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A Variance Decomposition Approach for Risk Assessment of Groundwater Quality

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Abstract

This research focuses on the assessment of fluoride doses in groundwater adopting the mathematical model employed by the USEPA. A total of 456 groundwater samples were tested to assess the spatial distribution of fluoride contamination in the study areas. Three age groups (children, teens and adults) were selected for two-way pathway exposure (potential dose and dermal dose) assessment. For uncertainty and sensitivity of inputs variables, a new emerging Sobol sensitivity analysis (SSA) technique was used to determine the relative importance of inputs using Monte Carlo simulation. Three types of effects, first-order effect (FOE), second-order effect (SOE) and total effect (TE) were calculated. The results showed that 96% of the samples analysed were within the standard acceptable level (1.5 mg l^{-1}) of WHO guidelines. The spatial distribution depicts that the eastern and south-eastern parts of the study area have the higher concentrations with the few spots of elevated concentration in the middle of the north and the south-west areas. The mean value of Hazard Index for children in the study region is less than 1, whereas the 95th percentile exceeded the value of 1 for both children and teens. The FOE shows the concentration of fluoride (C_w) is highly sensitive followed by exposure frequency (EF), intake rate (IR_w) and body weight (BW). The SOE scores revealed that IR_w–BW are the most important input parameters for the assessment of oral health risk. For the dermal model, the highest value of Sobol score was recorded for interactions C_w–SA for adults followed by teens and children. Further, the results show that the older-age groups have more dermal risk than the younger-age groups. The research explores the feasibility of SSA technique to investigate the effects of individual input parameters for health risk model and whether it can be applied to another contaminant.

Keywords Groundwater · Sobol sensitivity analysis · Fluoride · Mid-Gangetic plain

Introduction

Fluoride is ubiquitous in the environment in the form of fluoride ion in groundwater system, whether due to anthropogenic or natural addition (IPCS 2012). Poor water quality contributes to approximately 80% of the world diseases, 65% of which is contributed by endemic fluorosis alone (Adimalla and Venkatayogi 2017; Felsenfeld and Robert 1991; Karami et al. 2017; Miri et al. 2016; Narsimha and Sudarshan 2017; WHO 2011). In a developing country like India, more than 33% of the drinking resources are not suitable for consumption (Cronin et al. 2014). The fluoride contamination is affecting more than 400 million populations in the districts of Bihar, Rajasthan, Madhya Pradesh and West Bengal states of India (Chakraborti et al. 2010; Cronin et al. 2014).

In groundwater system, fluoride concentration depends upon various water-quality parameters such as pH, total solids, alkalinity and hardness (Baghania et al. 2017; Dehghani et al. 2017; Karthikeyan and Shunmugasundarraj 2000; Rostamia et al. 2017; Subba Rao et al. 1998). Samal et al. (2015) have investigated the fluoride contamination in two districts of West Bengal in water sources and agricultural soils and they had found insignificant association of total hardness and phosphate in relation with the fluoride. Furthermore, they have concluded high possibility of bioaccumulation of fluoride in the cultivated crops from contaminated soil and

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water. Marya et al. (2014) have determined the relationship between dental caries and dental fluorosis at different levels of fluoride in drinking water. From the analysis of results, they have found 1.13 ppm as the optimal level of fluoride with maximum caries reduction and minimum amount of aesthetical objection due to fluorosis. Kumar et al. (2017) have investigated the relationship between fluoride in water, urine, serum and fluorosis and found significant correlation between water, urine and serum fluoride. Edmunds and Smedley (2013) have investigated the association between fluoride, human bones and teeth considering fluoride ion existence. In addition, several studies by other researchers assessed its impact on human and plant considering biological, biochemical and clinical effects (Adimalla 2018; Adimalla et al. 2018a; Jacobson and Weinstein 1977; Li et al. 2018; NAS 1971; Ozsvath 2009; Singh et al. 1963).

Chilton et al. (2006) have stated that the fluoride-bearing minerals (local igneous and metamorphical rocks) are commonly found in Asia (India, Pakistan, Thailand, China and Sri Lanka) as well in African sub-continent (western and southern Africa). Li et al. (2014) have carried out hydrogeological investigation and estimated the fluoride concentration in confined aquifer underlying the first terrace of Weihe River in China. From the analysis of results, they have found fluoride enrichment in groundwater was controlled by geology especially fluoride-bearing minerals and hydrogeological conditions. They further stated that human intervention, ion exchange and mixing of different types of recharge water were the cause for fluoride enrichment.

In Indian context, granite rocks naturally contain fluoride-bearing minerals such as fluorite, apatite, biotite, mica and other minerals, which are the primary sources for fluoride (Cooper et al. 1991; Narsimha and Sudarshan 2017; Maithani et al. 1998; Jacks et al. 2005; Adimalla et al. 2018a, b). In groundwater system, high bicarbonate alkaline condition (pH range 7.5–8.6) is more conducive for fluoride dissolution (Saxena and Ahmed 2001). In addition, factors like temperature, pH, complexing and precipitating ions, cation exchange capacity and duration of water–rock interaction of minerals also influence the dissolution of fluoride in groundwater (Apambire et al. 1997). Narsimha and Sudarshan (2017) have investigated positive correlation between the pH and fluoride content in groundwater. In groundwater system, low calcium content and high fluoride concentration are found due to their low solubility (Apambire et al. 1997). In addition, sodium bicarbonate water type and high bicarbonate alkaline groundwater also ensure high probability of fluoride concentration.

It is a well-established fact that the ingestion of fluoride is associated with chemical toxicity that results in change of physical structure of bones (Miller et al. 1977; Riggs et al. 1990). Fluoride has dual effect (beneficial and detrimental effects) on the human health within narrow range. A small amount is needed to form bones, enamel and to prevent tooth decay, whereas high fluoride can adversely damage bones and teeth (Khorsandi et al. 2016; Petersen 2004; Podgorny and McLaren 2015). Some researchers reported destructive effects on the metabolism of soft tissues (kidney, liver and lungs) (Barbier et al. 2010; Yang and Liang 2011; Zhang et al. 2016) with tumbling intelligence quotient (IQ) in children (Tang et al. 2008). In addition, Bassin et al. (2006) and Choi et al. (2015) have reported possibility to induce skeletal cancer and neurotoxicological effects as a result of high fluoride concentration. Wu and Sun (2016) have investigated the groundwater fluoride contamination and potential risk associated to the local residents in alluvial plain of China. Both oral and dermal pathways were calculated due to intake of drinking of groundwater. From the analysis of results, they have found that children of alluvial plain (in China) were at the higher risk than the adults, and urgent and efficient measures are needed to take to reduce the health risk.

