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Toxicological Risk from Fluoride Exposure of a Population in Selected Areas of West Bengal, India

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Abstract: *This study was carried out to assess toxicological risk from the fluoride (F^-) exposure due to ingestion of drinking and vegetables/cereal crops such as mustard, rice and wheat, grown in potentially fluoridated area, of different age groups in Birbhum district, West Bengal, India. Among vegetables, maximum F^- concentration was found in spinach whereas in cereal crops, wheat accumulated more F^- than rice. With respect to both crops and vegetables the cumulative EDI values in children of 3–14 years are found to be higher as compared to the other age groups in both control and treated areas. Hazard risk assessment shows that adults are more susceptible to risk followed by children of 3 – 14 and 15 – 18 years. But in reference to crops/vegetables it is found that children of 3 – 14 years of age group are at a risk of developing fluorosis.*

Keywords*: Fluoride; Crops/vegetables; Exposure dose; Risk assessment*

I. Introduction

Fluoride (F^-) is an essential micronutrient for human beings, serving to strengthen the apatite matrix of skeletal tissues and teeth [1]. On the other hand, high F^- (>1.5 mg/L) results in dental and skeletal fluorosis; renal and neuronal disorders along with myopathy [2]. Fluoride is a lithophile element with atmophile affinities, and occurs in many common rock-forming minerals. Therefore, high F^- concentrations in water are expected in areas where fluorine-bearing minerals are abundant in the geologic substrate [3, 4, 5].

The disease 'fluorosis' has now become a global problem and the health impairment due to fluorosis has occurred in the citizens of about 25 nations across the globe, and more than 200 million people worldwide are at the risk of fluorosis [6]. It was thought previously that the principal source of F^- that causes fluorosis in human beings is the sources of drinking water. However, some food materials also contribute considerable amount to the total intake of F^- [7, 8, 9]. Several factors influence the level of fluorides in food. These include the sites in which the food is grown and whether there were sources of fluoride emissions in the area, the amount of fertilizer and pesticides applied and whether fluoridated water is used in cultivation and food preparation [10, 11, 12].

Fluoride is not having acute toxicity but its chronic toxicity if regularly consumed over an extended period of time can cause adverse effects. Fluoride toxicity depends for its severity on four factors *i.e.* total dose ingested, duration of fluoride exposure, nutritional status and body's response. In India very limited number of research works has carried out to evaluate the exposure assessment of F^- through different media like water, vegetable and crops in affected areas [12, 13].

Birbhum district (4,545 km^2) of West Bengal has been reported widely for the occurrence of F^- (1.5 - 17.9 mg/L) in the ground water, which is used for drinking as well as irrigating the vegetables and cereal crops [14, 15]. So far various research works have been done in the aspect of genesis of fluoride particularly in alluvial aquifer and quantitative accumulation of fluoride in the edible part of crops/vegetables [15, 16] in some endemic areas of the district. But no systematic research work has been done on the site specific exposure assessment of F^- intake through drinking water and crops/vegetables particularly in the potential fluoride contaminated area. So, in this study, an attempt was made to evaluate i) the site specific risk assessment due to F^- ingestion through drinking water as well as through vegetables and cereal crops grown with the aid of fluoride contaminated irrigation water in one of the endemic areas of the Birbhum district and ii) the risk to fluorosis in different age groups, using quantitative health risk assessment method.

II. Materials and Methods

2.1. Selection of Study area

Two adjacent villages namely Junidpur (24º 06' 07.5"N and 87º 46' 54.7"E) and Nowapara (24º 06' 18.0"N and 87º 47' 02.0"E) of Rampurhat –I block, Birbhum district are selected as a study area having an aerial coverage of 1.5 km^2. Western part of the Junidpur and Nowapara are contaminated with drinking water F^- (average F^- concentration 3.55 mg/L) [16]. Here villagers are not only using F^- contaminated groundwater as drinking purpose but also using it for agricultural purposes due to lack of awareness, education and overall non availability of alternate sources of water. The main occupations of local peoples are cattle farming and agriculture and there is no industrial enterprise in the vicinity of the study area. This area is chosen as a type area for this research because most of the villagers here not only exposed to F^- through drinking water but they also consume corps/vegetables, cultivated in their own agricultural plots with the aid of F^- contaminated irrigation water. Both dental and skeletal fluorosis is prevalent in this part of the study area [16]. So, in this research, this part of the study area is considered as a treated area. On the other hand, eastern portion of the Junidpur village has F^- concentration in groundwater within the [17] recommended limit of 1.5 mg/L. Thereby there is no chance of F^- input in crops/vegetables through irrigation water. That's why this portion of the study area is taken as control area (Figure. 1).

