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Fluoride contamination in groundwater resources in the southern Iran and its related human health risks

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ABSTRACT

The occurrence of excessive fluoride levels in groundwater has to be recognized as a threat to human health. Based on fluoride concentrations in groundwater samples in the Larestan County, Iran, the health-risk assessment such as chronic daily intakes (CDI) and hazard quotient (HQ) were computed to assess the suitability of groundwater for human consumption in 2018. In 48.27% of the water samples, the level of fluoride exceeds the desirable limits of 1.5 set by WHO. The results also showed that approximately 70.6, 48.2 and 34.4% of HQ values for children, teenagers and infants in these areas were above the safety level of 1, indicating that these age groups are facing to risk of fluoride through drinking water consumption. The health risk was in the order of: children > women > men. The study provides information to the government authorities, water and sewage organizations and health professionals concerned with water supply to provide water with optimum fluoride level.

Keywords: Groundwater; Fluoride contamination; Risk assessment; Larestan, Iran

1. Introduction

Sufficient, clean, and safe water supply is necessary for human life [1–3]. During the past few decades, the rapid population growth, urbanization, industrialization, and improper utilization of resources have led to the deterioration of soil and water quality in many regions of the world [4–6]. Sustainable access to safe potable water has been achieved in many developed nations of the world, but this is not true for different developing nations [7]. About 80% of the diseases around the world is due to unsafe drinking water quality [8,9].

Groundwater is a renewable resource and as the major, important fresh resource on earth specifically in arid and semi-arid regions [10]. Generally, groundwater comprises only 0.6% of the total water resources available on the earth [11]. Groundwater is usually considered as the best alternative water resource among rivers, streams, lakes, dams etc., due to natural protection from contamination when compared to surface and perceived normal filtration and treatment as water moves down during rainy days and also due to least treatment processes required including disinfection [12]. Despite the evident safety of groundwater for drink-

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ing, several agents like the geology and chemical characteristics of the aquifer, climate, and human activities can affect the quality of groundwater [13]. However, groundwater contamination has increasingly become a major concern throughout the world, specifically in regions where water shortage is considerable [10]. In Iran, a sparsely populated and almost dry country, much of the residents in urban and rural areas are reliant on groundwater for domestic purposes. Therefore, the prevention of groundwater pollution is necessary for successful groundwater resource management and vulnerability assessment.

Fluorine, fluoride (F^-) in aqueous environments, is the world's 13th abundant component and comprises about 0.08% of the earth's crust. High levels of fluoride in water may be due to natural processes (weathering fluoride containing rocks and minerals) or human activities (industries like aluminum and zinc smelters, fertilizer and pesticide industries, electronic industries) [14,15]. Fluoride is ubiquitous in nature and is considered worldwide as one of the main toxic inorganic contaminants in water [16,17]. The origin of fluoride in water is mostly geogenic [18,19]. Various rock forming minerals like fluorite, apatite, topaz, simple halides such as carobbite, silicates such as muscovite and a range of amphibole and mica have fluoride in their structures [20,21]. Physical, chemical and biological weathering of these rocks release fluoride into soils and subsequently into water [22]. Generally, soil contains about 330 ppm of fluoride. The little amount of fluoride normally exists in the water, air, plants and animals. Drinking water is the main source of naturally occurring fluoride intake [23]. Fluoride exists in all natural waters at some level [15]. Typical levels of fluoride in seawater, rivers and lakes are 1 mg/L and < 0.5 mg/L, respectively [24]. Its level in groundwater resources usually ranges from 0.1 to 10 mg/L [25]. Fluoride is rapidly absorbed from the digestive tract, with an absorption estimation in range of 75–100% [26]. The little amount of fluoride is necessary for maintenance and solidification of bones and prevention of dental cavity especially for young children [27,28]. Efficient fluoride intake during teeth development leads to long lasting protection against enamel fluorosis and dental cavity [24]. High intake of the element can damage teeth and skeletons and cause dental and skeletal fluorosis due to the attraction of negative fluoride ion by the positively charged calcium ion in bone and teeth. Furthermore, its exceeded concentrations can cause digestive and nervous system problems, kidney and respiratory problems, impaired development of intelligence in children, decreased birth rate, along with myopathy [9,29–32]. Fluorosis in its moderate forms usually appears as unnoticeable, tiny white streaks in the enamels of the teeth. In its extreme form, teeth become brown, pitted, rough and hard to remove. These spots are irreversible and may blacken over time [33]. The severity of dental fluorosis varies depending on the intake concentration of fluoride, the age, individual response, body weight, degree of physical activity, nutrition, and bone growth rate [34]. Long term intake of fluoride contaminated drinking water may increase crippling bone deformities and even cancer [35].