The World Health Organization had recommended a reference level 0.5 to 1.5 mg l^{-1} for fluoride and as per guidelines of US Public Health Service, the optimal concentration of fluoride in drinking water should be 0.7 mg l^{-1} (Kohn et al. 2001). Yadav et al. (2013) stated that more than 200 million people suffer from the deadly diseases called fluorosis due to intake of higher fluoride concentration in drinking water throughout the world. Developing countries have greater potential health consequences through intake of fluoride-containing drinking water (Adimalla 2018; Adimalla et al. 2018a, b; Huang et al. 2017; Li et al. 2018; Wu et al. 2015).

The objective of this study was to investigate concentration of fluoride in groundwater of mid-Gangetic plain in five districts of Bihar. After determination of fluoride concentration, a mathematical non-carcinogenic risk-assessment model proposed by USEPA was applied. Sobol sensitivity analysis (SSA) was applied to the Hazard quotient (HQ) model considering the three indices as first-order effect, second-order effect and total effect (TE) to the three age groups (children, teens and adults). For spatial distribution analysis of fluoride, ArcGIS 10.3 software was used.

Materials and methods

Study Area, Sampling and Analysis

Bihar state is situated in the mid-eastern region of India. Figure 1 shows location of the sampling districts in Bihar as the highlighted part as well as the geographical location of the state of Bihar in India. Aurangabad, Gaya, Jahenabad, Nalanda and Nawada lie in west-south Bihar (WSB) with geographical locations between 24°17′19.75″N to 25°27′40.91″N and 83°59′54.16″E to 86° 3′25.42″E

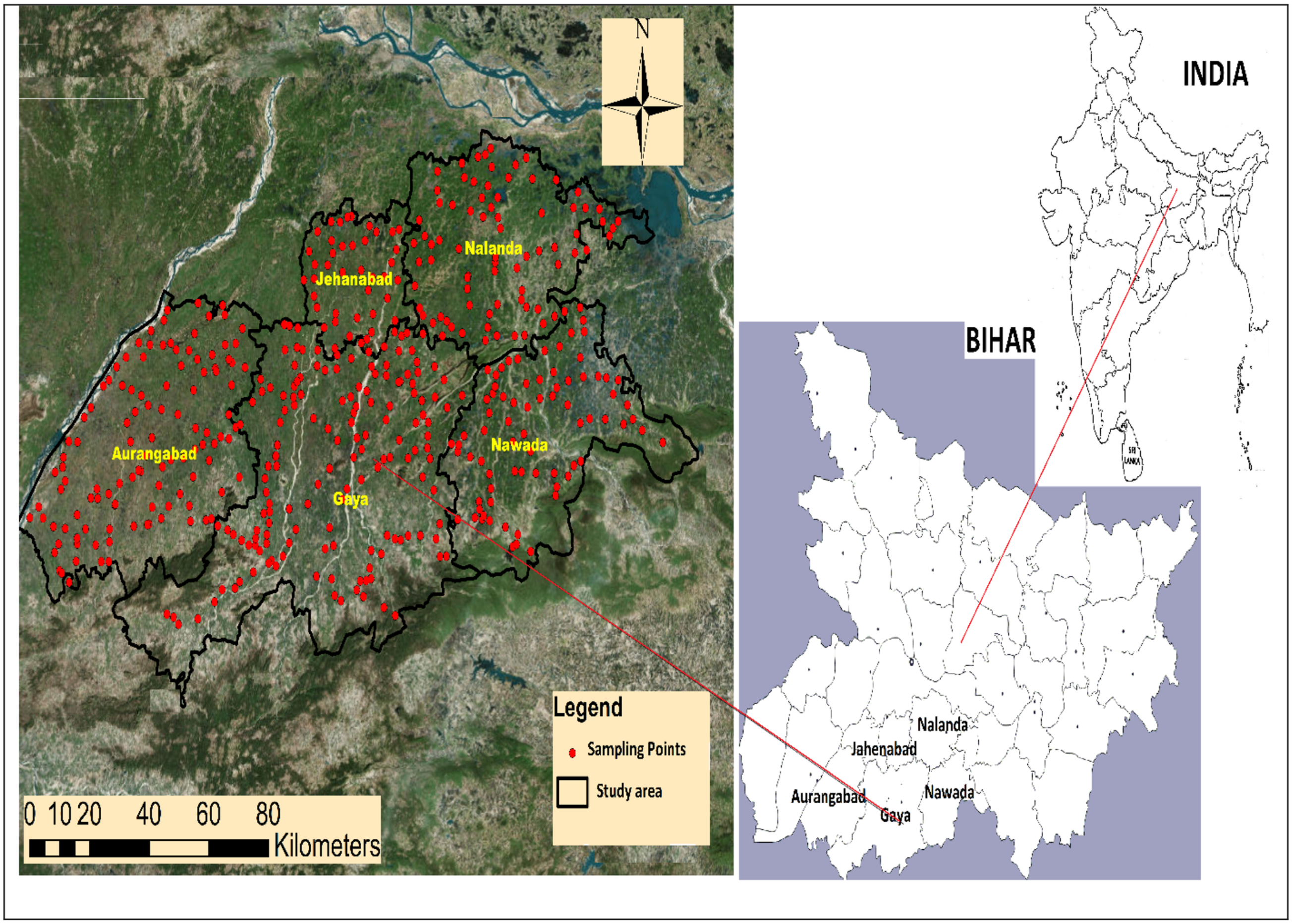

Fig. 1 Study area with Sampling Location

longitude covering the total area of 14,711 square kilometres. Sampling area is a region of the alluvial plain, situated in the southern portion of river Ganges. This area is underlain by recent unconsolidated alluvial deposits, which primarily consist of gravels, sand, silt and clay. Bedrock of this study area is composed of weathered granites with genesis and schists. The groundwater yield of the area has large variability that ranged from 5 to 200 cu m h^{-1}. In addition, the groundwater depth varies from 2 to 14 m below the ground level. The major activity in this area is agriculture with minor mineral exploration. The major industries found in this area include stone quarries, rice mills and sugar factories (Kumar et al. 2018).

