

Geo-spatial distribution of fluoride in drinking water resources in Eastern Iran

Nourahmad Nourafrouz, Ali Naghizadeh, Borhan Mansouri, Hamed Biglari, Elham Derakhshani and Vaibhav Thakkar

ABSTRACT

The purpose of this study was to determine the fluoride concentrations in drinking water of the Khaf County in Eastern Iran. Moreover, health risk assessment of three age groups (children, teens and adults), sensitivity analysis and uncertainties in the risk estimates were carried out using Monte Carlo simulation. For this reason, drinking water in 33 villages and 5 cities of the Khaf County were collected during March to September 2018. Fluoride contents in drinking water samples were assayed by using a HACH-DR6000 spectrophotometer. Fluoride content in drinking water from urban area and rural area ranged from 0.50 to 0.91 mg L⁻¹ and 0.24 to 2.31 mg L⁻¹. Among the population of the 33 villages, about 17 villages, corresponding to 51%, receive fluoride concentrations less than 0.5 mg L⁻¹ (minimum allowable concentration recommended by WHO), while the population of 4 villages, corresponding to 12%, receive fluoride concentrations higher than 1.5 mg L⁻¹ (the maximum allowable concentration of fluoride in drinking water recommended by the WHO). Moreover, our findings showed that the drinking water ingestion rate, fluoride concentration in water, and the fraction of skin in contact with water were the most important variable in calculating the Hazard quotient (HQ).

Key words | drinking water, fluoride, fluorosis, health risk

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INTRODUCTION

Nowadays, a readily available access to safe drinking water resources is an important aspect of public health in many countries around the world including Iran (Garg & Malik 2004; Fallahzadeh *et al.* 2018). Iran is a vast country, and has diverse climatic conditions in comparison to other countries. Khorasan Razavi is located in the northeast of Iran. In the southern part of this province, because of its close proximity to the desert, a dry climate is dominant (Keshavarzi *et al.* 2006; Madani 2014; Soltani *et al.* 2016). Providing safe and quality drinking water will have a significant impact on sustaining life and social health.

Generally, potable water contains both essential and non-essential elements. A lot of elements such as calcium, manganese, and fluoride that exist in drinking water are essential for metabolic activities as well as human health (Nerbrand *et al.* 2003; Li *et al.* 2009; Kazi *et al.* 2018a, 2018b). However, increasing the concentration of trace elements in relation to the required level in the body can lead to health problems. Contamination of drinking water with fluoride and its potential health implications remain a major public health issue in many countries, especially in Iran (Yousefi *et al.* 2018). Therefore continuous

monitoring and control of drinking water quality is very important (WHO 2004).

Fluoride (F) is one of the mineral elements that exist in the earth's crust. This F ion is highly reactive due to the presence of a free electron in its upper electron layer, and is mostly present in compounds such as apatite and cryolite in nature (Kır *et al.* 2016). Contamination of surface and groundwater by fluoride can be traced to natural resources (availability and solubility of fluoride minerals and fluoride-bearing rocks) and anthropogenic resources (hydrofluoric production, fertilization, electroplating of metals, the electronics manufacturing industry) (Tor 2006; Xu *et al.* 2011). The World Health Organization (WHO) has recommended the minimum and maximum allowable concentration of fluoride in drinking water as 0.5 and 1.5 mg L⁻¹ respectively. Defluoridation processes are recommended for water supplies where their fluoride concentration is higher than the standards (Kazi *et al.* 2018a, 2018b).

Also, according to the United States Public Health Service (USPHS) the maximum allowable concentration of fluoride in water depends on climate conditions because air temperature affects the amount of water consumed and consequently the exposure to the amount of fluoride (USPHS 1963; WHO 2004; Miretzky & Cirelli 2011). The optimal water fluoride concentration depends on several parameters such as methods of food processing and cooking, amount of food and water intake, dietary habits of the community, local climatic conditions, and physio-chemical parameters of water (Grimaldo *et al.* 1995; Kaseva 2006; Maheshwari 2006). Although drinking water is the main source of human exposure to fluoride, food, dental products, and pesticides are other important sources for dealing with fluoride (Garg & Malik 2004; National Research Council 2007).

Fluoride fluctuations can be associated with health problem of consumers, so that low fluoride concentrations cause tooth decay, and high concentrations of this element in water increase the risk of dental and bone fluorosis. According to previous studies, nearly 80% of prevalence and incidence of diseases all over the world are related to low water quality. Also contamination of drinking water with fluoride accounts for 65% of endemic fluorosis (World Health Organization 2011). Moreover, long term exposure to high fluoride concentrations may lead to various diseases such as dental and skeletal fluorosis, low

hemoglobin levels, skin rashes, gland damage, gastrointestinal problems, infertility, abortions, neurotoxicological effects, and Alzheimer's disease (Sehn 2008; Choi *et al.* 2012; Choi *et al.* 2015). Research reported that more than 30 million people in China suffer from fluorosis and about 100 million people are exposed to it (Chen *et al.* 2010). In another study conducted by Huang *et al.* (2017), the authors reported that the concentration of fluoride in groundwater in California is more than 1.5 mg L⁻¹ (Huang *et al.* 2017).

Moreover, the variations of fluoride concentration in drinking water are one of the main problems in drinking water quality in Iran. In some cities in Iran, the amount of fluoride in drinking water is higher than 1.5 mg L⁻¹, which can lead to fluorosis (Asgari *et al.* 2012). For example Qasemi *et al.* (2018) reported that 55% of the rural areas in Gonabad and 4.7% of the rural areas in Bajestan had fluoride levels below the minimum recommended value of WHO (Qasemi *et al.* 2018). In another study, Ghaderpoori *et al.* (2018) illustrated that the fluoride levels from all sampling stations in the water distribution network of Mashhad in Eastern Iran were below 0.5 mg L⁻¹ (Ghaderpoori *et al.* 2018). Therefore, the aim of this study was to determine the fluoride concentration of drinking water in 33 villages and five cities of the Khaf County, which is located in the southern part of Khorasan Razavi province, from March to September 2018. Moreover, we assessed the potential health risks from fluoride ingestion through drinking water for infants, children, teenagers and adults. The results of this study could be useful for future planning of water resources as well as public knowledge about health problems related to high fluoride concentrations.