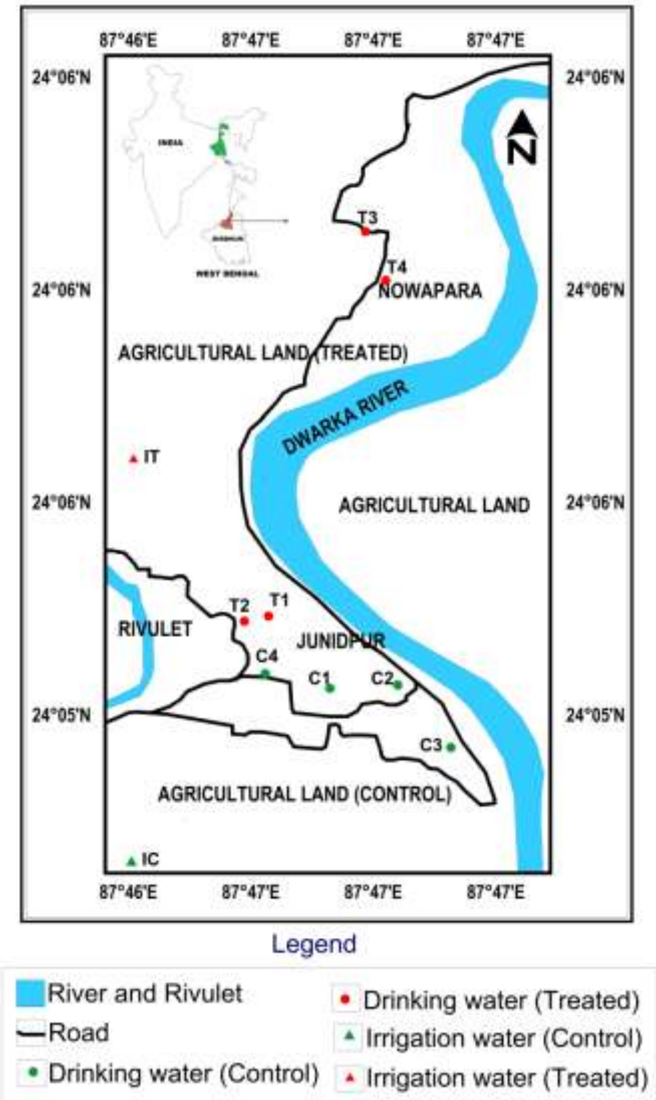

Figure 1: Map of the study area and sampling point on the Junnidpur and Nowapara area

In case of crop and vegetable, four (04) replicates were collected for each species and from each plot. The collected vegetables were washed with distilled water. Their edible part were then separated and further dried, chopped into pieces, and blended thoroughly. Next, 100-g samples of edible part were air-dried at 80ºC, ground, and passed through a 40 mm mesh sieve. The un-sieved material was sealed in polythene plastic bottles for further use. Similar kind of procedure was also followed for the husked rice, wheat and mustard.

2.2. Laboratory measurements

Water-soluble F (F_{H2O}) and total F (F_{Total}) from soil and crop/vegetables were determined by ion selective electrode technique (Orion 9609B NWP) followed by D'Alessandro et al., and McQuaker and Gurney [18, 19].

2.3. Nutritional Survey

The nutritional survey was conducted in the study area. Four households were selected each from control and treated areas in such a way so that the population was grouped into three categories according to their ages. The three age groups were children (3–14 years), children (15–18 years) and adults (19–70 years) with their average body weights taken as 25, 50 and 70 kg, respectively. A detailed dietary habit such as daily intake of drinking water, frequency of eating common vegetables and cereal crops such as rice and wheat were recorded. The survey was carried out by following the standard guidelines as prepared by National Institute of Nutrition (NIN), Hyderabad, India [20]. The survey was just based on the questionnaire and is not a bio-medical research in which human subjects are administered some medicine, injection or chemicals to study toxicity symptoms. This study satisfied all the criteria of ethical treatment of human subjects and kept the identity of human subject intact.