A total of 456 representative groundwater samples were collected from underground sources across the study area by adopting 6 × 5 km grid size. The groundwater sources were readily available for consumption as drinking water. Sampling site, geo-positions (latitude and longitude) were recorded using Global Positioning System (model: Garmin GPS 72H). For each location, 1-L high-density polyethylene (Thermo Scientific, 1131000BPC) sample bottles were treated with dilute nitric acid overnight and rinsed with ultrapure water (Milli-Q Ultrapure Water Purification System, Model Z00QSVC01). Sample Bottles were washed thoroughly with the sampled bore well/hand pumps water before collecting the water samples. Samples were collected during the pre-monsoon season from marked locations (Fig. 1). Soon after collection at the site, in situ water-quality parameters, such as pH and conductivity, were estimated in the field using a portable instrument (Thermo Scientific Orion™ 9609BNLSLN). Collected groundwater samples were filtered using 0.45-µm membrane filter (syringe filters). To avoid wall deposition, a portion of the samples was acidified after filtration. To avoid the matrix decomposition, the non-acidified samples were stored at 4 °C in dark until completion of analysis (APHA 2005).

Fluoride Estimation Using Ion-Selective Electrode (ISE)

Fluoride concentration of the groundwater samples were determined using fluoride ISE (Thermo Scientific Orion™

9609BNLSLN) in Thermo Scientific Orion VERSA STAR pH/ISE/Conductivity/RDO/Dissolved Oxygen Multiparameter Meter Kit. The reagents and redistilled water used for the solution preparation (or dilution) were, respectively, of analytical grade (Merk Millipore) and highest-purity milli-pore water. Prior to measurement of the unknown sample, the fluoride ISEs were calibrated by 0.1 to 5 mg l^{-1} standard solution prepared from 0.5 mg l^{-1} and 5.0 mg l^{-1} standard with TISAB II as per instructions provided by the vendor. The slope value for calibration curve was found to be −58 mV/(mg l^{-1}) at 23 °C. A Teflon-coated stirrer was placed in 100-ml plastic sampler having 50 ml of sample (25 ml standard solution/groundwater sample + 25 ml TISAB II), which was subjected to stirring at constant rate. After dipping the Fluoride ISE (9609BNWP) into the standard solution/groundwater sample, and once reading became stable, the concentration was recorded. All protocols followed for the analysis were as per standard methods prescribed for the examination of water and wastewater (APHA 2005).

Health-Risk Assessment Model

To assess the health risk for the populations of the mid-Gangetic plain, three groups [children (3–10 years old); teens (11–20 years old); adults (21–72 years old)] were considered. The exposure to fluoride-contaminated groundwater was evaluated in terms of potential dose and dermal dose, which were calculated using the Eqs. (1) and (2) introduced by USEPA (USEPA 1989; Li et al. 2016).

$$\mathrm{ADD}_{\mathrm{ingestion}} = \frac{C_{\mathrm{w}} \times \mathrm{IR}_{\mathrm{w}} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \tag{1}$$

$$\mathrm{ADD}_{\mathrm{dermal}} = \frac{\mathrm{C}_{\mathrm{w}} \times \mathrm{SA} \times K_{\mathrm{p}} \times F \times \mathrm{ET}_{\mathrm{S}} \times \mathrm{EF} \times \mathrm{ED} \times 10^{-3}}{\mathrm{BW} \times \mathrm{AT}}, \tag{2}$$

where ADD $_{\mathrm{ingestion}}$ denotes the average daily potential dose of fluoride ingested through drinking groundwater. The value $\mathrm{ADD}_{\mathrm{dermal}}$ calculates the amount of fluoride received by skin absorption in mg kg^{-1} day^{-1}. The C_{w} denotes concentration of fluoride in drinking water (mg l^{-1}), IR_{w} is daily intake rate of water (l day^{-1}), EF is exposure frequency (day year^{-1}), ED is the exposure duration (year), BW is the body weight (kg), AT is the averaging time (Days), SA is surface area of skin to get exposure (cm^2), K_{p} is coefficient of permeation (cm h^{-1}), F is the fraction of contact surface within skin and water, and ET is the exposure duration to shower (h day^{-1}).

The non-carcinogenic risk due to exposure of fluoride through groundwater is estimated in terms of Hazard quotient (HQ) for both ADD $_{\mathrm{ingestion}}$ and ADD $_{\mathrm{dermal}}$ using Eq. (3) (He and Wu 2018; He et al. 2018).

$$\mathrm{HQ} = \frac{\mathrm{ADD}_{\mathrm{ingestion.}} \text{ or } \mathrm{ADD}_{\mathrm{dermal}}}{\mathrm{R}f\mathrm{D}} \tag{3}$$

In this regard, R*f*D is the reference fluoride dose by specific pathway (mg kg^{-1} day^{-1}). According to USEPA's Integrated Risk Information system (IRIS), the R*f*D for oral and drinking ingestion is considered as 0.06 mg kg^{-1} day^{-1} (Huang et al. 2017). Reference dose for skin exposure is calculated by Eq. (4) based on USEPA conversation method from drinking $\mathrm{R}f\mathrm{D}_{\mathrm{w}}$ to $\mathrm{R}f\mathrm{D}_{\mathrm{dermal}}$ (Staff 2001).

$$\mathrm{R}f\mathrm{D}_{\mathrm{derm}} = \mathrm{R}f\mathrm{D}_{\mathrm{w}} \times \mathrm{ABS}_{\mathrm{gi}}, \tag{4}$$

where $\mathrm{R}f\mathrm{D}_{\mathrm{dermal}}$ and $\mathrm{R}f\mathrm{D}_{\mathrm{w}}$ are, respectively, the dermal and water reference doses, whereas $\mathrm{ABS}_{\mathrm{gi}}$ is the digestive absorption factor. The overall non-carcinogenic risk due to exposure of fluoride in groundwater is calculated in terms Hazard Index (HI) by Eq. (5)

$$\mathrm{HI} = \mathrm{HQ}_{\mathrm{overall}} = \mathrm{HQ}_{\mathrm{ing}} + \mathrm{HQ}_{\mathrm{derm}} \tag{5}$$

Sobol Sensitivity Analysis (SSA)

SSA is a global sensitivity analysis tool, in which all the input parameters are varied simultaneously over the whole input space with the increasing dimensionality (Saltelli et al. 1999; Sobol 1993). SSA evaluates the relative continuation of each influential input with their interaction to the variance of the model output. SSA is an innovative approach to understand which reaction and processes have more influence on the overall system. The basic framework of SSA is variance decomposition, which provides a quantitative measure of input contribution to the output variance using Monte Carlo integration. SSA tool is the one of the most powerful variance decomposition techniques compared to the available sensitivity analysis technique (weighted average of local sensitivity analysis, partial rank correlation coefficient, multiparametric sensitivity analysis, Fourier amplitude sensitivity analysis). It calculates the extent of variability in model output considering each input parameters as single and interaction among different parameters. SSA is not intended to identify the cause of the input variability. It just indicates the impact and the extent that the model output has. The basic feature of SSA is that it does not have any underlying assumption between the model input and output with full range of input variance and their interaction between parameters. The steps involved in SSA are depicted in brief in Fig. 2.