MATERIALS AND METHODS

The present study was descriptive-analytical and was performed experimentally on drinking water resources of Khaf County in Khorasan Razavi Province of Iran for determination of the fluoride ion concentration. The Khaf County contains five cities including Khaf, Nashtifan, Qasemabad, Salami, and Sangan. The climate of this region is hot and dry; that is, hot summers and cool winters. The mean annual temperature and rainfall in Khaf County are 18 °C and 120 mm respectively and the annual average of rainfall is about 255.7 mm. At the

2016 census, the county's population was 140,000 in 26,542 families. In the present study, all drinking water resources (38 wells and springs: five cities and 33 villages) were sampled in spring and summer of 2018 and their fluoride concentration was determined. The sampling locations as well as the map of Khaf County are shown in Figure 1.

Prior to sampling and accessing the drinking water resources, necessary coordination with the Water and Wastewater Administration was carried out and the required permissions were obtained. One-liter polyethylene containers were used to sample water from the studied areas. The containers were rinsed with acid and distilled water, and thoroughly cleaned before sampling. The date, time, and location of sampling were recorded on all the sampling containers. Then, samples were transferred to the laboratory by storing the cold chain (in 4 °C). Finally, fluoride concentrations in water samples were determined within 24 h after collection in the laboratory using a HACH-DR6000 spectrophotometer machine with SPADNS reagent. The detection limit for fluoride concentration was 0.02–0.2 mg L⁻¹ (method 8029). Moreover, in this study, Arc GIS 10.4.1 software (Esri, Berkeley, CA, USA) was used for spatial distribution of fluoride in the studied areas.

In this study, the daily exposure to fluoride by drinking water was estimated using Equations (1) and (2) (USEPA 2004; Fakhri *et al.* 2018):

$$EDI_{ing} = \frac{C_w \times IR_w \times EF \times ED}{BW \times AT} \quad (1)$$

$$EDI_{derm} = \frac{C_w \times SA \times K_p \times F \times ET_s \times EF \times ED \times 10^{-3}}{BW \times AT} \quad (2)$$

In this regard, EDI_{ing} estimates daily intake of fluoride consumed per day by drinking water and EDI_{derm} estimates the amount of fluoride received by skin absorption based on mg kg⁻¹ day⁻¹. C_w is the concentration of fluoride in drinking water in mg L⁻¹, IR_w is the drinking water ingestion rate based on L/day, EF is the exposure frequency based on day/year, ED is the exposure duration in terms of years, BW is the body weight in kg, AT is the averaging time in days, SA is the surface area of skin in terms of cm², K_p is the coefficient of skin permeation (Cm/h), fluoride is the fraction of the contact surface of the skin with water (without unit) and ET_s is the exposure time when

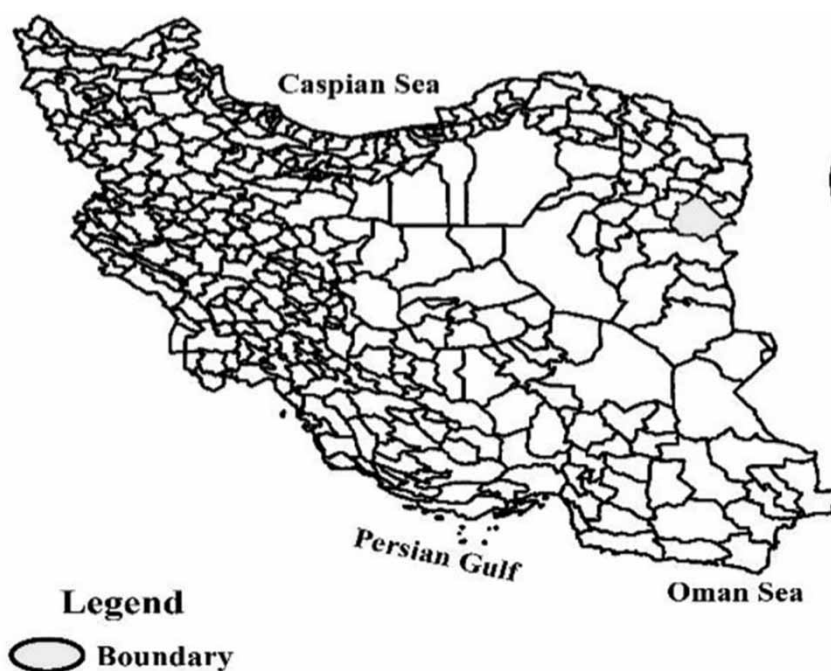


Figure 1 | Map of the study area and sampling stations in Khaf County, Eastern Iran.

showering (h/day). The Hazard quotient (HQ) of the non-carcinogenic risk estimate for fluoride exposure through drinking water and dermal exposure is calculated using Equation (3).

$$HQ = \frac{EDI}{RfD} \quad (3)$$

RfD in this equation expresses the reference fluoride dose by a specific exposure pathway in $\text{mg kg}^{-1} \text{ day}^{-1}$. Based on the USEPA database, the amount of *RfD*, oral reference dose, through drinking water consumption is $0.06 \text{ mg kg}^{-1} \text{ day}^{-1}$ (USEPA 2004). $HQ < 1$ implies a negligible risk of non-carcinogenic impacts, whereas $HQ > 1$ indicates potential non-cancer-causing health effects. Based on the USEPA, *RfD* represents the reference dose of fluoride in a specific exposure pathway ($\text{mg kg}^{-1} \text{ day}^{-1}$).