The fluoride contamination in drinking water is responsible for 65% of endemic fluorosis in the world [34]. Based on the world health organization (WHO) guidelines for drinking water, the acceptable range of fluoride in drinking water is between 0.5 and 1.5 mg/L. WHO reported that more than 200 million people from twenty five nations are using drinking water with a fluoride level more than the guideline value of 1.5 mg/L [25]. Fluoride exposure from water depends on the amount of water ingested through itself and the quantity of water ingested via foodstuffs by means of water used for cooking and their fluoride content [36,37]. Literature reviews have shown the occurrence of dental fluorosis even if the residents use drinking water with fluoride <1.0 mg/L [38,39]. High levels of fluoride in groundwater resources is reported in many countries especially in India, China, Sri Lanka, Iran, Pakistan,Syria, Jordan, Russia, Nigeria, Kenya, Morocco, Argentina, USA, Mexico, Brazil, and etc. [40,41]. There is wide variation in fluoride concentrations in the natural waters of different regions of Iran with concentrations higher and lower than the optimal range recommended by the WHO for drinking water encountered [16,42–48]. Successful and safe preventive fluoride strategies need a comprehensive awareness of the exact level of the element drinking water. Therefore, the magnitude and distribution of fluoride in groundwater/drinking water have gained universal attention, due to deleterious impacts on human health. The various chemical, biological and physical characteristics of drinking water should be monitored regularly to guarantee consumption of water within acceptable limits regarding elements present in the water [49–51]. Hence, there is an urgent need to evaluate water quality in Larestan County due to high levels of fluoride reported by health professionals in this area. Within the scope of this problem and needs, the main aims of this research was to assess the status of water quality from groundwater resources situated at Larestan County by analyzing fluoride concentration and determining potential health risk due to exposure of residents (infants, children, teenagers, and adults) to fluoride. This work can provide helpful information for future work planning and important to predict the potential health issues from fluoride exposure.

2. Materials and methods

2.1. Study area details

The study area comprises of rural and urban areas of Larestan County. This County is located in Fars province in southern Iran covering an area of 34720 km². Its geographical coordinates are 53°87′–55°44′ E and 27°21′–28°21′ N. The county has 9 cities including Lar, Evaz, Beyram, Banaruiyeh, Fishvar, Juyom, Khur, Latifi, and Emaddeh. This county has 6 districts: the central district, Beyram, Evazdistrict, Banaruiyeh, Sahray-ye Bagh, and Juyom. Its population was 226879 people. The climate is arid and dry with a mean temperature of 34°C. Mean elevation from sea level is 792 m. The study area has a limited amount of rainfall and high temperatures with an average annual rainfall of 150 mm. The geology map of the study area is shown in Fig. 2. The reasons for selection of the study area are high levels of fluoride reported by health professionals and previous studies [19,52–54] in drinking water samples and prevalence of dental fluorosis among residents living in these areas. In this research, both rural and urban areas of Larestan County were studied. The studied areas were coded as: 1-58.

