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DEVELOPING A MODEL FOR URINARY FLUORIDE LEVELS IN RELATION TO THE FLUORIDE CONTENT OF DRINKING WATER AND FOODSTUFFS IN A RURAL POPULATION IN LAKHIMPUR (KHERI), UTTAR PRADESH, INDIA

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ABSTRACT: The aim of the study was to develop a model for describing the relationship between the fluoride (F) content in drinking water and foodstuffs, and the urinary F level in residents, of different age groups, residing in low and high F exposure areas in Mitauli block, Lakhimpur (Kheri) district of Uttar Pradesh, India. The rural population at the study sites used groundwater for drinking and irrigation purposes without any quality test. The source of the F contamination in the groundwater was natural as no large industries were located in the study area which contained only a few agro-based industries.The F concentration was measured in the groundwater from 73 sites in Khanta, Khudania, Murai-tajpur, Salahpur, and Usari villages and in the urine of 548 residents. The range of the F concentration in the drinking water in Khanta, Khudania, Murai-tajpur, Salahpur, and Usari villages was 1.0–2.3, 0.5–1.4, 0.9–1.5, 1.8–2.8, and 2.7–4.3 mg L^{-1}, respectively. The urinary F level in the 548 villagers was found to vary in different age groups. The highest urinary F concentration (4.73 mg L^{-1}) was found in younger males, aged 20–40 yr, with the next highest group (3.72 mg L^{-1}) being older females, aged 60–70 yr.

Keywords: Groundwater contamination; Human; Physio-chemical properties; Urinary fluoride.

INTRODUCTION

Contamination of drinking water by fluoride (F) and the associated disease, fluorosis, is a growing problem across the globe and India is one of the countries affected. An excess of F in drinking water is a key aspect of water quality, especially in rural water supply systems, as it causes dental, skeletal, and non-skeletal fluorosis. An increasing scarcity of freshwater resources is driving the residents in the arid and semi-arid regions of many countries to use water of marginal quality for agriculture, drinking, and related activities.[1] Worldwide, more than 200 million people from 28 tropical countries are suffering from fluorosis, mainly due to the high F content in the drinking water.[2-6]

Although the World Health Organization set, in 1984 and reaffirmed in 1993, a guideline of 1.5 mg F/L (1.5 ppm) as a “desirable” upper limit, it also allows countries to set country standards, their own national standards or local guidelines.[7] The limit of 1.5 mg F/L has been seen to be unsuitable in some countries and lower country standards have been set of 1 mg/L in India and 0.6 mg/L in Senegal, West Africa.[8] A rider to the Indian limit is the “lesser the fluoride the better, as fluoride is injurious to health.”[8]

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In an update to the website of the Centers for Disease Control and Prevention, dated 24 April 2015,[9] it was noted that the US Department of Health and Human Services Federal Panel on Community Water Fluoridation has made a final recommendation on community water fluoridation that replaces the relevant parts of the 1962 Drinking Water Standards.[10,11] Whereas the earlier recommendation, based on the outdoor air temperature of geographic regions, involved a range of 0.7–1.2 mg F/L, the new recommendation, for community water systems, that currently fluoridate or plan to do so, is for an optimal F concentration in drinking water of 0.7 mg/L. The US Surgeon General, Dr VH Murthy, endorsed the recommendation and urged that communities adopt it.[12]

The maximum permissible limit (MPL) for F in the drinking water in India is 1.0–1.5 mg L^{-1}.[13] In India, more than 20 states have become endemic for fluorosis due to the ingestion of F-contaminated drinking water with levels up to 38.5 mg L^{-1}.[14-17] Higher F levels in water are found in different parts of the world, especially in the developing countries.[17] Besides causing dental fluorosis and skeletal fluorosis, chronic exposure to F may lead to non-skeletal manifestations such as kidney, liver, and brain damage.[18,19] It is reported that the excessive consumption of F may lead to muscle fibre degeneration, low haemoglobin level, excessive thirst, headache, skin rashes, nervousness, and depression.[20,21] It may also be harmful to the cardiovascular system.[22-24] Although chronic fluorosis may severely damage many systems of the human body, its pathogenicity is poorly understood.[18,25]

The present study was planned to help understand the pathogenicity of fluorosis due to high F ingestion through drinking water and foodstuffs by measuring and comparing the urinary F levels in the residents, of different age groups, residing in low and high F exposure areas. The aim of the study was to develop a model describing the relationship between the F content in drinking water and foodstuffs and the urinary F in people, of different age groups, residing in low and high F exposure areas in Mitauli block, Lakhimpur (Kheri) district, Uttar Pradesh, India.