Consider the following model

$$Y = f(X_1, \ldots, Xp), \tag{6}$$

where $X_1,\ldots,X_{\mathrm{p}}$ represents the independent random input variable having known probability distribution and Y scalar output. The Sobol method decomposes the output variances into the contributed associations of individual input factors.

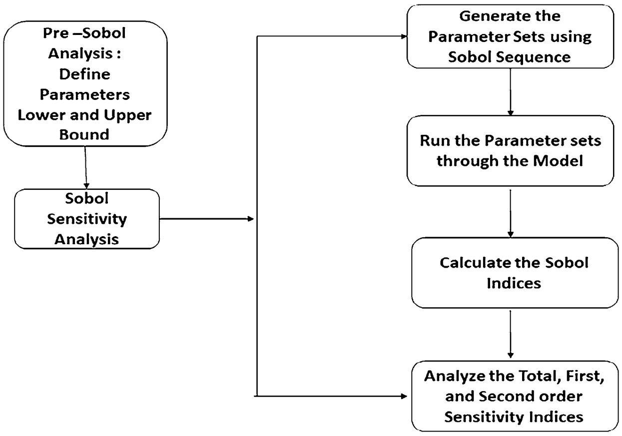

Fig. 2 The procedure flow chart of the Sobol sensitivity analysis (Zhang et al. 2015)

In order to estimate the influence of an input factor on the model output variance, we fix the value of X_i as x_i* and vary the other input variables $X_1, X_2, \ldots, X_p$ and then calculate the change in the variance of output (Y). This conditional variance is calculated using the Eq. (7).

$$V_{X-i}(Y|X_i = x *_i), \tag{7}$$

where the variance is taken over the ($p-1$) input parameter space X_{-1} consisting of all the inputs except the X_1. The basic framework of Sobol method's work based on law of total variance can be represented by Eq. (8).

$$V(Y) = V_{X_i}(E_{X_{-i}}(Y|X_i)) + E_{X_i}(V_{X_{-i}}(Y|X_i)) \tag{8}$$

After normalization, we have

$$1 = \frac{V_{X_i}(E_{X_{-i}}(Y|X_i))}{V(Y)} + \frac{E_{X_i}(V_{X_{-i}}(Y|X_i))}{V(Y)} \tag{9}$$

The first term in the Eq. (9) represents the first-order sensitivity index (FOSI) for the input X_i. i.e.

$$S_i = \frac{V_{X_i}(E_{X_{-i}}(Y|X_i))}{V(Y)} \tag{10}$$

Proceeding further through decomposition, it can be reduced to second order, third order and so on. The second-order sensitivity index represents the amount of variance of Y explained by interaction of the factor X_i and X_j (i.e. sensitivity to X_i and X_j) that is calculated based on Eq. (11).

$$S_{ij} = \frac{Vij}{V(Y)} \tag{11}$$

The total contribution to the output variance from the factor (first order plus all its interactions) is calculated through the total order sensitivity index (ST_i) proposed by Homma and Saltelli (1996), which is calculated using Eq. (12). The interested reader can find more details about the sensitivity analysis methodology in other studies (Saltelli et al. 1999; Sobol 1993; Zhang et al. 2015)

$$\mathrm{ST}_i = \sum_{k\#i} S_k$$

$$\mathrm{ST}_i = 1 - \frac{V_{X_{i-1}}\left(E_{X_i}\left(Y|X_{-i}\right)\right)}{V(Y)} \tag{12}$$

$$\mathrm{ST}_i = 1 - \frac{E_{X_{-i}}\left(V_{X_i}\left(Y|X_{-i}\right)\right)}{V(Y)}$$

Table 1 provides additional information on model parameters. The selection input parameters would be modelled as random variables (instead of fixed-value inputs). The models (HQ ingestion and HQ dermal) were developed in Python programming using "SALib 1.1.3 package".

Fluoride Spatial Distribution

Spatial distribution of fluoride was considered to allocate the potential elevated zone of fluoride using software ArcGIS 10.3. To map the fluoride zone, the inverse distance-weighted (IDW) method was used. IDW is a deterministic interpolation technique based on weighted mean of each parameter and the distance between the points. The basic assumption in this deterministic technique is that spatial features of points that are closer to each other are more alike than the features of those that are farther apart. The predicted value has been more influenced by the value of surrounding prediction location (Beg et al. 2011).

Results and Discussion

Analysis of Results and Spatial Distribution of Fluoride

Descriptive statistics of fluoride value of the groundwater samples of five districts are given in the Table 2. The fluoride concentrations in five districts varied from 0.1 to 3.6 mg l^{-1}. The highest fluoride value (3.6 mg l^{-1}) was recorded in the Nawada district followed by Gaya (2.3 mg l^{-1}), Aurangabad (2.1 mg l^{-1}), Jahenabad (1.6 mg l^{-1}) and Nalanda (1.5 mg l^{-1}). The permissible range for fluoride concentration in drinking water recommended by WHO (2004) is 0.5–1.5 mg l^{-1}. From the analysis of results, it was found that 2% (2/112) of Aurangabad, 6% (9/156) of Gaya, 2% (1/47) of Jahenabad, and 9% (7/75) of Nawada districts have exceeded the upper bound of the acceptable level of fluoride (> 1.5 mg l^{-1}). It was also observed that the majority of the sample had fluoride concentration below the lower bound of the recommended level (< 0.5 mg l^{-1}). About 59%

Table 1 Parameter values used for Sobol sensitivity for risk-assessment model (EPA 2011; Huang et al. 2017; USEPA 1992; WHO 2004)