Monte Carlo simulation and sensitivity analysis

We used sensitivity analysis in Monte Carlo simulation to identify important and effective parameters in the output model (Table 1). Crystal Ball software was used to simulate Monte Carlo. We also used 1,000 trails to analyze the

sensitivity analysis. In this study, Crystal Ball (version 11.1.1.1, Oracle, Inc., USA) was used to simulate Monte Carlo and perform sensitivity analysis with 1,000 trails.

RESULTS

Fluoride concentration in drinking water

Based on Table 2, overall fluoride levels from all sampling sites in rural and urban residents living were 0.69 and 0.70 mg L^{-1} respectively. The minimum and maximum contents of fluoride ions in urban regions were 0.50 and 0.99 mg L^{-1} respectively, while the minimum and maximum content of fluoride ions in rural regions were 0.18 and 2.31 mg L^{-1} respectively. The variations of fluoride levels in drinking water of rural areas were more than urban areas. Based on results from Table 2, the population of 17 of 33 villages (51%) receive fluoride concentrations less than the limit recommended by WHO of 0.5 mg L^{-1} , while the population of 4 of 33 villages (12%) receive fluoride concentrations higher than the maximum allowable concentration of fluoride in drinking water recommended by the WHO of 1.5 mg L^{-1} . Moreover, distribution of fluoride levels in two seasons is presented in Figures 2 and 3, so

Table 1 | Parameters used for the probabilistic risk model

Parameters (units)	Distribution type	Values		
		Children	Teens	Adults
Skin surface area (cm^2)	Lognormal	7422 ± 1.25	14321 ± 1.18	18182 ± 1.10
Body weight (kg)	Lognormal	16.68 ± 1.48	46.25 ± 1.18	57.03 ± 1.10
Ingestion rate (L/day)	Normal	1.25 ± 0.57	1.58 ± 0.69	1.95 ± 0.64
Average time (days)	Fixed value	2190	2190	9125
Exposure frequency (day/year)	Triangular	Min: 180 Mode: 345 Max: 365	Min: 180 Mode: 345 Max: 365	Min: 180 Mode: 345 Max: 365
Exposure duration (year)	Fixed value	6	6	6
Dermal permeability constant (cm/h)	Fixed value	1×10^{-3}	1×10^{-3}	1×10^{-3}
Exposure time in the shower (h/day)	Lognormal	0.13 ± 0.0085	0.13 ± 0.0085	0.13 ± 0.0085
Fraction of skin in contact with water	Uniform	Min: 0.4	Min: 0.4	Min: 0.4 Max: 0.9
Fraction of fluoride absorbed in gastrointestinal tract	Fixed value	1	1	1
Oral reference dose ($\text{mg kg}^{-1} \text{ day}^{-1}$)	Fixed value	0.06	0.06	0.06

Table 2 | Fluoride concentrations (mg L^{-1} ; mean and SD) in drinking water and frequency of hypertension in Khaf County, Eastern Iran

Stations	Latitude	Longitude	Mean	SD	Hypertension
Urban areas					
Khaf	34.58687672	60.16021906	0.50	0.233	463
Nashtifan	34.47256712	60.20494397	0.54	0.339	451
Salami	34.74197028	59.98430501	0.54	0.091	475
Sangan	34.40604183	60.25967174	0.99	0.466	602
Qasemabad	34.35702395	59.85749109	0.91	0.014	400
Overall			0.69	0.229	
Rural areas					
Sayed Abad	34.84537916	59.78127279	0.36	0.304	74
Sijavand	34.86352036	59.82868311	0.54	0.155	92
Chaman Abad	34.8323684	59.80180461	0.41	0.240	114
Mazrehshaikh	34.87756192	59.91496106	0.29	0.162	82
Sadeh	34.83145386	59.8949104	0.32	0.162	385
Khalil Abad	34.7886955	59.85509389	0.24	0.169	45
Hazarkhosheh	34.74454964	59.87032228	0.48	0.021	45
Ahmad Abad	34.77200424	59.90665737	0.35	0.141	140
Khair Abad	34.84628966	60.01281208	0.51	0.113	95
Hassan Abad	34.72589944	59.89459892	0.45	0.063	89
Chahar Deh	34.69690958	60.03558714	0.31	0.070	105
Razdab	34.81018533	60.12416025	0.27	0.219	73
Fayandar	34.66037219	60.0998979	0.46	0.077	70
Mahabad	34.58204424	60.06729215	2.31	0.487	12
Barakuh	34.68505963	60.17888137	0.58	0.205	35
Khargerd	34.54038693	60.18422183	0.56	0.056	116
Tizab	34.5160017	60.17971262	0.44	0.148	108
Barab	34.42904011	60.23412234	0.69	0.120	303
Nyaz Abad	34.23008266	60.24847463	1.40	0.148	83
Goryab	34.42357785	60.33274451	0.38	0.141	6
Dardoi	34.53296125	60.44891312	0.34	0.148	3
Zuzan	34.34673462	59.87040028	1.98	0.007	212
Ebrahimi	34.28437761	59.87041319	1.56	0.268	66
Baghbakhshi	34.45336889	59.52814272	0.65	0.148	80
Chahgachi	34.24257021	59.5531057	0.70	0.148	68
Kalshor (Abe-shirin)	34.02254292	59.79554483	0.29	0.070	57
Kalshor (Abe-shor)	34.02310912	59.79846006	0.68	0.049	44
Bonyabad (Abe-shor)	34.08077271	59.87445184	0.91	0.388	68
Bonyabad (Abe-shirin)	34.08007577v	59.87634548	0.18	0.084	17
Arg (Abe-shor)	34.1553623	59.91693782	1.85	0.155	10
Aliabad	34.21794791	59.96414062	1.32	0.021	17
Bias Abad	34.30870317	60.04716355	0.44	0.219	80
Baghcheh	34.43245196	60.10808853	1.04	0.410	6
Overall			0.70	0.161	