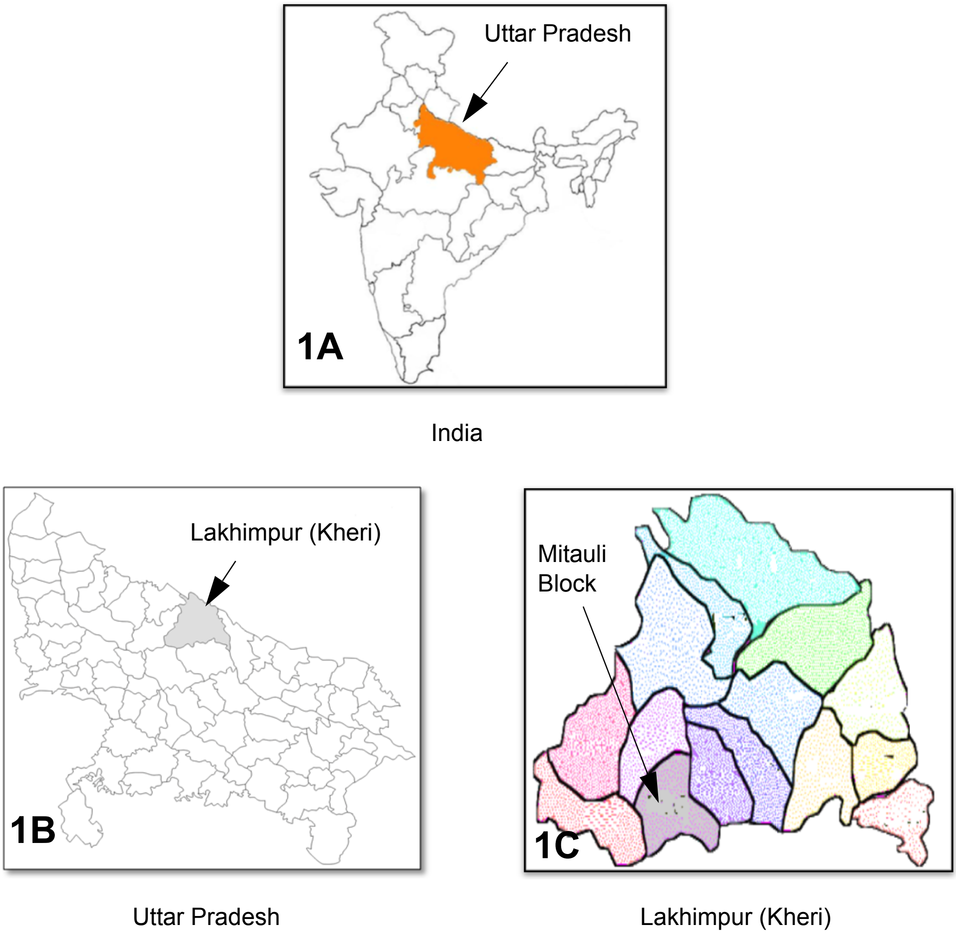
MATERIALS AND METHODS

Prior to the study commencing, a detailed research proposal was submitted to the Science and Engineering Research Board (SERB) Government of India, New Delhi, India. The study was conducted in high and low F contaminated areas after obtaining the approval of the SERB (File No. SB/YS/LS-167/2013). The consents of the participants, parents, and guardians were sought for the participation in the study. A questionnaire was administered to the parents/guardians to collect personal data, i.e., age, sex, height, residential history, medical history, educational qualifications, and socio-economic status of the family.

Ethical considerations: Ethical approval for the research experiment was obtained from the Committee on Human Research, Publication and Ethics (CHRPE) (CSAU/1302/14 and CHRPE/CSAU/163/14) of the C.S. Azad University of Agriculture and Technology, Kanpur, Uttar Pradesh, India.

Sample collection and analysis: The study area lies in the Tarai (wet) region of the State of Uttar Pradesh which stretches along the Himalayan foothills extending between 27.6ºN to 28.6ºN latitude and 80.34ºE to 81.30ºE longitude in India (Figure

1). It covers a total area of 7,680 km^2 with a population of 40,21,243 (2011 census). The district comprises 7 Tehsils and 15 Blocks.



Figures 1A–1C. 1A: The position of the state of Uttar Pradesh in India. 1B: The position of the district of Lakhimpur (Kheri) in the state of Uttar Pradesh; 1C: The position of the study area in the Mitauli Block in the district of Lakhimpur (Kheri).

The climate of the area is subtropical with moderate to heavy rainfall between mid-June and September. The majority of the population in the study area depends upon agriculture and approximately 62.08% of the area is covered with cultivated fertile agricultural land used for the multiple cropping of sugar cane, wheat, and paddy. There are a number of the small scale agro-based industries located in the district involving sugar cane factories and small sugar mills. Public-private hand pumps and tube wells are the major sources of drinking and irrigation water in the region.

The ground water samples were collected from bore wells/hand pumps (n=73) used for drinking purposes. The urine samples (n=548; 277 males and 271 females) were collected in polypropylene bottles separately and carried to the laboratory of the Department of Crop Physiology, C. S. Azad University of Agriculture and Technology, Kanpur (U.P.), India. The water quality parameters of temperature, pH, total hardness (TH), and dissolved oxygen (DO) were determined as per the methods prescribed by the American Public Health Association.[26]

The F in the groundwater and urine samples were analyzed using the method adopted by Gopal et al.[27] and Jha et al.[28] The F concentration in the groundwater and urine samples was analyzed using the F ion selective electrode of a Laqua F ion meter with an equal volume (1:1) of the samples and total ionic strength adjustment buffer-III (SIGMA ALDRICH TISAB-III). The buffer solution was added to the standard and sample to reduce matrix interference and ionic strength. The limit of detection (L.O.D.) was 0.03 mg L^{-1}.

