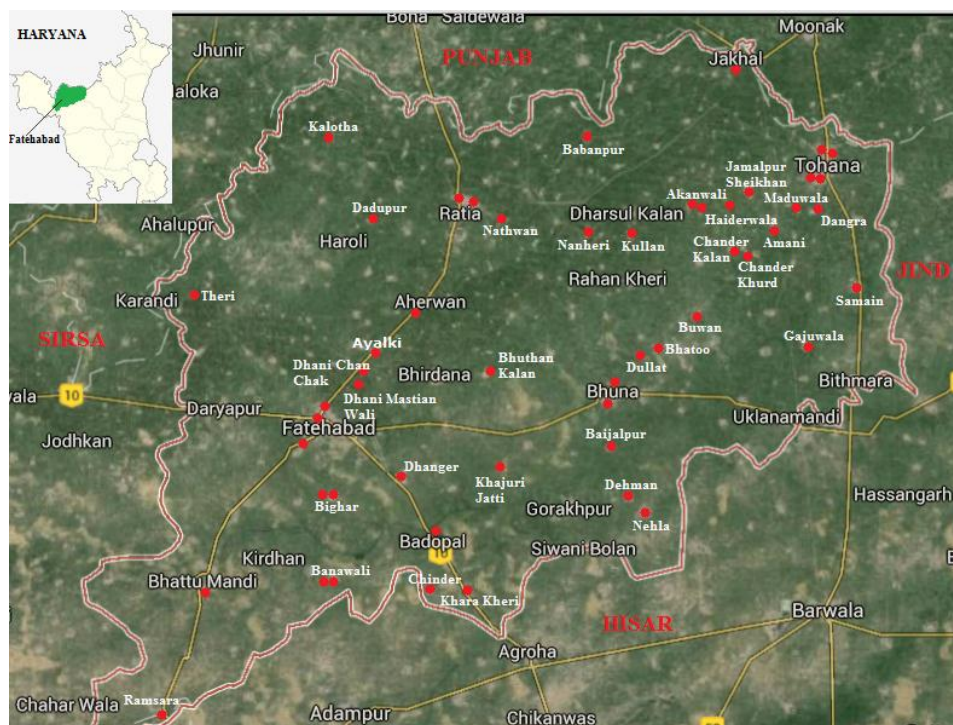


Fig. 1. Location map of the study area and sampling sites.

Table 1  
Concentration of uranium, pH, TDS and EC in groundwater from Fatehabad district, Haryana

Sr. No.	Sample Location	Latitude and Longitude	Source	Depth (Feet)	Uranium Conc. ( $\mu\text{g l}^{-1}$ )	pH	TDS ( $\text{mg l}^{-1}$ )	EC ( $\mu\text{S cm}^{-1}$ )
1	Ramsara	29.28 N 75.30 E	EM	160	11.54	8.3	1050	2120
2	Bhattu Mandi	29.38 N 75.34 E	TW	110	33.60	8.2	1980	3910
3	Chinder	29.38 N 75.54 E	HP	40	33.45	8.1	3960	8030
4	Khara Kheri	29.38 N 75.57 E	TW	60	61.25	7.8	4020	8300
5	Khara Kheri	29.38 N 75.58 E	TW	40	23.60	7.9	3680	7340
6	Banawali	29.39 N 75.43 E	Water works	—	1.11	8.0	196	382
7	Banawali	29.40 N 75.44 E	EM	50	25.08	7.9	3610	6940
8	Badopal	29.42 N 75.55 E	TW	80	18.26	7.7	726	1323
9	Nehla	29.44 N 75.74 E	HP	50	4.95	7.7	476	1026
10	Bighar	29.45 N 75.45 E	TW	80	9.84	8.2	2200	4310
11	Bighar	29.45 N 75.46 E	HP	60	8.21	8.5	2010	4090
12	Dehman	29.45 N 75.72 E	TW	60	2.29	8.0	230	442
13	Dhanger	29.46 N 75.52 E	TW	100	25.61	8.1	2090	4140
14	Khajuri Jatti	29.48 N 75.61 E	TW	80	23.75	7.9	1834	3625
15	Baijalpur (S-1)	29.49 N 75.70 E	Canal water	—	2.41	7.9	338	669
16	Baijalpur (S-2)	29.49 N 75.71 E	TW	150	5.78	8.0	403	819
17	Fatehabad City (S-1)	29.50 N 75.44 E	HP	60	21.43	7.8	710	1410
18	Fatehabad City (S-2)	29.50 N 75.47 E	EM	90	13.27	7.8	620	1230
19	Fatehabad City (S-3)	29.52 N 75.45 E	EM	80	7.45	8.0	5870	1127
20	Bhuna (S-1)	29.53 N 75.70 E	HP	80	10.41	7.7	1780	3650
21	Bhuna (S-2)	29.53 N 75.71 E	TW	105	68.0	7.6	3500	6540
22	Dhani Mastian Wali	29.54 N 75.48 E	EM	100	6.32	8.1	385	767
23	Bhuthan Kalan	29.55 N 75.60 E	EM	100	35.31	8.0	2050	4080
24	Dullat	29.56 N 75.73 E	TW	275	12.25	8.2	341	676
25	Ayalki	29.57 N 75.49 E	TW	425	11.51	8.2	660	1300
26	Gajuwala	29.57 N 75.89 E	TW	220	12.38	7.9	1356	2735
27	Dhani Chan Chak	29.58 N 75.51 E	TW	80	9.53	8.2	460	974
28	Bhatoo	29.58 N 75.77 E	TW	125	11.28	8.0	320	610
29	Buwan	29.59 N 75.78 E	EM	150	8.18	8.3	1480	3000
30	Theri	29.61 N 75.33 E	EM	100	20.21	7.9	879	1705