2.4. Moisture content in crops/vegetables

Fresh vegetable samples of about 50 g were chopped into pieces, and then pieces samples were first air dried for 2 days and then kept in a hot air oven at 70ºC - 80ºC for 3 days. The moisture percent was calculated using the following formula:

$$\text{Moisture }\% = \frac{(\text{W1} - \text{W2})}{\text{W1}} \times 100$$

Where W1 is the fresh weight of the vegetable and W2 is the weight of the vegetable after drying at 70ºC. Same procedure was also followed in case of crop. The values of the moisture content of crops/vegetables were required for the determination of average consumption of crops/vegetables on dry weight basis for each age group.

2.5. Exposure dose

The quantitative health risk assessment was measured by assessing the exposure doses of F^- due to the consumption of drinking water and various crops/vegetables in terms of estimated daily intake (EDI) using a generic equation [21]. The EDI with respect to water and crops/vegetables can be calculated as:

$$\text{EDI}_{\text{Water}} = \frac{C \times IR \times EF}{BW} \quad and \qquad \text{EDI}_{\text{Diet}} = \frac{C \times IR \times EF \times ED \times AF \times CF}{BW \times AT}$$

where EDI is the estimated daily intake (mg/ kg d), C is the F^- concentration in drinking water (mg/L) and crops/vegetables (mg/kg), IR the ingestion or intake rate (mg/ d), EF the exposure frequency (d/ year), ED the exposure duration (year), AF the absorption factor (unit less), CF the conversion factor (10^{-6} kg/ mg), BW the body weight (kg) and AT the averaging time (d). Here the absorption factor was taken as 75% and the body weights of children (3–14 years), children (15–18 years) and adults (19–70) years were taken as 25, 50 and 70 kg, respectively.

The site specific cumulative EDI was also calculated in the study area as follows:

$$\text{EDI}_{\text{Cumulative}} = \text{EDI}_{\text{water}} + \text{EDI}_{\text{vegetable}} + \text{EDI}_{\text{cereal crops}}$$

2.6. Risk assessment

The assessment of risk due to F^- exposure dose (EDI) in various age groups was also carried out by comparing with the reference dose of fluoride by determining the hazard index (HI), which is defined as the ratio of cumulative EDI to the reference dose (R_fD), represents total F^- intake risk:

$$\text{HI} = \frac{EDI_{Cumulative}}{R_f D}$$

R_fD is the reference dosage (oral toxicity reference value). It is 0.08 mg/kg/d for drinking water F^- [22] and 0.01 mg/kg/d for vegetable F^- [23].

2.7. Statistical analysis

Statistical analysis was carried out to determine the mean values and standard deviation (means ± SD) of the data of nutritional survey and fluoride concentration in soils and vegetables and cereal crops. Interrelationship among the analytical parameters was done by Pearson correlation co-efficient using XLSTAT v 2013 software.

III. Results and Discussion

3.1. Fluoride concentration in vegetables and cereal crops

The fluoride concentrations in vegetables and cereal crops such as rice, wheat, mustard and lentil were determined and are presented in Table 1.

Table 1: Fluoride concentration in vegetables and crops

Vegetables and cereal crops	Scientific name	Total F (mg/kg)
Vegetables		
Spinach C	*Spinacia oleracea*	47±2.160
Spinach T		55±2.944
Cabbage C	*Brassica oleracea Capitata*	22.6±2.570
Cabbage T		29.8±.787
Cauliflower C	*Brassica oleracea*	17.36±0.682
Cauliflower T		20.6±1.030
Potato C	*Solanum tuberosum*	29.6±0.966
Potato T		42±1.826
Carrot C	*Daucus carota sativus*	20±2.582
Carrot T		23.8±.258
Beet C	*Beta vulgaris*	17.36±.084
Beet T		20.6±.594
Cucumber C	*Cucumis sativa*	12.52±0.108
Cucumber T		18.64±0.360
Pea C	*Pisum sativum*	25.4±0.497
Pea T		27±1.826
Celeriac C	*Apium graveolens rapaceum*	29.6±.529
Celeriac T		42±2.160
Onion C	*Allium cepa*	24±2.582
Onion T		26.2±1.867
Onion (leaf)C		20.8±1.992
Onion T		31.6±.627
Garlic C	*Allium sativum*	8.26±.196
Garlic T		27.4±1.866
Cereal crops		
musur C	*Lens culinaris*	21.4±0.294
Musur T		25.2±0.337
Masturd C	*Brassica juncea*	20.8±0.572
Mastard T		43.6±0.572
WheatC	*Triticum sp*	15.06±0.127
Wheat T		28.4±0.497
Rice C	*Oryza sativa*	14.9±0.294
Rice T		17.44±0.029