Hypothesis: The F exposure and risk vary to a large extent between the different age groups. The F concentration in infants' urine is low due to high milk consumption and good nutritional status and increases as the age increases due to increased dietary and water intake and a lower milk consumption. In elderly people, the F concentration in the urine declines due to a low dietary intake. Accordingly, a hypothesis for explaining the variation in the F level in the urine samples can be derived by observing the F concentration in the urine and relating this to the age. The F concentration in human urine is dependent on the ingestion of F through drinking water and contaminated foodstuffs. The hypothesis for the present study was:

The rate of change of F concentration in human urine with respect to age is directly proportional to the concentration of F in the drinking water or foodstuffs consumed at that time and the age of the human being.

Mathematically the hypothesis can be expressed as in Equation 1:

$$\frac{dF_{at}}{dt} \propto F\,t \qquad \text{...........................Equation 1}$$

Where:

F	=	Fluoride concentration in drinking water and/or foodstuffs (mg L^{-1})
F_{at}	=	Fluoride concentration in human urine at the age of t years (mg L^{-1})
t	=	Age (years)
$\propto$	=	Proportionality symbol (is proportional to)

The F concentration in human urine is a function of the F concentration in the drinking water or foodstuffs being consumed. A linear correlation between the F concentration in human urine and the foodstuffs consumed can be considered as shown in Equation 2.

$$F_{at} \propto \lambda F \qquad \text{...Equation 2}$$

Where:

F	=	Fluoride concentration in drinking water and/or foodstuffs (mg L^{-1})
F_{at}	=	Fluoride concentration in human urine at the age of t years (mg L^{-1})
$\propto$	=	Proportionality symbol (is proportional to)
λ	=	A constant

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Substituting Equation 2 into Equation 1 and removing the proportionality constant gives the following governing equation (Equation 3):

$$\frac{dF_a}{dt} = \alpha \lambda F_{at}$$ ....Equation 3

Where:

F	=	Fluoride concentration in drinking water and/or foodstuffs (mg L^{-1})
F_{at}	=	Fluoride concentration in human urine at the age of t years (mg L^{-1})
α	=	Proportionality constant
λ	=	A constant

The Equation 3 can be also written as Equation 4:

$$\frac{dF_a}{dt} = k F_{at}$$ ....Equation 4

Where:

k = $\alpha\lambda$, a new constant

Rearranging Equation 4 gives Equation 5:

$$\frac{dF_a}{F_{at}} = -k t$$ ....Equation 5

Integrating Equation 5, gives the following solution (Equations 6 and 7):

$$\ln F_{at} = -k \frac{t^2}{2} + C$$ ....Equation 6

$$\ln F_{at} = -k \frac{t^2}{2} + C$$ ....Equation 7

Where:

C = Integration constant

The value of k can be worked out as below.

At t = 0, $F_{at} = F_{ao}$ (value of F_{at} at central point)

$$F_{ao} = F'_{at} e^0$$

$$F'_{at} = F_{ao}$$

Substituting the value of $F_{at}^{/} = F_{ao}$, gives following equation (Equation 8):

$$F_{at} = F_{ao}\, e^{-k\frac{t^2}{2}} \qquad \text{....................................Equation 8}$$

By considering k as unity, translating the normal distribution rightward by a distance "b", and stretching the standard normal distribution by a factor "c", Equation 8 can be written as Equation 9.

$$F_{at} = F_{ao}\, e^{-\frac{1}{2}(\frac{t-b}{c})^2} \qquad \text{.........................Equation 9}$$

RESULTS

The values obtained for the various physio-chemical parameters after the analysis of the groundwater samples are presented in Table 1.

Table 1. Assessment of drinking water in Khanta, Khudania, Murai-tajpur, Salahpur, and Usari villages of Mitauli block, district Lakhimpur (Kheri), Uttar Pradesh, India. (DL=Desirable limit; PL=Permissible limit; NR=Not relevant; Values with ± are mean±SEM)

Characteristic	DL	PL	Village				
			Khanta (n=16)	Khudania (n=12)	Murai-tajpur (n=10)	Salahpur (n=20)	Usari (n=15)
Temperature (ºC)	–	–	32	31	35	32	33
pH	6.5–8.5	NR	8.20 ±0.04	7.85 ±0.02	7.83 ±0.06	8.30 ±0.03	7.86 ±0.08
Total hardness (mg L^{-1})	300	600	146.83 ±5.08	182.80 ±9.31	184.09 ±6.01	173.60 ±8.51	106.31 ±4.33
Dissolved oxygen (mg L^{-1})	>6	NR	7.9 ±0.03	8.44 ±0.05	8.48 ±0.03	6.91 ±0.02	8.20 ±0.06
Fluoride (mg L^{-1})	1.0	1.5	1.70 ±0.06	1.01 ±0.04	0.98 ±0.03	1.88 ±0.05	3.42 ±0.09

The average temperature of the groundwater samples ranged from 31–35ºC and the pH was slightly alkaline varying between 7.83–8.30. The average total hardness of the groundwater was 106.31–184.09 mg L^{-1}. The dissolved oxygen ranged from 6.91–8.48 mg L^{-1}, which is above the minimum desirable limit (6 mg L^{-1}) and indicates the presence of a fairly good amount of oxygen in the groundwater. The average F concentration in the groundwater of the villages Khanta (1.70±0.06), Salahpur (1.88±0.05), and Usari (3.42±0.09) exceeded the permissible (1.5 mg L^{-1}) and the desirable limits (1.0 mg L^{-1}) for India while the concentration of F in the villages of Khudania (1.01±0.04) and Murai-tajpur (0.98±0.03) were less than the desirable upper limits.[13,29]

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The relationship between the F level in the drinking water found in the five villages and the desirable upper limit (1.0 mg F^{-1}) and the permissible upper limit (1.5 mg F^{-1}) for India are shown in Figure 2.