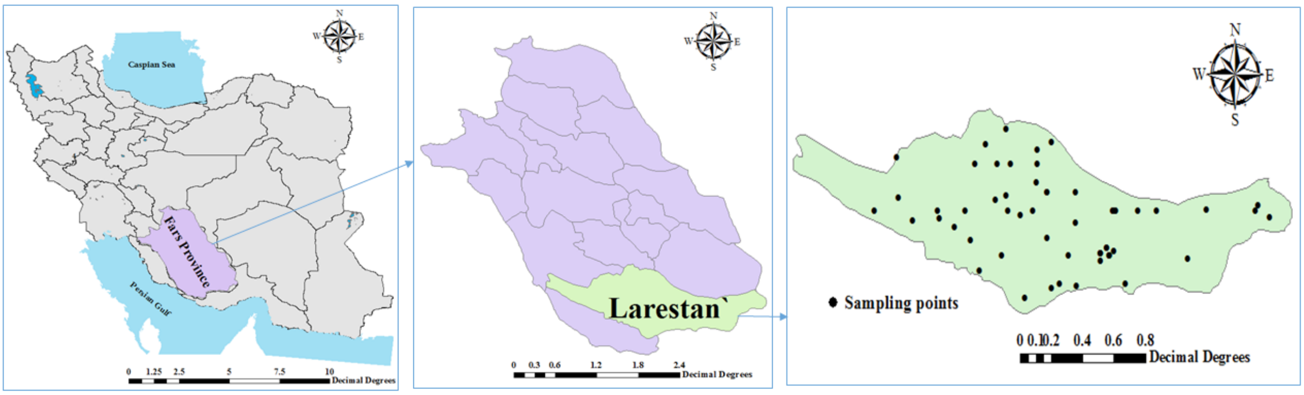

Fig. 1. Map of Larestan County showing the location of sampling sites.

2.2. Sampling and analysis

Groundwater is the main water resource of the communities in the study area. A total of 58 groundwater samples were collected from 49 wells in April and May, 2018. In some places two samples were taken due to having more than one groundwater resource used. To ensure obtaining a good representative sampling of the entire county, sampling sites were selected considering the distribution of rural and urban areas, and verified and confirmed by health experts. Locations of sampling sites in the current work are shown in Fig. 1. In this study, groundwater samples were collected from the main sources of water, applied for drinking, cooking and other domestic purposes. The samples were collected in clean, sterilized 500 ml high-density polyethylene bottles and were immediately carried to the laboratory for chemical analysis. The sampling containers were rinsed at least three times with the sampled water before collection. The bottles were kept at 4°C before being analyzed. The analysis was carried out according to Standard Methods for the Examination of Water and Wastewater book [55]. Finally, fluoride concentrations of the samples were analyzed using Spectrophotometer (DR-5000, USA) and obtained limits of determination (LOD) and quantification (LOQ) were 0.12 mg/L and 0.37 mg/L, respectively.

2.3. Health risk assessment

Fluoride is a naturally occurring element that has been shown to cause significant effects in population both in low and high concentrations [56]. Therefore, it is very important to know the health risks induced by the daily intake of excessive levels of fluoride through drinking water. Human health risk assessment is the most common methodology proposed by the United States Environmental Protection Agency (USEPA) and is described as the process to estimate the nature and probability of harmful health impacts in people who drink highly contaminated water over a specified time period. This method was introduced to determine the degree of fluoride exposure and its tendency based on the toxicity and the response on human health. The health risks induced by natural and man-made contaminants can be categorized into carcinogenic risk and non-carcinogenic risk based on their characteristics [57–59].

Generally, human exposure risk routes of a person to contaminants could occur via 3 main routes including inhalation through nose and mouth, direct intake (ingestion) and dermal absorption via skin contact [60]. Generally, drinking water ingestion and skin adsorption are the predominant routes to induce health risks to people for groundwater. Many researches have demonstrated that the health risks of contaminated groundwater via dermal absorption can be ignored in comparison with the drinking water ingestion [61,62]. One of the main exposure routes to fluoride is through drinking water consumption. Therefore, in this work fluoride exposure to human through water consumption (ingestion) has been assessed based on chronic daily intake (CDI) and hazard quotient (HQ) equations. The CDI value per kilogram of body weight per day (mg/kg/day) for non-carcinogenic and carcinogenic contaminants through direct ingestion pathways of fluoride was computed by considering the rate of water intake, the frequency of exposure, the exposure duration, the averaging time, and the body weight of residents using the following formula proposed by USEPA [51,63,64]:

$$\text{CDI} = \frac{\text{C.DI.F.ED}}{\text{BW.AT}} \quad (1)$$

where C is fluoride concentration in water (mg/L), DI is daily water intake (L/d), F is exposure frequency (d/y), ED is exposure duration (y), BW is average body weight (kg), and AT is averaging time (d).