31	Aherwan	29.61 N 75.54 E	EM	150	8.39	8.0	1050	2090
32	Samain	29.62 N 75.93 E	TW	250	22.36	7.7	2310	2640
33	Chander Kalan	29.65 N 75.82 E	TW	225	1.395	8.0	308	624
34	Chander Khurd	29.65 N 75.83 E	TW	250	9.07	7.8	2430	4890
35	Nanheri	29.66 N 75.68 E	TW	400	11.98	7.8	604	730
36	Kullan	29.66 N 75.73 E	TW	250	4.85	8.0	350	704
37	Dadupur	29.67 N 75.49 E	EM	180	12.38	7.9	938	1880
38	Ratia (S-1)	29.67 N 75.57 E	TW	250	23.49	8.0	910	1836
39	Ratia (S-2)	29.67 N 75.58 E	TW	200	21.43	8.2	889	1759
40	Nathwan	29.67 N 75.61 E	TW	400	17.07	8.3	940	1803
41	Amani	29.67 N 75.85 E	TW	180	20.15	8.5	669	1992
42	Akanwali (S-1)	29.68 N 75.78 E	TW	550	47.92	8.3	1380	2600
43	Akanwali (S-2)	29.68 N 75.79 E	TW	350	8.90	7.4	629	1007
44	Maduwala	29.68 N 75.88 E	TW	230	43.53	8.3	1500	3030
45	Dangra	29.68 N 75.89 E	TW	200	18.59	8.3	860	1693
46	Haiderwala	29.69 N 75.80 E	TW	310	20.31	8.1	926	1978
47	Jamalpur Sheikhan	29.70 N 75.83 E	TW	350	22.25	8.1	1230	2390
48	Tohana (S-1)	29.70 N 75.90 E	Water works	—	2.70	8.1	182	362
49	Tohana (S-2)	29.71 N 75.89 E	EM	320	112.97	8.0	1155	3120
50	Tohana (S-3)	29.71 N 75.90 E	EM	290	92.78	8.1	1120	2280
51	Tohana (S-4)	29.72 N 75.91 E	EM	250	65.20	7.9	310	625
52	Babanpur	29.73 N 75.69 E	TW	250	25.26	8.0	934	1860
53	Kalotha	29.74 N 75.45 E	TW	150	13.34	7.6	3210	6470
54	Jakhal	29.80 N 75.82 E	EM	200	42.21	7.9	1056	2100

Table 2

Age-dependent annual effective radiation dose due to intake of uranium through the ingestion of groundwater

Life Stage Group	Age Group	DWI ( $\text{I d}^{-1}$ )	Annual Effective Radiation Dose ( $\mu\text{Sv y}^{-1}$ )			
			Min	Max	Mean	SD
Infants	0–6 months	0.7	2.4	248	49	49
	7–12 months	0.8	2.8	284	56	56
Children	1–3 y	1.3	1.6	163	32	32
	4–8 y	1.7	1.4	142	28	28
Males	9–13 y	2.4	1.7	170	34	33
	14–18 y	3.3	2.3	231	46	45
Females	Adults	3.7	1.7	174	34	34
	9–13 y	2.1	1.5	149	29	29
Pregnancy	14–18 y	2.3	1.6	161	32	31
	Adults	2.7	1.2	127	25	25
Lactation	14–18 y	3.0	2.1	210	41	41
	19–50 y	3.0	1.4	141	28	28
Lactation	14–18 y	3.8	2.6	266	53	52
	19–50 y	3.8	1.7	178	35	35

Table 3

Uranium activity concentration, cancer mortality and morbidity risks, LADD and HQ due to ingestion of uranium in groundwater