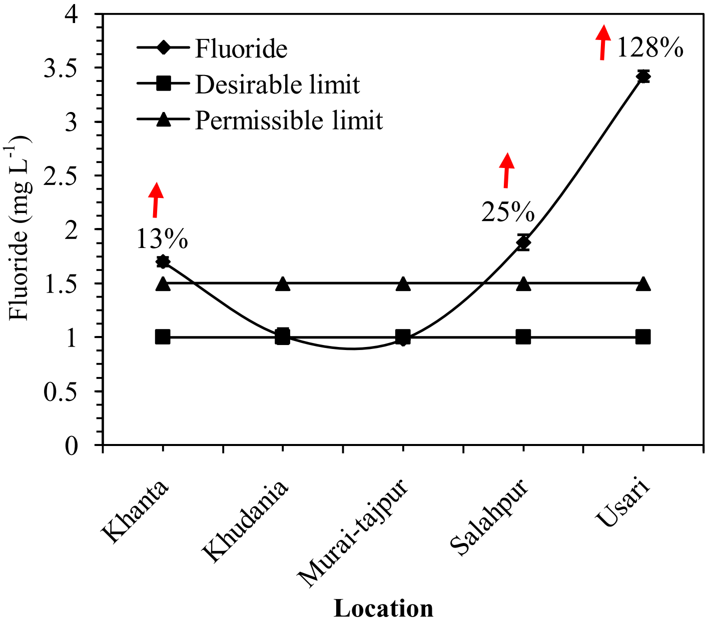

Figure 2. The relationship between the drinking water fluoride level in the five villages of Mitauli block, in district Lakhimpur (Kheri), in the state of Uttar Pradesh, India, and the desirable upper limit (1.0 mg F^{-1}) and the permissible upper limit (1.5 mg F^{-1}) for India.

The variations in the urinary F concentrations in the male and female residents are well explained by the derived model as shown in Figures 3A–3B to 7A-7B and Table 2. The observed data fitted well with the model with the highest value of r = 0.999 and S = 0.181 for male and r = 0.989 and S = 0.145 for female residents as shown in Figures 7A and 7B. The values of r ranged from 0.941–0.999 and of S from 0.009–0.181. The value of F_{fluo} is actually the peak value of urinary F. The values of F_{fluo} for the male residents, in the five villages of Khanta, Khudania, Murai-tajpur, Salahpur, and Usari were found to be 1.875, 1.476, 1.534, 2.283, and 4.481 mg L^{-1}, respectively, while for the female residents the corresponding observed values were 1.792, 1.480, 1.739, 1.976, and 4.461 mg L^{-1}. The model and the observed values of F_{fluo} are identical. The values of ‘b’ against all fittings, the translation distance by which the peak of the normal distribution curve is shifted rightward, were 31.092, 29.779, 35.546, 38.228, and 34.610 for the male residents and 34.155, 30.948, 37.922, 30.850, and 39.773 for the female residents. The stretching factor ‘c’ for urinary F was found to be 30.235, 60.416, 42.191, 30.883, and 34.165 for the male residents and 31.997, 83.192, 35.323, 56.608, and 37.535 for the female residents.

The urinary F concentrations of the different age groups in the male and female residents were found to be within the ranges of 1.08–1.84 mg L^{-1} (Khanta), 1.34–1.50 mg L^{-1} (Khudania), 1.10–1.73 mg L^{-1} (Murai-tajpur), 1.21–2.23 mg L^{-1} (Salahpur), and 2.76–4.41 mg L^{-1} (Usari) as shown in Figures 3–7. A higher F concentration was found in the drinking-water (3.42±0.09 mg L^{-1}) and urine (2.76–4.48 mg L^{-1}) in the Usari village residents (Figures 2–7) as compared to the residents