The studied people were classified into four groups: adults (≥16 years), teenagers (6 to < 16 years), children (2 to < 6 years) and infants (less than 2 years). The parameters used for the calculation of CDI values are obtained from a previous study [65]. The average water consumption rates in adults, teenagers, children, and infants were considered 2.5, 2, 0.85, and 0.08 L/d, respectively. Body weights of target groups were considered 78, 50, 15 and 10 kg, respectively.

The hazard quotient (HQ) is calculated using the following equation [66,67]:

$$\text{HQ} = \frac{\text{CDI}}{\text{RfD}} \quad (2)$$

where RfD is the reference dosage of fluoride in mg/kg/d.

Considering oral fluoride intake and its non-carcinogenic character based on the USEPA risk assessment technique, a recommended RfD of 0.06 mg/kg/d was applied

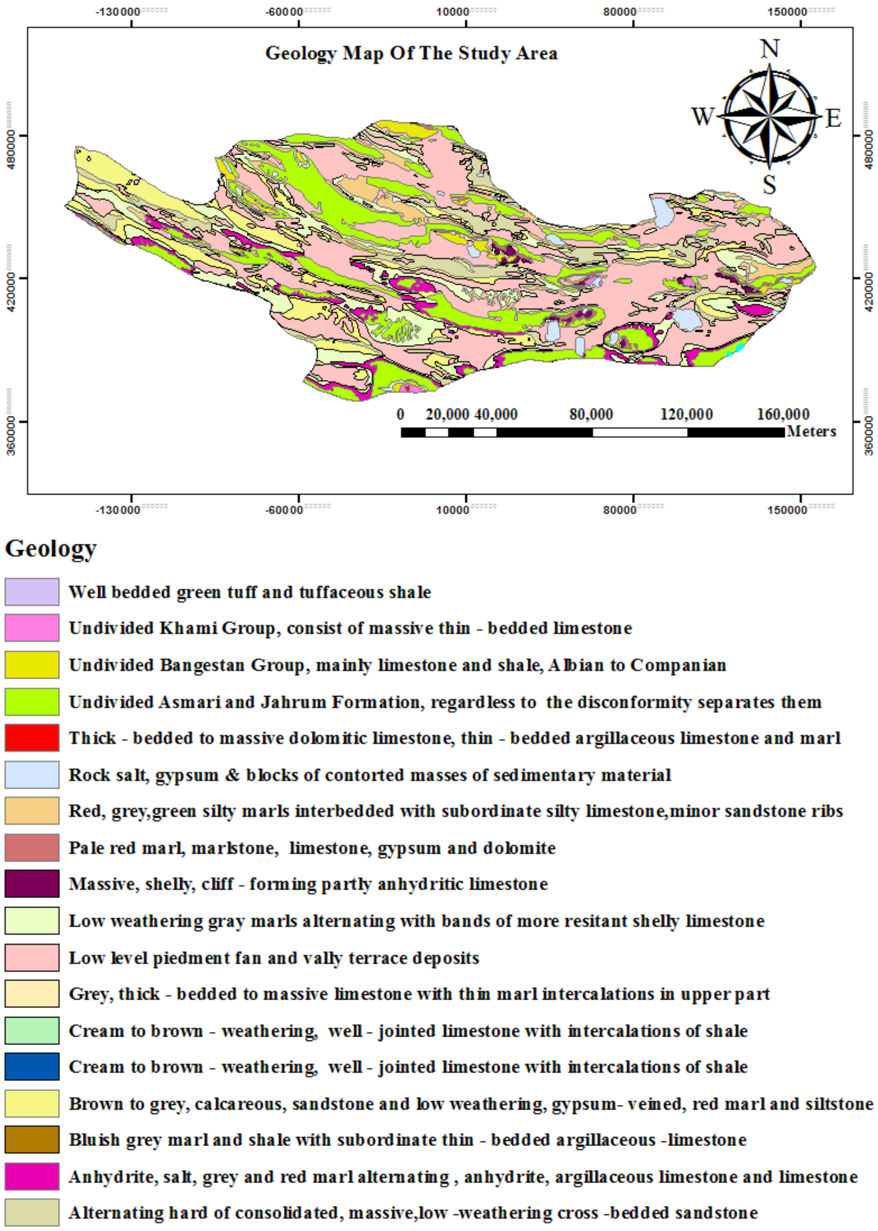

Fig. 2. Geology map of study area.

for HQ calculations. Deleterious health impacts should be considered cautiously, and if HQ > 1, it indicates a high risk of the contaminant with long term health risks; if HQ < 1, it shows there is no health risk [65]. The higher the HQ values, the higher is the risk level of the contaminant indicating long term health hazard impacts increasing.