Parameters*	Unit	Population value ± SD		
		Children	Teen	Adults
Water Intake rate	l day^{-1}	1.25 ± 0.57	1.58 ± 0.69	1.95 ± 0.64
Average time	Days	2190	2190	9125
Exposure frequency	Day $year^{-1}$	Min: 180, max: 345, mode: 365	Min: 180, max: 345, mode: 365	Min: 180, max: 345, mode: 365
Exposure duration	Year	6	6	6
Body weight	kg	16.68 ± 1.48	46.25 ± 1.18	57.03 ± 1.10
Skin surface area	cm^2	7422 ± 1.25	14,321 ± 1.18	18,182 ± 1.10
Dermal permeability	cm h^{-1}	1×10^{-3}	1×10^{-3}	1×10^{-3}
Exposure time during shower	h day^{-1}	0.13 ± 0.0085	0.13 ± 0.0085	0.13 ± 0.0085
Fraction of skin in contact with water	Unit less	Min: 0.4 Max: 0.9	Min: 0.4 Max: 0.9	Min: 0.4 Max: 0.9
Fraction of fluoride absorbed in gastrointestinal tract	Unit less	1	1	1
Oral reference dose	mg kg^{-1} day^{-1}	0.06	0.06	0.06

*Input parameters for integrated Monte Carlo simulation

Table 2 Descriptive statistics of fluoride in Middle Gangetic Plain of India

Districts		Min	Max	1st Quartile	Median	3rd Quartile	Mean	Standard deviation	Skewness	Kurtosis	Geometric mean	Harmonic mean
Aurangabad	*F*	0.1	2.1	0.3	0.5	0.6	0.5	0.3	2.2	6.1	0.5	0.4
	pH	6.4	7.5	7.0	7.1	7.2	7.1	0.21	−0.72	1.05	7.08	7.07
	EC	207	3001	530	618	847	744	418	2.5	8.6	643.4	230.7
Gaya	*F*	0.1	2.30	0.31	0.50	0.70	0.59	0.40	1.71	3.06	0.48	0.39
	pH	5.8	7.6	6.9	7.1	7.2	7.0	0.26	−1.56	5.37	7.02	7.01
	EC	188	3891	556	714	997	866	535	2.40	8.5	738.3	488.4
Jahenabad	*F*	0.2	1.60	0.34	0.51	0.68	0.59	0.32	1.25	1.22	0.51	0.45
	pH	6.8	7.7	7.2	7.3	7.4	7.3	0.20	−0.50	0.28	7.26	7.26
	EC	306	1646	575	655	786	742	305	1.52	1.6	692.4	652.0
Nalanda	*F*	0.2	1.50	0.32	0.45	0.62	0.52	0.28	1.51	2.25	0.46	0.41
	pH	6.7	7.7	7.2	7.3	7.4	7.3	0.20	−0.52	0.60	7.29	7.29
	EC	188	1856	281	334	458	412	232	3.54	17.6	373.7	348.3
Nawada	*F*	0.1	3.60	0.48	0.60	0.92	0.84	0.72	2.46	6.11	0.65	0.48
	pH	6.4	8.0	6.99	7.13	7.27	7.12	0.27	0.00	2.02	7.12	7.11
	EC	136	1974	455	597	747	665	363	1.55	2.7	579.2	493.3

samples of Aurangabad, 50% samples of Gaya, 49% samples of Jahenabad, 62% samples of Nalanda, 27% samples of Nawada had concentrations below the lower bound level of 0.5 mg l^{-1}. The pH value in the study region varied from 5.8 to 8.0. The highest standard deviation of pH was found for Nawada districts followed by Gaya, Aurangabad, Jahenabad and Nalanda. The value of EC ranged in the regions from 136 to 3891 µS cm^{-1}. The highest standard deviation was recorded for the Gaya district. The large variations of the in situ parameters may be due to large geographical variability.

The spatial distributions of fluoride in drinking water in five districts in Bihar are shown in Fig. 3. The eastern and south-eastern parts of the study area mostly have the higher concentrations with a few spots of high concentrations in the middle north and south-west. The Nawada district located in the south-east region has the highest concentration of fluoride in terms of spatial extent. Groundwater in the southern and northern districts (Aurangabad, Gaya;

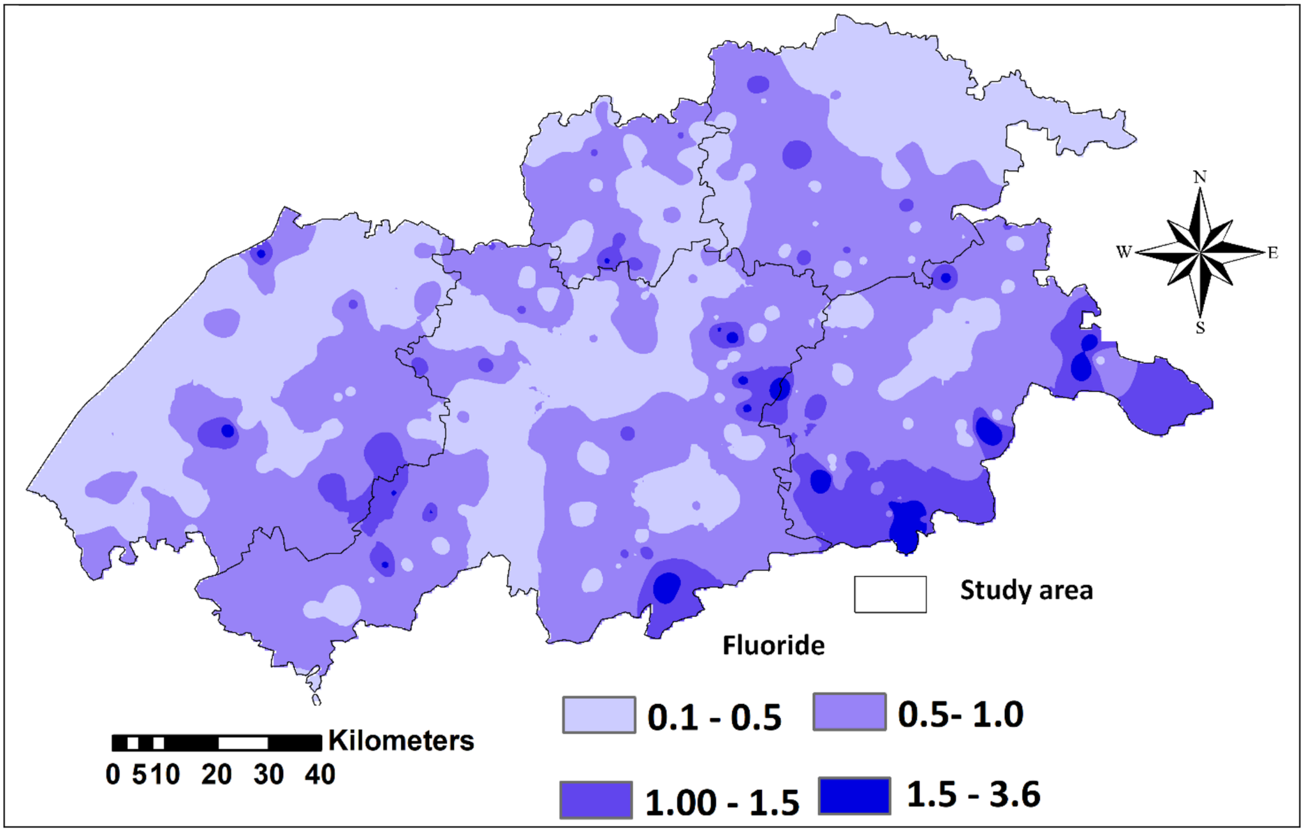

Fig. 3 Spatial distribution of fluoride in groundwater in the studied areas

and Jahenabad and Nalanda) has fluoride concentrations lower than 0.5 mg/l, which is less than the WHO guidelines (WHO 2004), and may lead to increased tooth decay as harmful health effect.