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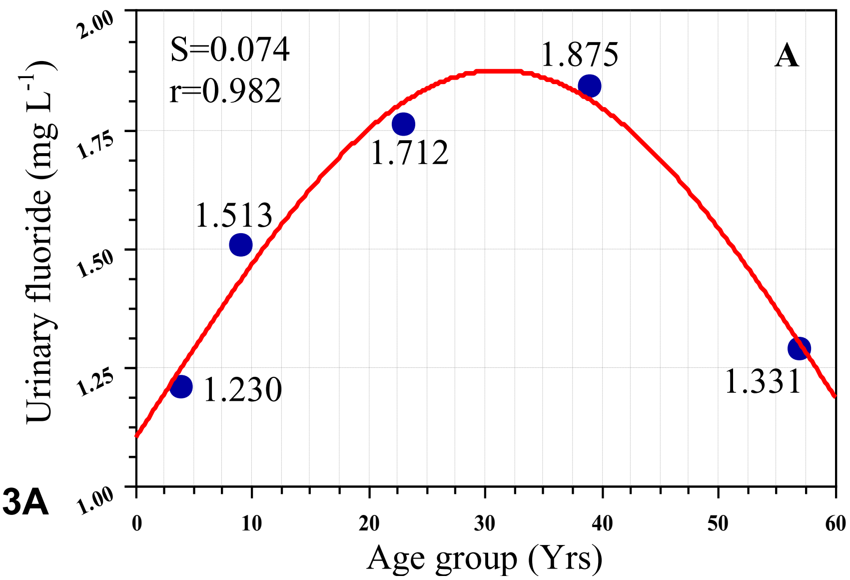
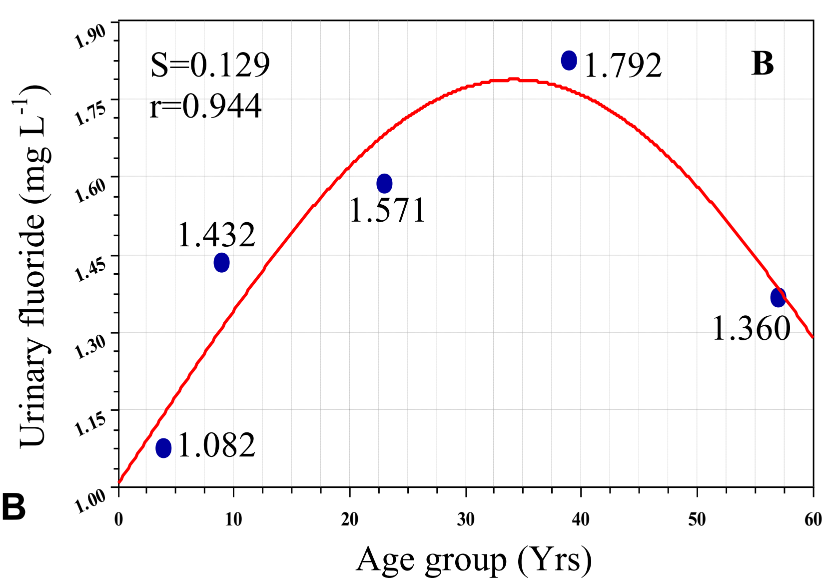
of the other villages. The model hypothesis successfully spelt out the variations of the urinary F with age for the male and female residents.

$$F_{fluo\_m} = 1.875^{e^{-\frac{1}{2}\left(\frac{t-31.092}{30.235}\right)^2}}$$



3A

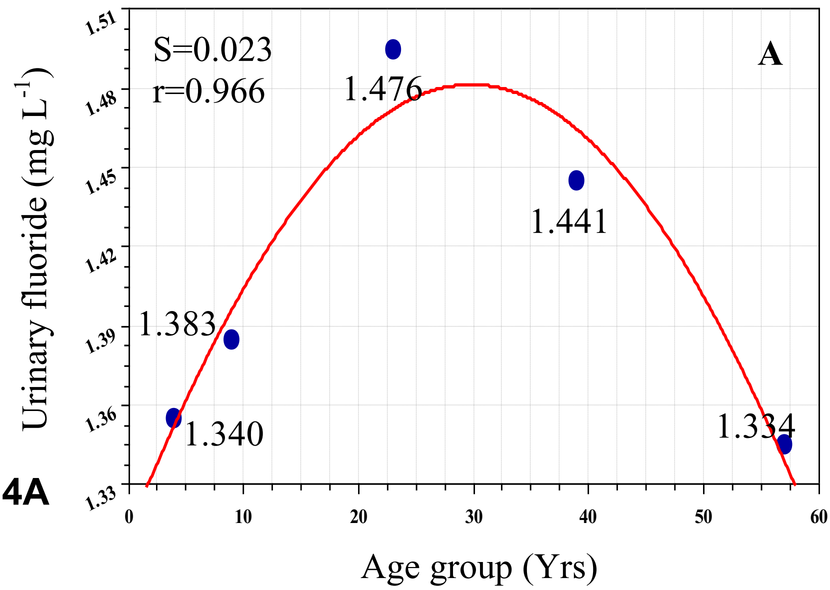
$$F_{fluo-f} = 1.792^{e^{-\frac{1}{2}\left(\frac{t-34.155}{31.997}\right)^2}}$$



3B

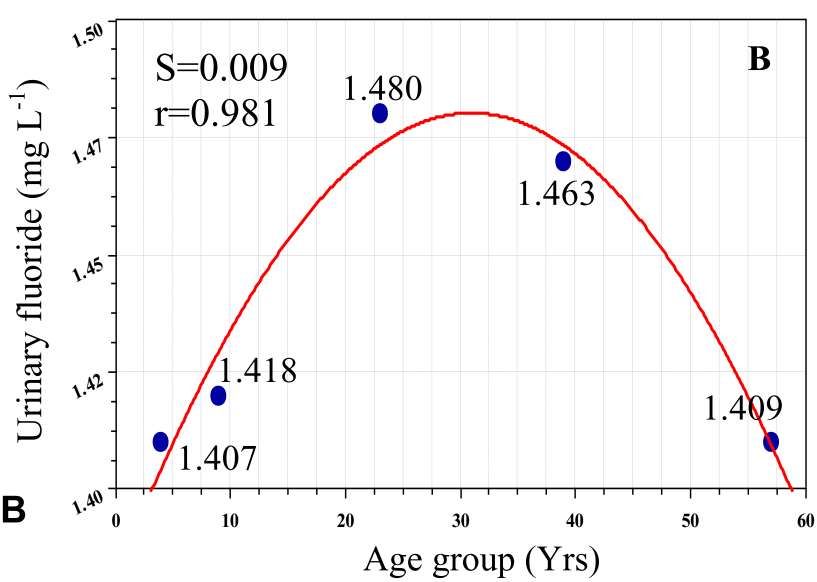
Figures 3A and 3B: The variation of the urinary fluoride concentration with age in the residents of Khanta village. 3A: male residents; 3B: female residents.