n= 32 (16 each from control and treated field; No. of replicates for each sample is 4)

It was found that F^- concentration is higher in the vegetables and cereal crops grown under the treated area. The rate of F^- accumulation in vegetables and crops is in the order of spinach > celeriac > potato > cabbage > pea > onion > garlic > carrot > cauliflower > beet > cucumber and mustard > wheat > lentil > rice respectively in both control and treated condition. The range of F^- concentration in vegetable are varied between 8.26±0.19 mg/kg to 47±2.16 mg/kg in control condition and 18.64±0.36 mg/kg to 55±2.94 mg/kg in treated condition with the mean values are 22.88 and 30.55 respectively where as in case of crops the values varies between 14.9±0.29 mg/kg to 21.4±0.29 mg/kg with the mean 18.04 in control condition and 17.44±0.03 mg/kg to 43.6±0.57 mg/kg with the mean value 28.66 in treated condition. The results of higher fluoride accumulation in spinach (47±2.16 mg/kg in control and 55±2.94 mg/kg treated condition) may be due to the fact that spinach is known to be a good accumulator of fluoride [24]. It was also found that, F^- concentration higher in leafy vegetable rather than the other vegetables. Among the underground rooted vegetables potato is the higher accumulator of F^- in treated condition.

3.2 Nutritional survey

The season-wise nutritional survey among the population of different age groups children (3–14 years), children (15–18 years) and adults (19–70 years) was conducted in the study area and presented as mean fresh wt consumption/d (Table 2). The mean value of vegetable and crops intake in children 3–14 years, children 15–18 years and adults 19–70 years are taken as 52 g/d, 83 g/d, 135 g/d and 46 g/d, 63 g/d and 122 g/d, respectively. A wide variation in the consumption pattern of vegetable and cereal crops among population of different age groups was found in the area.

Table 2: Consumption pattern of vegetables and cereal crops among different age groups

Food items	**Per capita consumption (g/d)**					
	Children (3 -14 years)		**Children (15 -18 years)**		**Adult (19 – 70 years)**	
	Intake frequency (g_{fwt}/d)	**Duration intake/EF (d/year)**	**Intake frequency (g_{fwt}/d)**	**Duration intake/EF (d/year)**	**Intake frequency (g_{fwt}/d)**	**Duration intake/EF (d/year)**
Vegetables						
Spinach	58 (twice in a week)	2 months (16)	125 (twice in a week)	2 months (16)	223 (4 times in a week)	2 months (32)
Cabbage	83 (3 time in a week)	2 months (24)	133 (3 time in a week)	2 months (24)	206(4 time in a week)	2 months (32)
Cauliflower	75 (3 time in a week)	3 months (36)	150 (3 time in a week)	3 months (36)	225 (3 time in a week)	3 months (36)
Potato	116 (Daily)	12 months (365)	180 (Daily)	12 months (365)	234 (Daily)	12 months (365)
Carrot	30 (3 time in a week)	3 months (36)	50 (3 time in a week)	3 months (36)	75 (3 time in a week)	3 months (36)
Beet	32 (Once in a week)	3 months (13)	43 (Once in a week)	3 months (13)	60 (Once in a week)	3 months (13)
Radish	58 (5 time in a week)	3 months (60)	92 (5 time in a week)	3 months (60)	184 (5 time in a week)	3 months (60)
Radish leaf	40 (twice in a week)	2 months (16)	73 (twice in a week)	2 months (16)	146 (twice in a week)	2 months (16)
Cucumber	83 (5 time in a week)	4 months (80)	117 (5 time in a week)	4 months (80)	170 (5 time in a week)	4 months (80)
Tomato	48 (4 time in a week)	4 months (64)	60 (4 time in a week)	4 months (64)	110 (4 time in a week)	4 months (64)
Pea	58 (twice in a week)	2 months (16)	82 (twice in a week)	2 months (16)	130 (twice in a week)	2 months (16)
Celeriac	20 (4 time in a week)	2 months (32)	25 (4 time in a week)	2 months (32)	46 (4 time in a week)	2 months (32)
Onion	45 (5 time in a week)	12 months (240)	65 (5 time in a week)	12 months (240)	104 (5 time in a week)	12 months (240)
Onion leaf	31 (twice in a week)	2 months (16)	46 (twice in a week)	2 months (16)	98 (twice in a week)	2 months (16)
Garlic	8 (4 time in a week)	12 months (192)	15 (4 time in a week)	12 months (192)	28 (4 time in a week)	12 months (192)
Cereal crops						
Lentil	21 (4 time in a week)	12 months (192)	30 (4 time in a week)	12 months (192)	83 (4 time in a week)	12 months (192)
Mustard	33 (Daily)	12 months (365)	43 (Daily)	12 months (365)	78 (Daily)	12 months (365)
Wheat	50 (Daily)	12 months (365)	75 (Daily)	12 months (365)	142 (Daily)	12 months (365)
Rice	83 (Daily)	12 months (365)	106 (Daily)	12 months (365)	186 (Daily)	12 months (365)