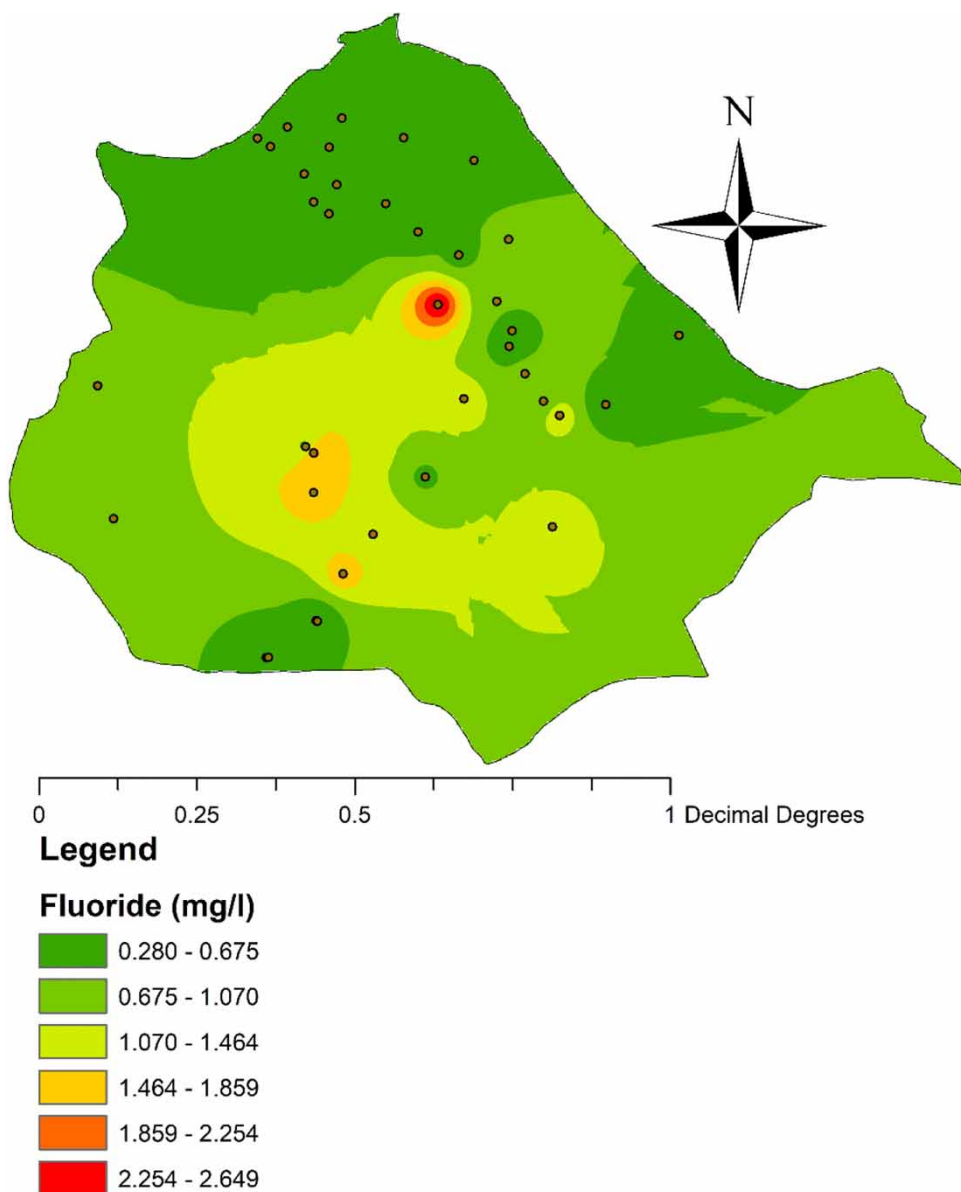


Figure 2 | Distribution of fluoride concentrations in spring in Khaf County, Eastern Iran.

that the highest concentrations of fluoride in drinking water were in the center and south locations of the studied area.

Calculation of chronic daily intake of fluoride

Mean value of EDI_{ing} for children, teens and adults in urban areas was 0.025, 0.011, and 0.002 $mg\ kg^{-1}\ day^{-1}$ respectively, and for rural areas, the mean value of EDI_{ing} for children, teens and adults was 0.026, 0.011, 0.002 $mg\ kg^{-1}$

day^{-1} respectively (Table 3). Furthermore, the mean value of the EDI_{derm} for fluoride in urban samples for children, teens and adults was 7.964×10^{-6} (6.218×10^{-6} to 1.129×10^{-5}), 0.0055 (0.0040 to 0.0078), and 0.0013 (0.0009 to 0.0019) $mg\ kg^{-1}\ day^{-1}$ respectively (Table 3), and for rural areas, the mean value of the EDI_{derm} for fluoride in rural samples for children, teens and adults was 8.087×10^{-6} (8.044×10^{-6} to 2.641×10^{-5}), 0.0056 (0.0019 to 0.0183), and 0.0013 (0.0003 to 0.0045) $mg\ kg^{-1}\ day^{-1}$ respectively.