$$F_{fluo\_m} = 1.476^{e^{-\frac{1}{2}\left(\frac{t-29.778}{60.416}\right)^2}}$$



4A

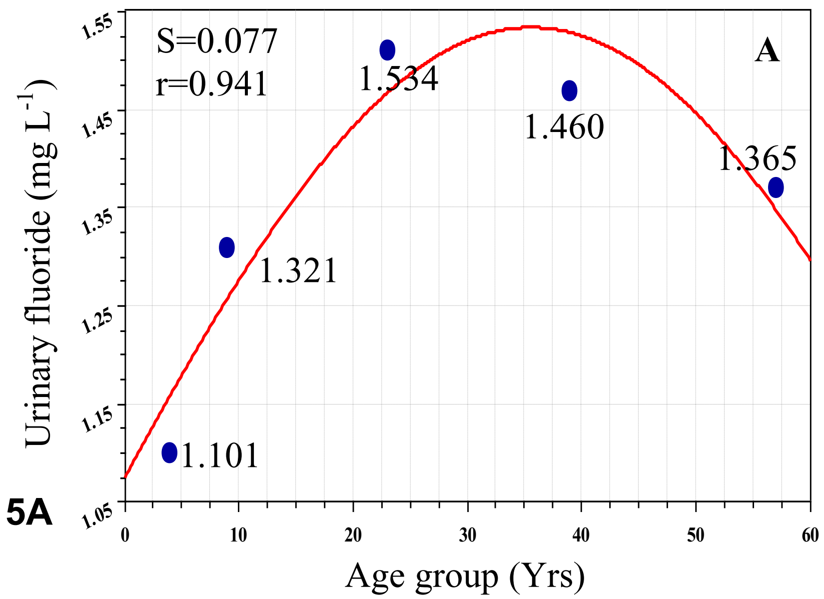
$$F_{fluo\_f} = 1.480^{e^{-\frac{1}{2}\left(\frac{t-30.948}{83.192}\right)^2}}$$



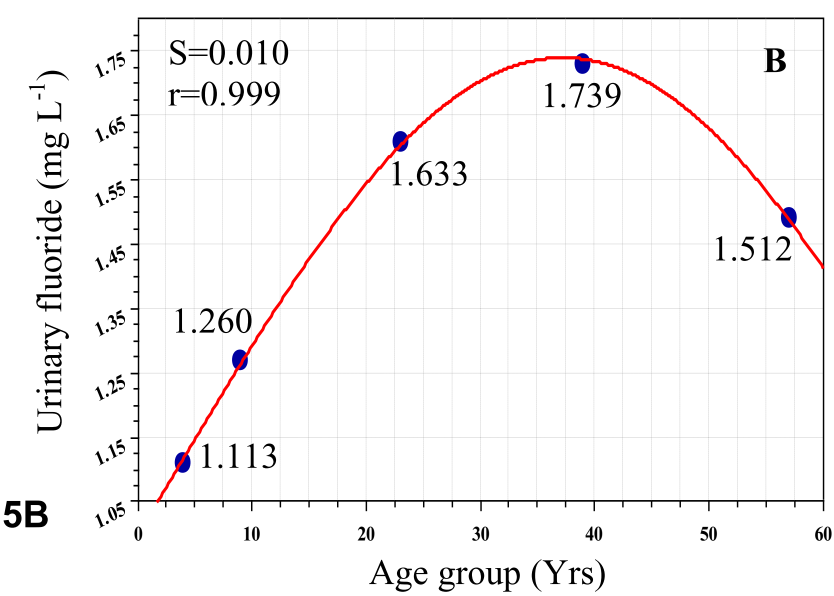
4B

Figures 4A and 4B: The variation of the urinary fluoride concentration with age in the residents of Khudania village. 4A: male residents; 4B: female residents.

$$F_{fluo}\_m = 1.534^{e^{-\frac{1}{2}\left(\frac{t-35.546}{42.191}\right)^2}}$$



$$F_{fluo}\_f = 1.739^{e^{-\frac{1}{2}\left(\frac{t-37.922}{35.323}\right)^2}}$$



Figures 5A and 5B: The variation of the urinary fluoride concentration with age in the residents of Murai-tajpur village. 5A: male residents; 5B: female residents.