Risk Assessment of Fluoride

In this section, the non-carcinogenic health risk of fluoride in drinking water to the population Mid-Gangetic plain has been estimated. The ADD was estimated for different age groups that get exposure to fluoride through drinking water consumption. Table 3 shows the results of non-carcinogenic potential doses for the three age groups of study area.

Table 3 $ADD_{ingestion.}$ for different age groups in the studied area

Age group	$ADD_{ingestion.}$	Aurangabad	Gaya	Jahenabad	Nalanda	Nawada
Child	Min	4.46E−03	4.60E−03	1.42E−02	1.27E−02	3.19E−03
	Mean	3.67E−02	4.15E−02	4.15E−02	3.67E−02	5.97E−02
	95th	8.11E−02	1.13E−01	7.79E−02	8.50E−02	1.74E−01
	Max	1.49E−01	1.63E−01	1.13E−01	1.06E−01	2.55E−01
Teen	Min	2.03E−03	2.09E−03	6.44E−03	5.80E−03	1.45E−03
	Mean	1.67E−02	1.89E−02	1.89E−02	1.67E−02	2.71E−02
	95th	3.69E−02	5.15E−02	3.54E−02	3.86E−02	7.89E−02
	Max	6.76E−02	7.41E−02	5.15E−02	4.83E−02	1.16E−01
Adults	Min	4.89E−04	5.04E−04	1.55E−03	1.40E−03	3.49E−04
	Mean	4.02E−03	4.54E−03	4.54E−03	4.02E−03	6.53E−03
	95th	8.88E−03	1.24E−02	8.53E−03	9.31E−03	1.90E−02
	Max	1.63E−02	1.78E−02	1.24E−02	1.16E−02	2.79E−02

The average daily dose of non-carcinogenic risk of fluoride was found to be higher for the children compared to teens and adults. The 95th percentile of potential dose was found higher for children in Nawada (0.174 mg kg day^{-1}) followed by Gaya (0.113 mg kg day^{-1}), Nalanda (0.085 mg kg day^{-1}), Aurangabad (0.081 mg kg day^{-1}) and Jahenabad (0.078 mg kg day^{-1}). In addition, it was also found that the ADD scores are higher than dermal absorption. Therefore, it can be concluded that the ingestion is the primary route of exposure to fluoride through drinking water (WHO 2004). Table 4 shows the descriptive statistics of HI values estimated for ingestion and dermal contact pathways for three age groups (Children, Teens and Adults). The mean HI value for children in all the study regions were less than 1, whereas the 95th percentile exceeded the value of 1 for both children and teens. The results lead us to conclude that children of the study are at the highest risk followed by teens and adults. The cause behind the high risk is the low BW, which results in higher exposure dose for the lower age groups (Huang et al. 2017). Akiniwa (1997) has reported 0.3 mg F kg^{-1} BW dose as threshold value for inducing acute intoxication to human body. None of the samples of the studied regions exceeded this threshold value. In Nawada district, the HI value of children (mean = 1.49, 95th percentile = 4.34) followed by teens (mean = 1.06, mean = 2.32) showed that these groups were at high non-carcinogenic risk due to exposure of fluoride-contaminated groundwater. Based on the analysis of results, it can be concluded that the lower age groups in the study region are specifically prone to the non-carcinogenic risk. Furthermore, it can also be concluded that a large section of the study area is also affected by the low concentration of fluoride (Fig. 3). The low concentration of fluoride may be of geogenic nature. However, this was not investigated, as the work was beyond the scope of study. The groundwater pH values in all districts are of acidic to neutral in nature, which might resist the dissolution/mobilization of fluoride in groundwater. In addition, the absence of geological minerals especially fluoride-bearing minerals like fluorite, appatite, etc., might also contribute to low fluoride concentrations. Since large variations in hydrogeology and hydrogeological conditions prevail in this region, it is difficult to come to any concrete conclusion, and therefore there arises the need for root-cause analysis. Therefore, water-quality monitoring and assessment should be carried out for preventing non-carcinogenic health effect. The alternate way to minimize this high exposure of fluoride concentration is through adoption of deep well as sources for drinking water. Huang et al. (2017) have also emphasized the preference for adoption of deep well as sources where the shallow groundwater is contaminated by high fluoride concentration.

Model Sensitivity: Sobol Scores

The uncertainty within any model output is strongly affected by availability and quality of input data. For any risk-assessment model, uncertainty occurring during the whole process of sampling and analysis can be minimized but not removed completely. Higher uncertainty is obtained, when single-point values are used to estimate the risk of a given population. To overcome such effects and calculate the most influential inputs of model, the variance decomposition SSA was performed considering sample size of 10,000 simulations. The Sensitivity analysis of model output is strongly influenced by the specific range of input parameters. The authors have selected the values listed in Table 1, appropriate for the present analysis, which were published by Huang et al. (2017). SSA calculates the effects of each input through

Table 4 Hazard Index (HI) scores for different age groups in the studied area

Age groups	HI	Locations				
		Aurangabad	Gaya	Jahenabad	Nalanda	Nawada
Child	Min	1.12E−01	1.15E−01	3.55E−01	3.19E−01	7.98E−02
	Mean	9.27E−01	1.04E+00	1.04E+00	9.20E−01	1.49E+00
	95th	2.03E+00	2.84E+00	1.95E+00	1.98E+00	4.34E+00
	Max	3.72E+00	4.08E+00	2.84E+00	2.66E+00	6.38E+00
Teen	Min	5.98E−02	6.17E−02	1.90E−01	1.71E−01	4.27E−02
	Mean	4.96E−01	5.56E−01	5.56E−01	4.92E−01	7.99E−01
	95th	1.09E+00	1.52E+00	1.04E+00	1.06E+00	2.32E+00
	Max	1.99E+00	2.18E+00	1.52E+00	1.42E+00	3.42E+00
Adults	Min	1.46E−02	1.50E−02	4.62E−02	4.16E−02	1.04E−02
	Mean	1.21E−01	1.35E−01	1.35E−01	1.20E−01	1.95E−01
	95th	2.65E−01	3.70E−01	2.54E−01	2.58E−01	5.66E−01
	Max	4.85E−01	5.32E−01	3.70E−01	3.47E−01	8.32E−01