The average consumption of vegetables and cereal crops on dry weight basis in different age groups was calculated on the basis of moisture contents and is presented in Table 3.

Table 3: Average consumption of vegetables and cereal crops on dry weight basis in different age groups

Vegetables and cereal crops	**Water content (g/100 g)**	**Age groups 3 -14 years**		**Age groups 15 – 18 years**		**Age groups 19 -70 years**	
		Average consumption (g_{fwt}/d)	**Average consumption (g_{dwt}/d)**	**Average consumption (g_{fwt}/d)**	**Average consumption (g_{dwt}/d)**	**Average consumption (g_{fwt}/d)**	**Average consumption (g_{dwt}/d)**
Vegetables							
Spinach	93.84	58	3.57	125	7.70	223	13.74
Cabbage	95.04	83	4.12	133	6.60	206	10.23
Cauliflower	92.02	75	5.98	150	11.96	225	17.95
Potato	86.57	116	15.58	180	24.17	234	31.43
Carrot	88.78	30	3.37	50	5.61	75	8.42
Beet	93.37	32	2.12	43	2.85	60	3.98
Radish	95.11	58	2.84	92	4.50	184	9.00
Radish leaf	90.26	40	3.90	73	7.11	146	14.22
Cucumber	96.52	83	2.89	117	4.07	170	5.92
Tomato	94.57	48	2.61	60	3.26	110	5.97
Pea	63.35	58	21.26	82	30.05	130	47.65
Celeriac	82.82	20	3.44	25	4.30	46	7.90
Onion	89.76	45	4.61	65	6.66	104	10.65
Onion leaf	93.55	31	2.00	46	2.97	98	6.32
Garlic	96.52	8	0.28	15	0.52	28	0.97
Cereal crops							
Lentil	10.19	21	18.86	30	26.94	83	74.54
Mustard	13.19	33	28.65	43	37.33	78	67.71
Wheat	6.73	50	46.63	75	69.95	142	132.44
Rice	11.24	83	73.67	106	94.09	186	165.09