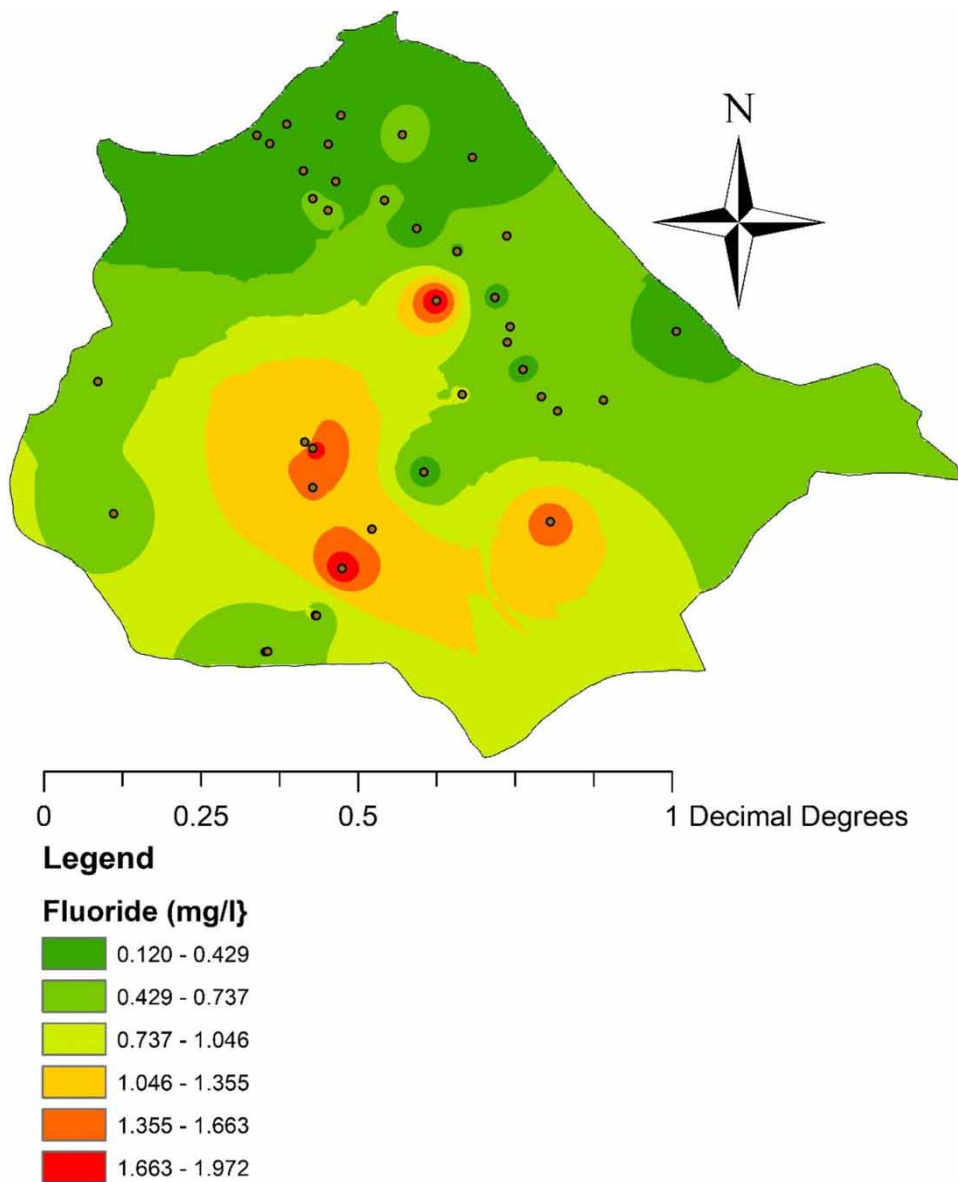


Figure 3 | Distribution of fluoride concentrations in summer in Khaf County, Eastern Iran.

Sensitivity analyses

Findings showed that the HQ value for consuming water containing fluoride for drinking was higher than the HQ level for dermal contact (Figure 4). Moreover, the most important variables affecting the value of HQ_{ing} in the three age groups are drinking water ingestion rate (IRw) and fluoride concentration in water (CW), while concentration of fluoride in water and the fraction of skin in

contact with water (F) are the most important variables in the value of HQ_{derm} in dermal contact. In the three age groups, IRw was the most important variable affecting $HQ_{overall}$. Figure 5 shows the results of the sensitivity analysis to assess the non-carcinogenic risk for the three age groups exposed to fluoride. In urban areas, IRw was the most important variable affecting the amount of health risk in the three age groups of children, teens, and adults. After IRw, fluoride concentration in drinking water (C) and exposure frequency

Table 3 | EDI ($\text{mg kg}^{-1} \text{ day}^{-1}$) for different age groups in the studied areas