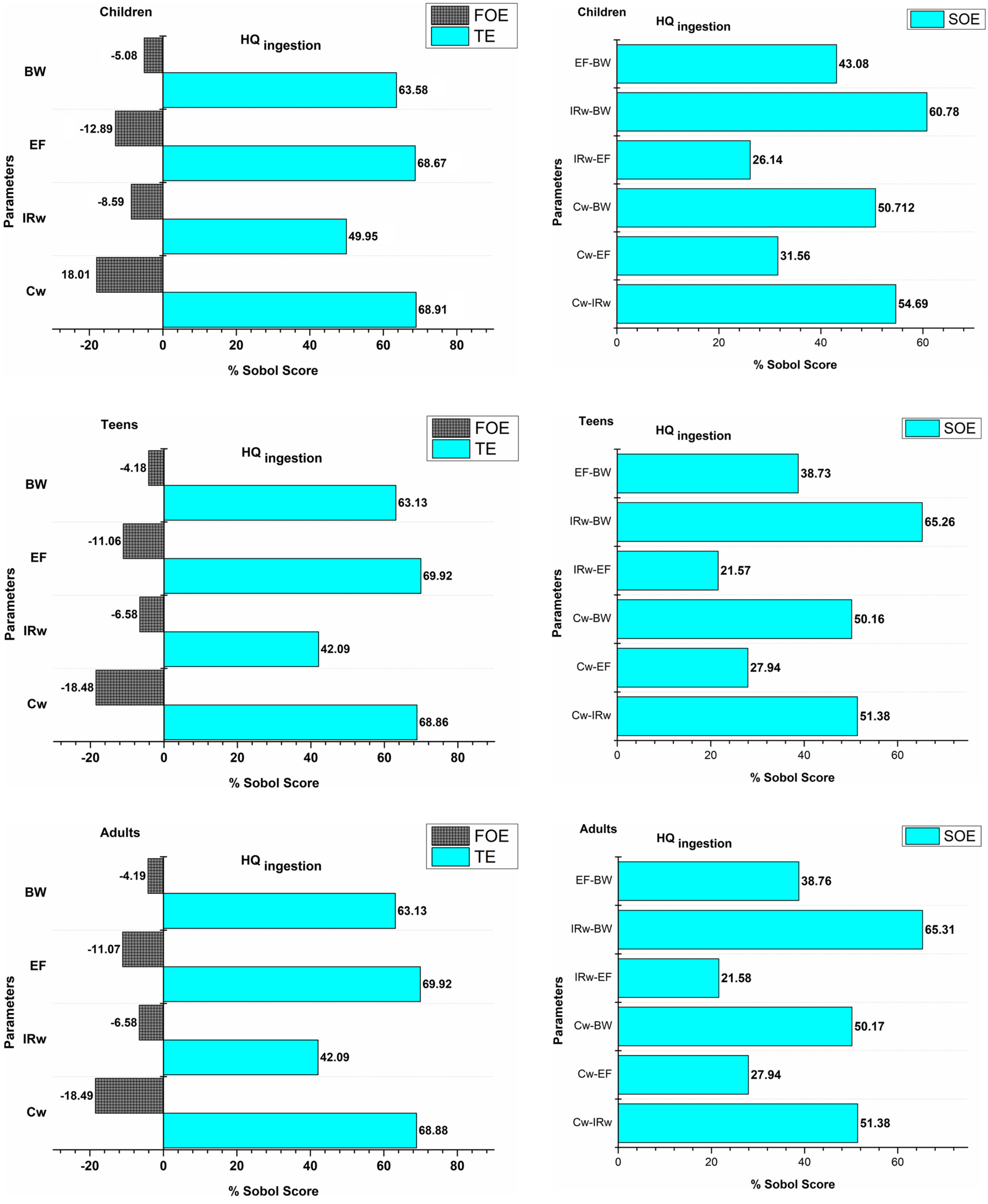

Fig. 4 Sensitivity analysis based on oral HQ model for different age groups considering first-order effect (FOE), second-order effect (SOE) and total effect (TE)