* *g_{fwt}, g_{dwt} indicates fresh weight and dry weight respectively*

3.3 Exposure dose due to consumption of drinking water and crops/vegetables

The equation for the estimated daily intake (EDI) gives the dose of F^- that ingested into gastrointestinal tract through the daily consumption of different vegetable and crops in different age groups. For the purpose of health risk assessment, the exposure dose is more useful than the absorbed dose, as the absorbed dose is neither known for humans nor for animals. The assessment of the human health risk from ingestion of F^- from vegetables and cereal crops requires the information on quantities of F^- contaminated vegetables and cereal crops consumed and the extent of F^- contamination present in these food materials. The estimation of EDI also requires the information on absorption (or bio-availability factor). In this study the absorption factor was taken as only 75% to assess F^- exposure at the minimum level of absorption, whereas the absorption of 100% is considered only in those cases when the F^- is ingested as sodium fluoride tablets on fasting stomach. The estimated daily intake (EDI) of F^- through vegetables and crops were calculated to assess their exposures in different age groups. Assessment of the human health risk from ingestion of contaminated food requires information on the quantities of contaminated food supplements especially some common vegetables and crops were consumed and the extent of contamination present in foods. The cumulative mean EDI for drinking water and the different vegetables and crops in various age groups is presented in Table 4. The estimated daily intake (EDI) of F through drinking water was calculated to assess their exposures in different age groups. It was found that EDI in drinking water for children 3–14 years, 5–18 years and adults19–70 years were 0.0241, 0.0181 and 0.1676, 0.1275, 0.1795 mg/kg d in control and treated areas respectively. It was found that with respect to control adults of 19-70 years of age group in treated area are more exposed to drinking water F^- which may be due to higher intake of water and lower metabolic rate. Among children 3-14 years age group of treated area are more exposed in comparison to 15 – 18 years. This is because children of this age group frequently use drinking water after boiling this may give rise to more concentration of F^- [25].

Table 4: The cumulative mean EDI for drinking water and the different vegetables and crops among various age groups

	Age groups		
	3 -14 years	**15 – 18 years**	**19 -70 years**
		Control	
$EDI_{Vegetable}$	0.0026	0.0020	0.0022
EDI_{Crop}	0.0060	0.0041	0.0055
$EDI_{Drinking\ Water}$	0.0241	0.0181	0.0259
$EDI_{Cumulative\ Vegetable + Crop}$	0.0086	0.0061	0.0077
$EDI_{Cumulative\ Vegetable + Crop + drinking\ water}$	0.0328	0.0242	0.0336
$HI_{Vegetable + Crop}$	0.8584	0.6075	0.7698
$HI_{Vegetable + Crop + drinking\ water}$	0.3017	0.2262	0.3232
		Treated	
$EDI_{Vegetable}$	0.0029	0.0023	0.0025
EDI_{Crop}	0.0079	0.0055	0.0074
$EDI_{Drinking\ Water}$	0.1676	0.1275	0.1796
$EDI_{Cumulative\ Vegetable + Crop}$	0.0108	0.0077	0.0099
$EDI_{Cumulative\ Vegetable + Crop + drinking\ water}$	0.1784	0.1334	0.1895
$HI_{Vegetable + Crop}$	1.0820	0.7711	0.9889
$HI_{Vegetable + Crop + drinking\ water}$	2.0950	1.5713	2.2446

The mean concentration of F^- in vegetable and crops can be used to calculate EDI. Information on individual vegetable and crops consumption history in under study areas has collected by verbal questionnaire, based on the interviews of eight selected families. It was found that cumulative EDI in vegetables for children 3 – 14 years,15 –18 years and adults 19 –70 years were 0.0025, 0.0020, 0.0021 and 0.0029, 0.0023, 0.0025 mg/kg d in control and treated areas respectively, where as in crops, the cumulative EDI is 0.0060, 0.0041, 0.0055 and 0.0079, 0.0054 and 0.0074 respectively. With respect to both crops and vegetables the cumulative EDI values in children of 3–14 years are found to be higher as compared to the other age groups in both control and treated areas. It was also found that the exposure from the crop (rice and wheat) in all the age groups was more than the exposure due to consumption of vegetable. This was mainly because of the fact that per capita consumption (g dwt/d) for cereal crop is more as compared to vegetables.

It was observed that the cumulative EDI values in children aged 3–14 years are found to be higher (0.011) than the limits laid down for children as total dietary intake by IOM [26]. However, the cumulative values in the children (15–18 years age group) are 0.0077 and adults (19–70 years) are 0.0099 (Table 4), EDI is lesser than the prescribed limits by both IOM and Hargreaves [26, 27]. The prescribed limit for adults by Hargreaves [27] is 1.5–4.0 mg F/d or 0.0214–0.0571mg/kg d. It was also found that exposure from the cereal crop (rice and wheat) in all the age groups viz., children (3–14 years), children (15–18 years) and adults (19–70 years) as more (0.0079 mg/kg d, 0.0055 mg/kg d and 0.0074 mg/kg d respectively) than the exposure due to consumption of vegetable (0.0029 mg/kg d, 0.0023 mg/kg d and 0.0025 mg/kg d respectively).