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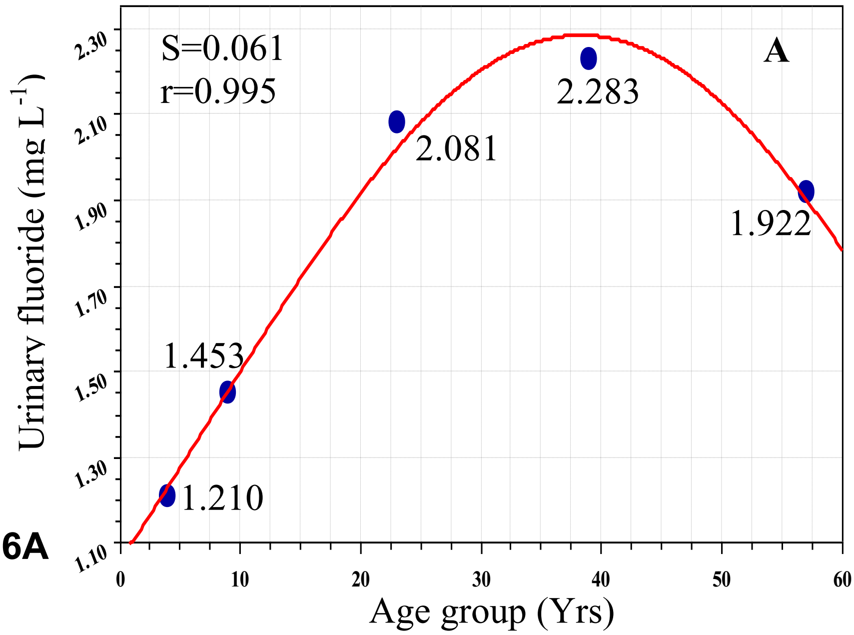
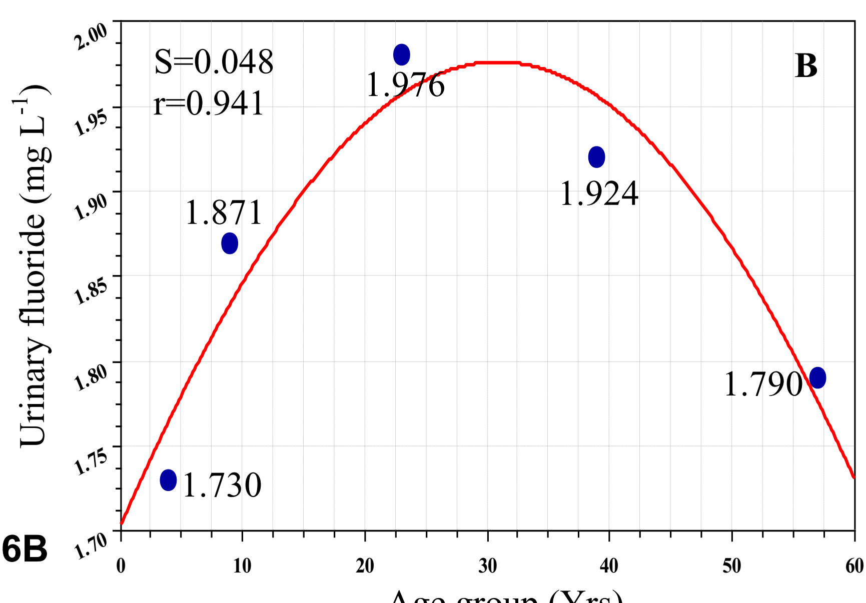
$$F_{fluo}\_m = 2.283^{e^{-\frac{1}{2}\left(\frac{t-38.288}{30.883}\right)^2}}$$

6A



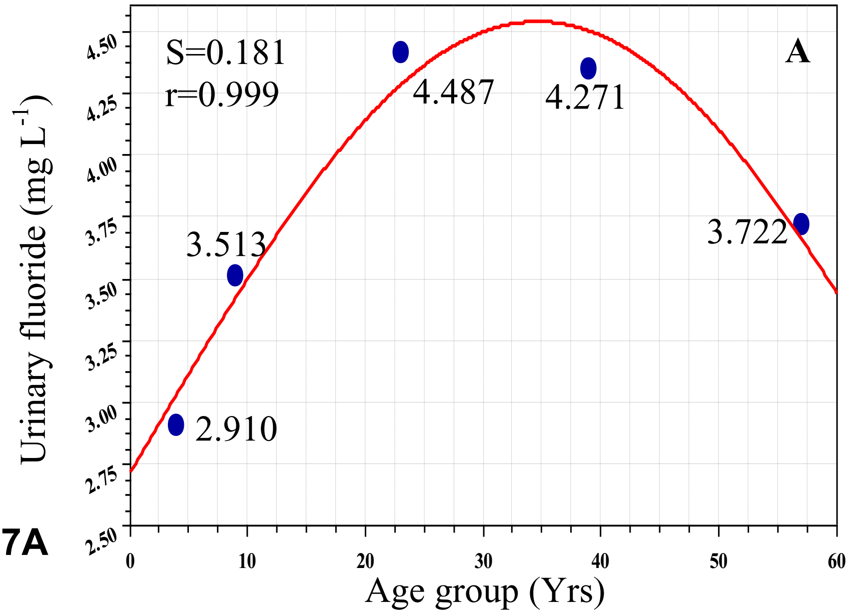
$$F_{fluo}\_f = 1.976^{e^{-\frac{1}{2}\left(\frac{t-30.850}{56.608}\right)^2}}$$