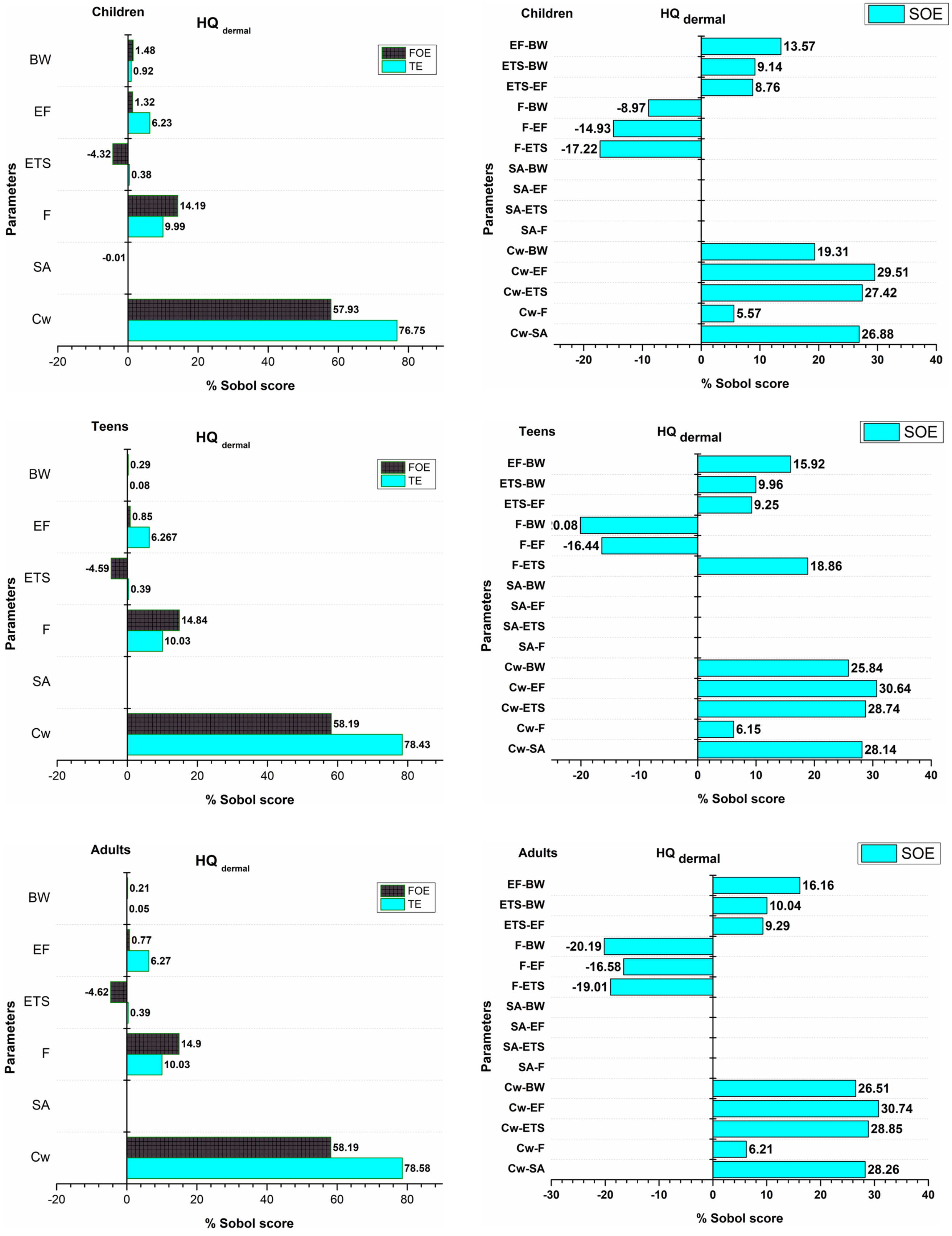

Fig. 5 Sensitivity analysis based on dermal HQ model for different age groups considering first-order effect (FOE), second-order effect (SOE) and Total effect (TE)

minimizing the uncertainty in calculations through variance decomposition. SSA was performed on HQ model and the Sobol indices were calculated for four input parameters (C_w, IR_w, EF, and BW) that were considered as potential dose for inducing the ingestion effect. Further, for dermal health risk, HQ_{dermal} model was used considering six inputs (C_w, F, ET_S, EF, BW, and SA) parameters. To investigate sensitivities of both the models, Sobol scores were calculated for each inputs (4 for oral model) and (6 for dermal model). Three types of Sobol scores; first-order effect (FOE), second-order (SOE) and TE were calculated.

Figure 4 shows the Sobol scores for all four input variables of the model considering three age groups (children, teens and adults). Interesting point to be noted here is that all the first-order scores for four inputs have values less than zero. Since these values are negative, we cannot interpret them in terms of relative contributions to the variance. This may be overcome by increasing the sample size, and that could be the scope of further research in this area. As stated above, first-order score does not give any clear idea about each input contribution to the output of the model. Therefore, SOE (i.e. interaction effect) and TE were further estimated for oral model. For the FOE, the highest value was recorded for C_w followed by EF, IR_w and BW. That clearly shows the relative importance of each features for estimation of oral risk value. But for the TE the inputs C_w (68.91%) and EF (68.68%) approximately shows the equal effect, whereas as BW has high Sobol score than the IR_w. To know the interaction effects of each variable for risk value, SOE was calculated. The high value of interaction score was found for IR_w–BW followed by C_w–IR_w, C_w–IR_w, EF–BW, C_w–EF, IR_w–EF. This result reveals that IR_w–BW scores are the important input parameters for the assessment of oral health risk than the C_w alone. Similar relative importance trends were also observed for others age groups as well. The interaction effects score was recorded high for Adults for IR_w–BW followed by teens and children (Fig. 4).

Figure 5 shows the Sobol scores of six variables for dermal model considering age groups (children, teens and adults). For all age groups, the highest value Sobol scores was found for the C_w followed by those of fraction of skin contact (*F*) and EF and BW. The ET values for FOE were found negative which indicated the presence of possible higher-order or interaction effects. In addition, skin surface area (SA) has no contribution for the determination of dermal risk value. For the FOE, the highest value was recorded for C_w followed by fraction of skin contact, EF and BW. This clearly shows the relative importance of each features for estimation of dermal risk value. For the TE the inputs C_w contributes 78.58% for adults; 78.43% teens 76.75% for children and C_w approximately shows the equal effect both teens and adults. The SOE shows interaction effects between the contributed variables. The higher value of interaction was found for the C_w -EF followed by C_w–ETS, C_w–SA. In addition, significant interaction effect (i.e. SOE) was found between C_w and SA, which was absent during FOE and TE. The higher value of C_w–SA was recorded for adults followed by teens and children. That shows the older age groups have more dermal risk than the younger age groups.

Conclusions

In this research, a total of 456 groundwater samples were investigated to assess the fluoride concentrations of the study regions. Of the 456 groundwater samples taken from the five districts, 96% of the samples are within the maximum acceptable limit of WHO guidelines. The results of spatial distribution show that Nawada district has the high concentration of fluoride distribution. In addition, the eastern and south-eastern parts of the studied areas have the higher concentrations of fluoride with a few spots in the middle north and south-west. Groundwater in the southern and northern districts (Aurangabad, Gaya and Jahenabad and Nalanda) has a fluoride concentration of lower than 0.5 mg l^{-1} which is less than the WHO guidelines (WHO 2004) and leads to increased tooth decay. For the FOE, the highest value was recorded for C_w followed by EF, IR_w and BW. This clearly showed the relative importance of each feature for estimation of oral risk value. However, for the TE, the inputs C_w (68.91%) and EF (68.68%) are approximately same. The Sobol score of BW was found to be higher than that of the IR_w. For the interaction effect (SOE), the highest value was found for IR_w–BW followed by C_w–IR_w, C_w–IR_w, EF-BW, C_w–EF, IR_w–EF. This result revealed that IR_w–BW effects are the important input parameters for the assessment of oral health risk. The highest Sobol scores were recorded for Adults followed by teens and children. The SOE values for dermal model showed that the fluoride concentration and skin surface area have significant contribution towards dermal risk value. In addition, the highest value of C_w–SA was recorded for adults followed by teens and children. This result revealed that the older age groups have more dermal risk than the younger age groups. The future scope of this research can be devoted to find the effects of exposure to fluoride through other ways of contact (e.g. food) with seasonal variation.

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Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

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