3.4 Risk assessment

The hazard index (HI) was calculated for various age groups in the study area, as shown in Table 4. It was found that in comparison to the HI of F^- through drinking water in control area age group the treated area has higher HI (>1) among all the studied age group. On the basis of these findings adults are more susceptible to risk followed by children years 3 – 14 and 15 – 18 years. But in reference to crops/vegetables it is found that children of 3 – 14 years of age group are at risk of developing fluorosis. The other age groups are not at the risk of developing fluorosis in the study areas. In addition to high level of F^- in drinking water and crops/vegetables other problems especially poverty, malnutrition, low facilities of medication and presence of other toxicant in food stuff and drinking water, which may have synergistic effects on the health of people of the study area. This hypothesis is corroborated with the findings in other areas [28, 29, 30, 31].

IV. Conclusions

The exposure doses of F^- obtained by the consumption of crops were found to be higher in lower age group 7–15 years than elders. The hazard index (HI) calculation also obtained that the children 3-14 years more affected than the other age groups. So the youngsters are more likely at the risk of fluorosis via consumption of contaminated vegetable and crops in the studied area.

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References

[1] P.B. Maithani, R. Gurjar, R. Banerjee, B.K. Balaji, S. Ramachandran and R. Singh, Anomalous fluoride in ground water from western part of sirohi district, Rajasthan and its crippling effects on human health, Current Science, 74, 1998, 773–777.

[2] S. Ayoob, and A.K. Gupta, Fluoride in drinking water a review on the status and stress effects, Critical Reviews in Environmental Science and Technology, 36, 2006, 433–487.

[3] K. Tirumalesh, Isotope hydrochemical approach to understand fluoride release into groundwaters of Ilkal area, Bagalkot district, Karnataka, India, Hydrogeology Journal, 15, 2006, 589–598.

[4] E. Shaji, J. Bindu, Viju and D. S. Thambi , High fluoride in groundwater of Palghat District, Kerala, Current Science, 92, 2007, 240–245.

[5] S. Naseem, T. Rafique, E. Bashir, M.I. Bhanger, A. Laghari and T.H. Usmani, Lithological influences on occurrence of high-fluoride groundwater in Nagar Parkar area, Thar Desert, Pakistan, Chemosphere, 78(11), 2010, 1313–1321.

[6] United Nation International Children Education Fund *(*UNICEF), States of the art report on the extent of fluoride in drinking water and the resulting endemicity in India, Report by Fluorosis and Rural Development Foundation for UNICEF, New Delhi, 1999.

[7] L. Singer, and R. Ophauge, Total fluoride intake in children, Pediatrics, 63, 1979, 460–466.

[8] V. Singh, M.K. Gupta, P. Rajwanshi, S. Srivastava and S. Dass, Studies on ingestion of fluoride through tobacco pan masala and toothpaste, Indian Journal of Environmental Health, 35, 1993, 215–220.

[9] S. Gupta, and S. Banerjee, Fluoride accumulation in crops and vegetables and dietary intake in a fluoride endemic area of West Bengal, Fluoride, 44, 2011, 153–157.

[10] F.J. McClure, Fluorine in foods. Public health reports. U.S. Public Health Service. Public Health Reports, 64, 1949, 1061–1074.

[11] H.M. Myers, Fluorides and dental fluorosis. Monogr, Oral Science, 7, 1978, 1–76.

[12] S.K. Jha, A.K. Nayak and Y.K. Sharma, Site specific toxicological risk from fluoride exposure through ingestion of vegetables and cereal cropsin Unnao district, UttarPradesh, India, Ecotoxicology and Environmental Safety, 74, 2011, 940–946.

[13] A. Amalraj, and A. Pius, Health risk from fluoride exposure of a population in selected areas of Tamil Nadu South India, Food Science and Human Wellness, 2, 2013, 75–86.

[14] Central Groundwater Board (ER)(CGWB), Hydrogeology and groundwater resources of Birbhum district,West Bengal,Technical report, 1985, series D, No. 30.