6B



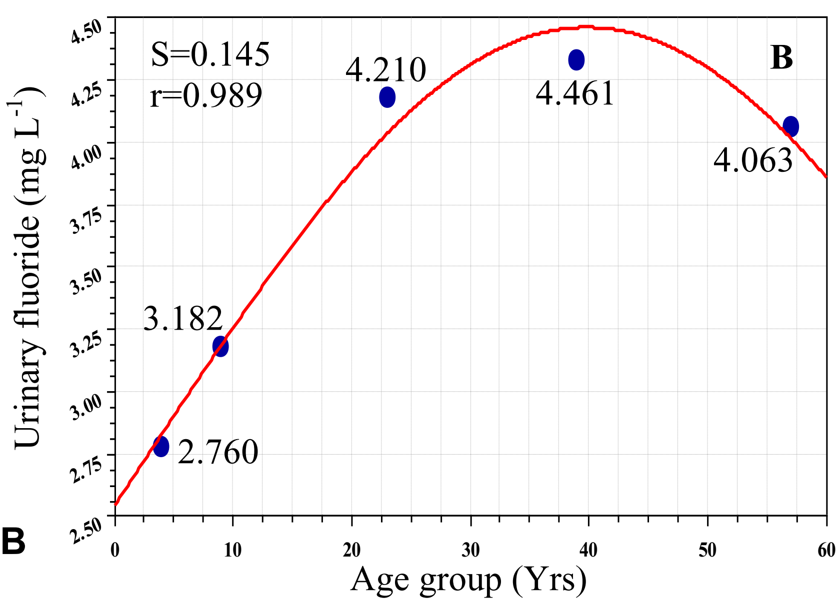
Figures 6A and 6B: The variation of the urinary fluoride concentration with age in the residents of Salahpur village. 6A: male residents; 6B: female residents.

$$F_{fluo}\_m = 4.537^{e^{-\frac{1}{2}\left(\frac{t-34.610}{34.165}\right)^2}}$$



7A

$$F_{fluo}\_f = 4.461^{e^{-\frac{1}{2}\left(\frac{t-39.773}{37.535}\right)^2}}$$



7B

Figures 7A and 7B: The variation of the urinary fluoride concentration with age in the residents of Usari village. 7A: male residents; 7B: female residents.

Table 2. Model parameters in the different villages (F_{fluo} = mg L^{-1})

Village	Category	F_{fluo}	b	c	r	S
Khanta	Male	1.875	31.092	30.235	0.982	0.074
	Female	1.792	34.155	31.997	0.944	0.129
Khudania	Male	1.476	29.778	60.416	0.966	0.023
	Female	1.480	30.948	83.192	0.981	0.009
Murai-tajpur	Male	1.534	35.546	42.191	0.941	0.077
	Female	1.739	37.922	35.323	0.979	0.010
Salahpur	Male	2.283	38.288	30.883	0.995	0.061
	Female	1.976	30.850	56.608	0.941	0.048
Usari	Male	4.481	34.610	34.165	0.999	0.181
	Female	4.461	39.773	37.535	0.989	0.145

DISCUSSION

Ailments such as skeletal deformation, weakened joints, and teeth mottling have been observed in a rural Indian population and attributed to being mainly due to the presence of a high concentration of F in the ground water.[30] Fluorine is the lightest and the most electronegative element and is extremely reactive with the most of the elements. It has an extraordinary tendency to attract positively charged ions such as calcium. Chronic exposure to F during the development of mineralized tissues, such as bone and teeth, may lead to serious developmental ailments.[31] Dental fluorosis may occur when the teeth are exposed to F during development, with the first 6 to 8 years of life appearing to be the critical period of risk.[32]

High concentrations of F in ground water are being reported continuously over the globe. However, the developing countries are at a greater risk of fluorosis due to poorer economies and dietary intakes. The problem is becoming increasingly worse due to the over exploitation of ground water. F concentrations in ground water in India have been reported up to 38.5 mg L^{-1} in endemic fluorosis areas.[2-6] Nearly, 42 and 60 million people are seriously affected by fluorosis in China and India, respectively.[33,34] Although the country standard limit for India of 1 mg L^{-1} has the rider the "lesser the fluoride the better, as fluoride is injurious to health,"[8] some consider that there is narrow range of F in drinking water, at approximately 0.6 mg L^{-1}, in which F is beneficial for human health.[18,35] The guidelines for the safe limit of F may vary from country to country depending on the dietary composition of the people. The WHO had set a limit of 1.5 mg L^{-1} while the Bureau of Indian Standards (BIS) norm prescribes only 1.0 mg L^{-1} of F in drinking water.[13,36] The severity of fluorosis has a relationship with the concentration of fluorine in drinking water and the duration of ingestion (exposure). The overall risk of fluorosis developing is the result of the duration of F exposure and the level of F ingestion through the use of medicaments and the consumption of the drinking water and foodstuffs which make up the composition of the daily diet.

The kidney is the principal vital organ for the excretion of F. The exposure levels of F can be checked by analyzing urinary samples.[37,38] The maximum F consumption has been found when the F concentration in drinking water is extremely high.[39,40] The F concentration in the drinking water in Khanta, Salahpur, and Usari villages was found to be higher than the recommended limit (1.5 mg L^{-1}) of the World Health Organization.[41] The exposure risk from using F contaminated drinking water is dependent on the age of the consumer and a knowledge of the relationship between the urinary F concentrations and age could be useful for understanding the pathogenicity of the F contaminated drinking water in humans. A mathematical model explaining the variation of F concentration in human urine with age could provide a basis for this understanding.

There is a great need to monitor the health of fluorosis affected people in the fluorosis endemic block of Mitauli of district Lakhimpur (Kheri), Uttar Pradesh, India. Investigations are needed in terms of ingestion, exposure, dietary composition, and precipitating health problems in different age groups in order to minimize the associated health hazards of F and to sustain the health of future generations.

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