Stations	EDI _{ing}			EDI _{derm}		
	Children	Teens	Adults	Children	Teens	Adults
Urban areas						
Khaf	0.0186	0.0085	0.0020	5.8E-06	0.0040	0.0009
Nashtifan	0.0199	0.0090	0.0021	6.2E-06	0.0042	0.0010
Salami	0.0201	0.0091	0.0022	6.2E-06	0.0043	0.0010
Sangan	0.0365	0.0166	0.0040	1.1E-05	0.0078	0.0019
Qasemabad	0.0336	0.0153	0.0036	1.0E-05	0.0072	0.0017
Overall	0.0257	0.0117	0.0028	8.0E-06	0.0055	0.0013
Rural areas						
Sayed Abad	0.0134	0.0061	0.0014	4.2E-06	0.0028	0.0007
Sijavand	0.0199	0.0090	0.0021	6.2E-06	0.0042	0.0010
Chaman Abad	0.0151	0.0069	0.0016	4.7E-06	0.0032	0.0008
Mazrehshaikh	0.0109	0.0049	0.0011	3.4E-06	0.0023	0.0005
Sadeh	0.0120	0.0054	0.0013	3.7E-06	0.0025	0.0006
Khalil Abad	0.0088	0.0040	0.0009	2.7E-06	0.0019	0.0004
Hazarkhosheh	0.0179	0.0081	0.0019	5.5E-06	0.0038	0.0009
Ahmad Abad	0.0129	0.0058	0.0014	4.0E-06	0.0027	0.0006
Khair Abad	0.0188	0.0085	0.0020	5.8E-06	0.0040	0.0010
Hassan Abad	0.0168	0.0076	0.0018	5.2E-06	0.0036	0.0008
Chahar Deh	0.0114	0.0052	0.0012	3.5E-06	0.0024	0.0006
Razdab	0.0101	0.0046	0.0011	3.1E-06	0.0021	0.0005
Fayandar	0.0171	0.0078	0.0018	5.3E-06	0.0036	0.0009
Mahabad	0.0855	0.0390	0.0093	2.6E-05	0.0183	0.0045
Barakuh	0.0216	0.0098	0.0023	6.7E-06	0.0046	0.0012
Khargerdeh	0.0206	0.0094	0.0022	6.4E-06	0.0044	0.0010
Tizab	0.0164	0.0074	0.0018	5.1E-06	0.0035	0.0008
Barab	0.0256	0.0117	0.0028	7.9E-06	0.0055	0.0013
Nyaz Abad	0.0519	0.0236	0.0056	1.6E-05	0.0111	0.0027
Goryab	0.0140	0.0064	0.0015	4.3E-06	0.0030	0.0007
Dardoi	0.0127	0.0058	0.0013	3.9E-06	0.0027	0.0006
Zuzan	0.0733	0.0334	0.0080	2.3E-05	0.0157	0.0038
Ebrahimi	0.0576	0.0262	0.0063	1.8E-05	0.0123	0.0030
Baghbakhshi	0.0242	0.0110	0.0026	7.5E-06	0.0052	0.0012
Chahgachi	0.0260	0.0118	0.0028	8.0E-06	0.0055	0.0013
Kalshor (Abe-shirin)	0.0107	0.0048	0.0011	3.3E-06	0.0023	0.0005
Kalshor (Abe-shor)	0.0253	0.0115	0.0027	7.8E-06	0.0054	0.0013
Bonyabad (Abe-shor)	0.0338	0.0154	0.0037	1.0E-05	0.0072	0.0017
Bonyabad (Abe-shirin)	0.0066	0.0030	0.0007	2.1E-06	0.0014	0.0003
Arg (Abe-shor)	0.0683	0.0311	0.0074	2.1E-05	0.0146	0.0036
Aliabad	0.0489	0.0223	0.0053	1.5E-05	0.0105	0.0025
Bias Abad	0.0164	0.0074	0.00180	5.1E-06	0.0035	0.0008
Baghcheh	0.0384	0.0175	0.0042	1.2E-05	0.0082	0.0020
Overall	0.0261	0.0119	0.0028	8.2E-06	0.0056	0.0013

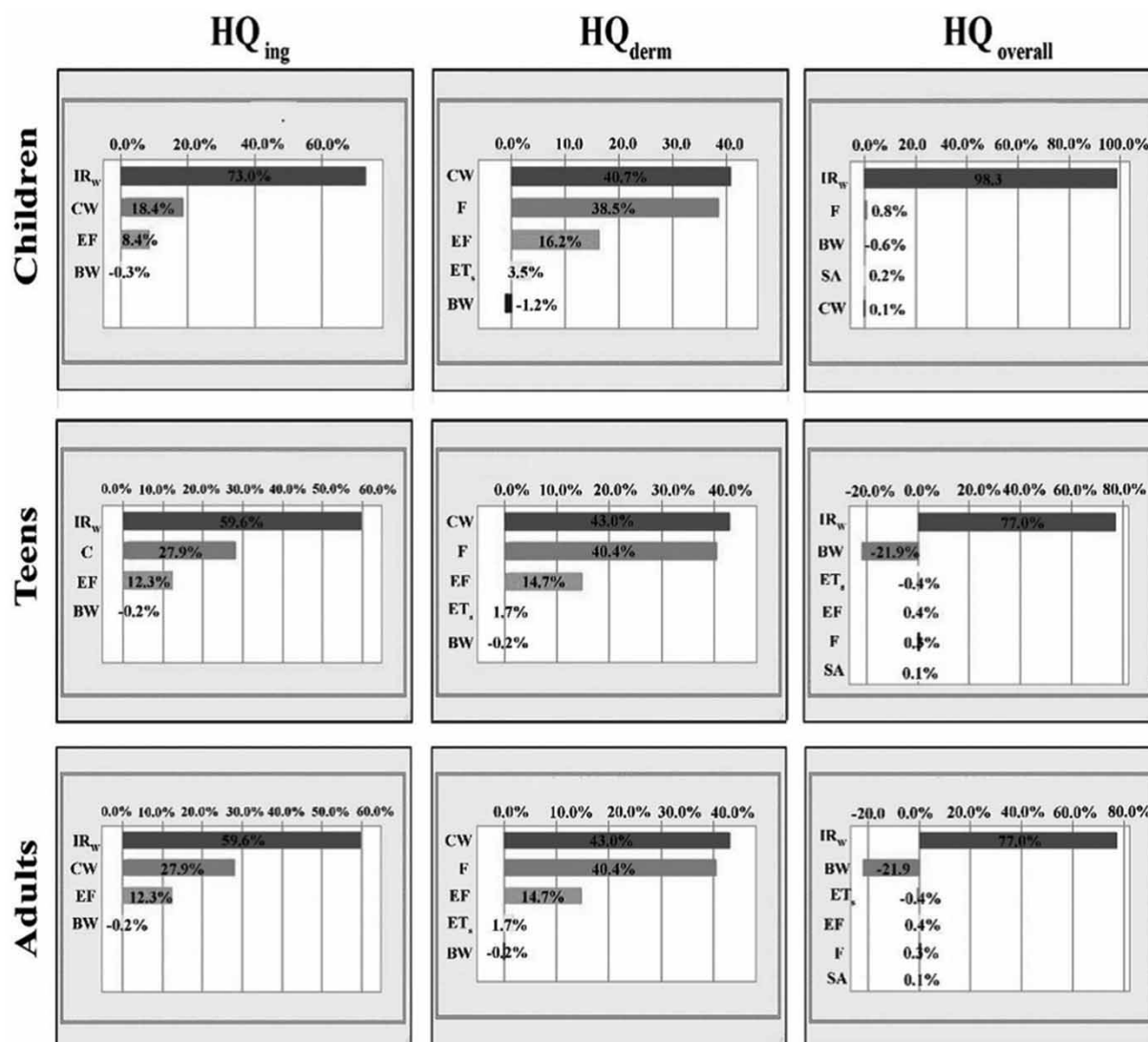


Figure 4 | Sensitivity analysis based on the type of dermal contact and drinking water ingestion for different age groups.