[15] S. Gupta, and S. Banerjee, Fluoride accumulation in paddy (*Oryza sativa*) irrigated with fluoride-contaminated groundwater in an endemic area of the birbhum district, west Bengal, Fluoride, 42, 2009, 170–173.

[16] D. Mondal, S. Gupta, D.V. Reddy and P. Nagabhusanam, Geochemical controls of fluoride concentrations in groundwater from alluvial aquifer of the Birbhum district, West Bengal, India. Journal of Geochemical Exploration, 145, 2014, 190–206.

[17] World Health Organisation (WHO) (2004). Fluoride in Drinking Water - Background Document for Development of WHO Guidelines for Drinking Water Quality, Geneva.

[18] A. D'Alessandro, S. Bellomo, and F. Parello, Fluoride speciation in top soils of three active volcanoes of sicily (Italy). Environmental Geology, 56, 2008, 413–423.

[19] N.R. McQuaker, and M. Gurney, Determination of total fluoride in soil and vegetation using an alkali fusion–selective ion electrode technique, Analytical Chemistry, 49, 1977, 53–56.

[20] Thimmayamma, B.V.S. and Rau, P. (1987). Diet survey methods. In: Thimayamma, B.V.S. (Ed.), A Handbook of Schedule and Guidelines in Socio-Economic and Diet Survey, National Institute of Nutrition, Indian Council of Medical Research (pp. 1–8).

[21] United State Environmental Protection Agency (USEPA), Guidelines for exposure assessment. Federal Register Notice 57, Environmental Protection Agency (United State, 1992), 22887–22938.

[22] Environmental Protection Agency (EPA), Fluoride: Dose-Response Analysis for Noncancer Effects. EPA-820-R-10-019, Office of Water, U.S. Environmental Protection Agency, 2010b.

[23] R.K. Rattan, S.P. Datta, P.K. Chhonkar, K. Suribabu, and A.K. Singh, Long-term impact of irrigation with sewage effluents on heavy metal content in soils, crops and groundwater—a case study. Agriculture, Ecosystems and Environment, 109, 2005, 310–322.

[24] R. Sheldrake, E.D. George, E. Leigh, E. St John Jr, and J.L. Donald, Lime and charcoal amendments reduced fluoride absorption by plants cultured in perlite peat medium, Journal of the American Society for Horticultural Science, 103(2), 1978, 268–270.

[25] Martinez-Mier, E.A. Fluoride: Its metabolism, toxicity, and role in dental health, Journal of Evidence-Based Complementary and Alternative Medicine. 1–5, 2011. Doi: 10.1177/2156587211428076.

[26] Institute of Medicine (IOM), Dietary Reference Intakes for Calcium, Magnesium, Vitamin D, and Fluoride, National Academy Press, , Washington, DC, 1999.

[27] J.A. Hargreaves, Water fluoridation and fluoride supplementation: consid- erations for the future, Journal of Dental Research, 69, 1990, 765–770.

[28] T. Roychowdhury, T. Uchino, H. Tokunaga, and M. Ando, Survey of arsenic in food composites from an arsenic-affected area of West Bengal, India, Food and Chemical Toxicology, 40, 2002, 1611–1621.

[29] H.K. Das, A.K. Mitra, P.K. Sengupta, A., Hossain, F. Islam, and G.H. Rabbani, Arsenic concentrations in rice, vegetables and fish in Bangladesh: a preliminary study, Environment International, 30, 2004, 383–387.
[30] M.B. Arain, T.G. Kazi, J.A. Baig, M.K. Jamali, H.I. Afridi, A.Q. Shah, N. Jalbani, and R.A. Sarfraz, Determination of arsenic levels in lake water, sediment, and foodstuff from selected area of Sindh, Pakistan: estimation of daily dietary intake. Food and Chemical Toxicology, 147, 2009, 242–248.
[31] J.A. Baig, T.G. Kazi, A.Q. Shah, H.I. Afridi, G.A. Kandhro, S. Khan, N.F. Kolachi, S.K. Wadhwa, F. Shah, M.B. Arain, and M.K. Jamali, Evaluation of arsenic levels in grain crops samples irrigated by tube well and canal water, Food and Chemical Toxicology, 49, 2011, 265–270.