Sr. No.	Sample Location	Uranium activity concentration ( $\text{Bq l}^{-1}$ )	Lifetime cancer risk		Chemical toxicity risk	
			Cancer mortality risk	Cancer morbidity risk	LADD ( $\mu\text{g kg}^{-1} \text{d}^{-1}$ )	HQ
1	Ramsara	0.29	1.7E-05	2.6E-05	0.33	0.38
2	Bhattu Mandi	0.85	4.9E-05	7.5E-05	0.96	1.12
3	Chinder	0.85	4.9E-05	7.5E-05	0.96	1.12
4	Khara Kheri	1.55	8.9E-05	1.4E-04	1.75	2.04

5	Khara Kheri	0.60	3.4E-05	5.3E-05	0.67	0.79
6	Banawali	0.03	1.6E-06	2.5E-06	0.03	0.04
7	Banawali	0.63	3.7E-05	5.6E-05	0.72	0.84
8	Badopal	0.46	2.7E-05	4.1E-05	0.52	0.61
9	Nehla	0.13	7.2E-06	1.1E-05	0.14	0.17
10	Bighar	0.25	1.4E-05	2.2E-05	0.28	0.33
11	Bighar	0.21	1.2E-05	1.8E-05	0.23	0.27
12	Dehman	0.06	3.3E-06	5.1E-06	0.07	0.08
13	Dhanger	0.65	3.7E-05	5.7E-05	0.73	0.85
14	Khajuri Jatti	0.60	3.5E-05	5.3E-05	0.68	0.79
15	Baijalpur (S-1)	0.06	3.5E-06	5.4E-06	0.07	0.08
16	Baijalpur (S-2)	0.15	8.4E-06	1.3E-05	0.17	0.19
17	Fatehabad City (S-1)	0.54	3.1E-05	4.8E-05	0.61	0.71
18	Fatehabad City (S-2)	0.34	1.9E-05	3.0E-05	0.38	0.44
19	Fatehabad City (S-3)	0.19	1.1E-05	1.7E-05	0.21	0.25
20	Bhuna (S-1)	0.26	1.5E-05	2.3E-05	0.30	0.35
21	Bhuna (S-2)	1.72	9.9E-05	1.5E-04	1.94	2.27
22	Dhani Mastian Wali	0.16	9.2E-06	1.4E-05	0.18	0.21
23	Bhuthan Kalan	0.89	5.2E-05	7.9E-05	1.01	1.18
24	Dullat	0.31	1.8E-05	2.7E-05	0.35	0.41
25	Ayalki	0.29	1.7E-05	2.6E-05	0.33	0.38
26	Gajuwala	0.31	1.8E-05	2.8E-05	0.35	0.41
27	Dhani Chan Chak	0.24	1.4E-05	2.1E-05	0.27	0.32
28	Bhatoo	0.29	1.6E-05	2.5E-05	0.32	0.38
29	Buwan	0.21	1.2E-05	1.8E-05	0.23	0.27
30	Theri	0.51	3.0E-05	4.5E-05	0.58	0.67
31	Aherwan	0.21	1.2E-05	1.9E-05	0.24	0.28
32	Samain	0.57	3.3E-05	5.0E-05	0.64	0.75
33	Chander Kalan	0.04	2.0E-06	3.1E-06	0.04	0.05
34	Chander Khurd	0.23	1.3E-05	2.0E-05	0.26	0.30
35	Nanheri	0.30	1.7E-05	2.7E-05	0.34	0.40
36	Kullan	0.12	7.1E-06	1.1E-05	0.14	0.16
37	Dadupur	0.31	1.8E-05	2.8E-05	0.35	0.41
38	Ratia (S-1)	0.59	3.4E-05	5.3E-05	0.67	0.78
39	Ratia (S-2)	0.54	3.1E-05	4.8E-05	0.61	0.71
40	Nathwan	0.43	2.5E-05	3.8E-05	0.49	0.57
41	Amani	0.51	2.9E-05	4.5E-05	0.58	0.67
42	Akanwali (S-1)	1.21	7.0E-05	1.1E-04	1.37	1.60
43	Akanwali (S-2)	0.22	1.3E-05	2.0E-05	0.25	0.30
44	Maduwala	1.10	6.4E-05	9.7E-05	1.24	1.45
45	Dangra	0.47	2.7E-05	4.2E-05	0.53	0.62
46	Haiderwala	0.51	3.0E-05	4.5E-05	0.58	0.68
47	Jamalpur Sheikhan	0.56	3.3E-05	5.0E-05	0.64	0.74
48	Tohana (S-1)	0.07	3.9E-06	6.0E-06	0.08	0.09
49	Tohana (S-2)	2.86	1.7E-04	2.5E-04	3.23	3.77
50	Tohana (S-3)	2.35	1.4E-04	2.1E-04	2.65	3.09
51	Tohana (S-4)	1.65	9.5E-05	1.5E-04	1.86	2.17
52	Babanpur	0.64	3.7E-05	5.6E-05	0.72	0.84
53	Kalotha	0.34	1.9E-05	3.0E-05	0.38	0.44
54	Jakhal	1.07	6.2E-05	9.4E-05	1.21	1.41