(EF) were the most effective variable on the value of health risk assessment in cities and rural areas.

DISCUSSION

The fluoride concentrations in drinking water of Khaf County ranged from 0.50 mg L^{-1} – 0.99 mg L^{-1} in urban areas whereas the ranges in rural region were 0.18 mg L^{-1} – 2.31 mg L^{-1} . Although the fluoride levels determined in some rural areas were outside the range of that

recommended by WHO, in temperate regions, where water intake is low, a fluoride level up to 1.5 mg L^{-1} is acceptable (World Health Organization 2011). Populations of 4 villages in the studied area were exposed to fluoride content higher than the WHO's maximum allowable concentration. On the other hand, it is illustrated that a higher concentration of fluoride in drinking water than 0.7 mg L^{-1} can pose some adverse health effects such as fluorosis, so that fluorosis prevalence was reported in different provinces of Iran (Fallahzadeh et al. 2018; Yousefi et al. 2018; Keramati et al. 2019). Moreover, some studies done in

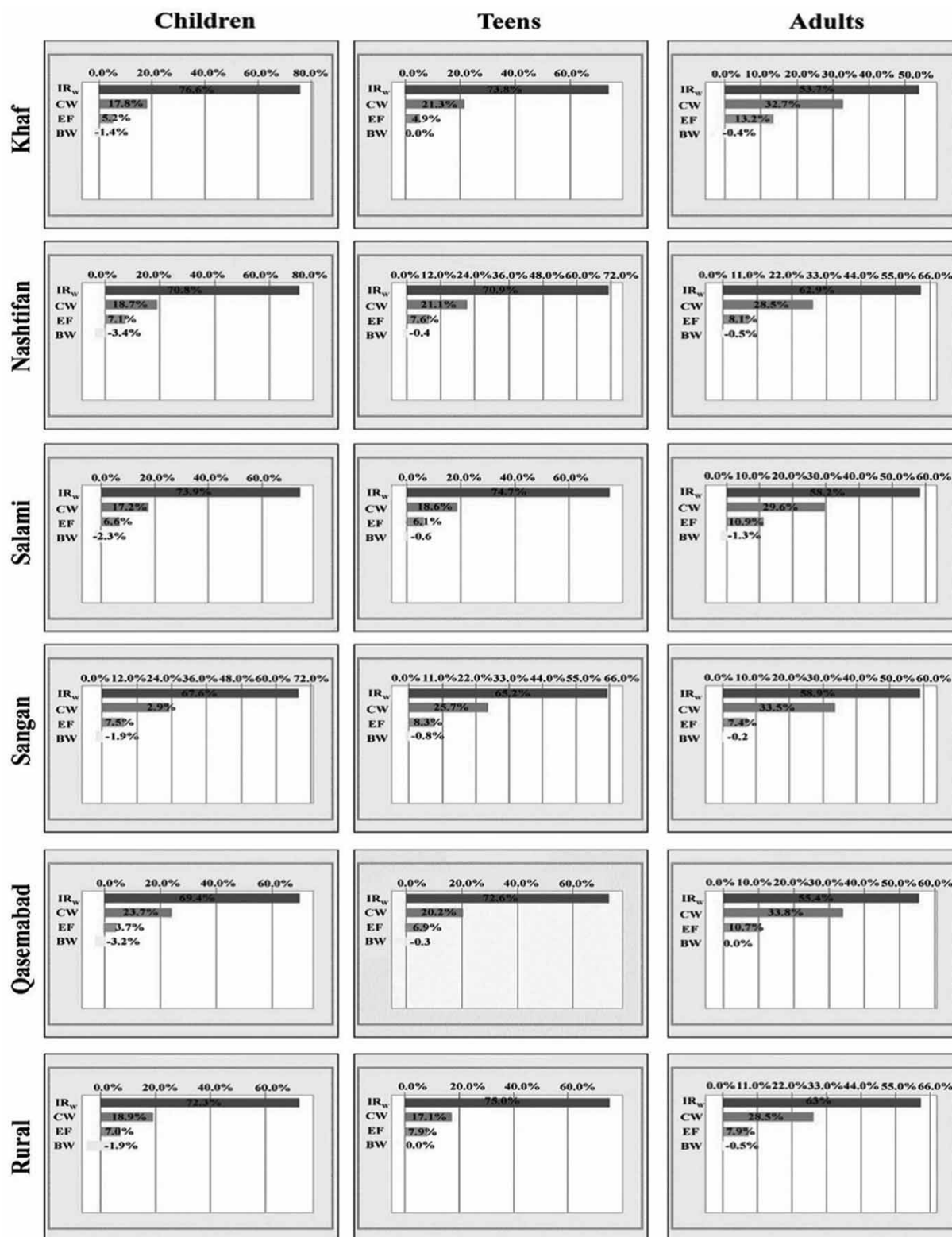


Figure 5 | Sensitivity analysis results for three age groups of children, teens and adults in studied regions.

Saudi Arabia and Mexico also reported the presence of fluoride levels in drinking water exceeding the permissible limits (Hurtado *et al.* 2000; Brahman *et al.* 2014). Almost 200 million people from 25 countries are at risk of health problems for high concentration of fluoride in their drinking water (Ayooob & Gupta 2006; Amouei *et al.* 2012; Huang *et al.* 2017). There is a pressing need for de-fluoridation activities to be carried out in high-risk regions whose drinking water contains higher fluoride concentration (Chidambaram *et al.* 2013; Habuda-Stani *et al.* 2014).

Hard tissue deformities like dental and skeletal fluorosis are seen in populations exposed to high levels of fluoride in drinking water (Fordyce 2011). Higher concentrations cause damage to the soft tissues like liver, kidney, lung, and testis and may also induce skeletal cancer and neurotoxicological effects (Bassin *et al.* 2006; Barbier *et al.* 2010; Zhang *et al.* 2016). Fluoride levels of up to 1.5 mg L^{-1} in drinking water are acceptable for maximum benefits and minimum risks, which is also recommended by WHO, other organizations in other countries, such as the European Union (DECLG), Canada (Ottawa), Australia (NHMRC), Ozsvath and India (BIS) (Ozsvath 2009; European Union 2014; Government of Canada 2010; World Health Organization 2011; NHMRC 2011; IS10500 B 2012). The US Public Health Service has set the optimum concentration of fluoride in drinking water at 0.7 mg L^{-1} (Kohn *et al.* 2001).

Fluoride levels in drinking water of 17 villages in Khaf County were less than the limit recommended by WHO. Hence, it was observed that different areas within the same county demonstrated differing levels of fluoride in drinking water. When the fluoride level in drinking water is less than 0.5 mg L^{-1} , the risk of dental caries increases (Dissanayake 1991; Ozsvath 2009). The low level of fluoride in drinking water observed in these regions was similar to those observed in various other studies (Huang *et al.* 2017; Ram *et al.* 2017). The villages with fluoride content less than 0.5 mg L^{-1} may require fluoridation programs, health education initiatives, fluoride supplements or any other combined strategies for prevention of dental caries in their population. Another recommendation is to establish a drinking water supply from deep wells with adequate fluoride content in that region with suitable conditions. Prevention of dental caries and strengthening of bones is observed when a population is exposed to low concentration of

fluoride ($0.5\text{--}1.5 \text{ mg L}^{-1}$) in drinking water (Dissanayake 1991; Ozsvath 2009).

In our study, we also observed that children exposed to high levels of fluoride in drinking water demonstrated dental fluorosis. In similar studies, Huang *et al.* (2017), Guissouma *et al.* (2017), and Fallahzadeh *et al.* (2018) reported that the children age group was the population more at potential health risk of fluoride than other age groups (Guissouma *et al.* 2017; Huang *et al.* 2017; Fallahzadeh *et al.* 2018). The chronic daily intake of fluoride (daily exposure) was calculated in this study for three different age groups and both in rural as well as urban areas. The mean value of both EDI_{ing} and EDI_{derm} was more in the children group compared to teens and adults, both in rural as well as urban areas. Signifying higher daily exposure of fluoride content in the children's age-group, this may have higher risk of adverse effects in them. Humans could be exposed to fluoride in drinking water through oral intake, dermal absorption, and inhalation. It was observed that health risk values (HQ) were higher for exposure to fluoride through consumption of water containing fluoride (HQ_{ing}) than for exposure through dermal contact (HQ_{derm}). Out of all the variables considered for this analysis, the most important variables affecting the value of HQ_{ing} in all the three age groups were drinking water ingestion rate (IRW) and fluoride concentration in water (CW), while concentration of fluoride in water and the fraction of skin in contact with water (F) were the most important variables in the value of HQ_{derm} in dermal contact. These observations were in accordance with the previous studies done in different populations in the same as well as different countries. For more accurate health risk assessment of fluoride exposure in further studies, there should be more focus on these parameters identified by sensitivity analysis (Huang *et al.* 2017; Fallahzadeh *et al.* 2018).

In our study, the overall HQ value was less than 1, indicating that there was negligible risk of non-carcinogenic effect of exposure to fluoride for the population of Khaf County. Similarly, in a study done by Zhang *et al.* (2016), the calculated national mean non-carcinogenic risk for Chinese residents were lower than 1, thus indicating the absence of potential health effects at the national level. However, the HQs of fluoride were >1 in some areas, which may pose possible health risks to local residents

(Zhang *et al.* 2016). Among the three age groups assessed in this study, children were at the highest non-carcinogenic risk possibly because children have the lowest BW, and other exposure parameters are similar to those of teens and adults. These findings are consistent with the study done by Huang *et al.* (2017). In a study conducted by Guissouma *et al.* (2017), it was observed that consumers of drinking water in areas where the HQ is higher than the guidelines suffer from dental fluorosis (Guissouma *et al.* 2017). The fluoride level in drinking water is not the recommended standard for the control of dental caries; on the basis of the socioeconomic and climatic conditions and the dietary and oral hygiene habits of their population, each country should determine the concentrations of fluoride in drinking water (Khan *et al.* 2004). Although fluoride in drinking water is commonly considered the greatest contributor to daily intake, other fluoride sources, including beverages, food-stuffs and fluoride supplements, may also significantly contribute to daily fluoride intake (Erdal & Buchanan 2004; Li *et al.* 2009).

Hence, in this study, the non-carcinogenic risk due to exposure to fluoride could be underestimated because only the drinking water and dermal absorption exposure pathway was considered. Additional data related to fluoride exposure should be collected from each exposure pathway, and by considering additional resources and time, accurate and precise estimates should be further investigated. In future studies, to obtain accurate data on health risk estimates of fluoride exposure on the population of Khaf County, fluoride exposure via inhalation should be investigated. Measures should be implemented to strengthen the timely monitoring of fluoride levels in drinking water in Khaf County.

CONCLUSION

The findings of this study indicated that the fluoride levels in drinking water in rural and urban areas was 0.70 and 0.69 mg L⁻¹ respectively, although fluoride concentrations in rural areas showed more variation than urban areas. Moreover, our results showed that the HQ value for consuming water containing fluoride for drinking was higher than the HQ level for dermal contact. Sensitivity analysis illustrated that the drinking water ingestion rate and

fluoride concentration in water were the most important variables affecting the value of HQ_{ing}, while the concentration of fluoride in water and the fraction of skin in contact with water were the most important variables in the value of HQ_{derm} in dermal contact in three age groups. As a result, the overall HQ in the studied areas was less than 1, indicating that there was negligible risk of non-carcinogenic effect of exposure to fluoride.

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