3. Results and discussions

3.1. Groundwater fluoride concentration and its distribution in Larestan County

Contamination of the natural waters especially groundwater resources by fluoride is a worldwide issue because it is indestructible and has deleterious impacts on human health when it exceeds a certain concentration. Therefore, monitoring this water contaminant is essential for safety assessment. In the current research, the value of fluoride in the study area groundwater was in the range 0.35–3.46 mg/L, with a mean of 1.64 mg/L (Table 1). As clearly shown in the table, in 48.27% of the total water samples, the concentration of fluoride was higher than its permissible limit of 1.5 mg/L for drinking water set by WHO. High fluoride concentrations were determined in the villages of KohneBorhan (3.46 mg/L), Baluchi (3.44 mg/L), and Mehrabad (3.28 mg/L). The main reason for the high level of fluoride in water in many areas in the current research is the existence of an abundant source of fluoride-rich minerals. The distribution of sampling sites, as well as fluoride concentrations in these areas is shown in Fig. 3.

Table 1
Sampling points and population in studied sites in Larestan

Code	Location	Population	Fluoride (mg/L)	Code	Location	Population	Fluoride (mg/L)
1	Emaddeh	6547	2.28	30	Juyom	19601	1.33
2	Emaddeh	6547	2.28	31	Kargah	768	0.45
3	Bagh	1236	1.55	32	Kirishki	433	1.48
4	Zaravan	1036	2.66	33	Beyram	12389	0.50
5	Hormud	472	2.84	34	Najafabad	130	1.12
6	Dashti	1421	2.66	35	Baladeh	3936	1.34
7	Khalur	503	2.86	36	Biram	12389	0.51
8	KohnehBorhan	412	3.46	37	Gerash	10270	1.26
9	Baluchi	200	3.44	38	Khur	6370	0.83
10	Darz	1357	2.36	39	Berak	2355	1.44
11	Mehrabad	576	3.28	40	Latifi	5731	1.71
12	Ramijan	150	2.08	41	Baen	105	1.57
13	Dowlatabad	1069	2.28	42	Shah Gheyb	606	1.07
14	Dehmian	531	2.95	43	Hasanabad	107	1.30
15	Lar	55365	1.84	44	Damcheh	801	1.41
16	Kahneh, Evaz	2553	1.62	45	Fereshtejan	601	0.91
17	Eslamabad	597	2.40	46	Mansurabad	1905	0.95
18	Eslamabad	597	0.80	47	TangeHajiabad	100	1.28
19	Bidshahre	13111	0.64	48	Marme	471	2.73
20	Hud	1248	0.96	49	Khur	6370	1.75
21	Khur	6370	0.84	50	Dehkuyeh	3560	2.03
22	Ahmad Mahmudi	1078	0.67	51	Fadagh	5210	2.02
23	Ahrari	109	0.62	52	Khalili	1438	2.10
24	Karyan	2068	0.35	53	Hormud	563	2.88
25	KhordehDarreh	224	0.63	54	Hasanabade	107	1.23
26	Chaghan	1108	0.64	55	Ghelar	791	1.67
27	Lagharan	1601	0.65	56	Galat	2613	1.46
28	Jalalabad	137	1.65	57	Karmowstaj	1626	1.49
29	Evaz	14315	1.00	58	Aghoseh	1266	2.73

3.2. Human health risk assessment

3.2.1. The chronic daily intakes (CDI)

The chronic daily intakes (CDI) of fluoride through drinking water consumption were computed based on Eq. (1). CDI values are necessary for the calculation of carcinogenic and non-carcinogenic health risks. The mean CDI values via oral intake of drinking water for infants, children, teenagers, and adults were 0.0131 (0.0028–0.0277) mg/kg/d, 0.0927 (0.0198–0.1961) mg/kg/d, 0.0654 (0.0140–0.1384) mg/kg/d and 0.0524 (0.0112–0.1109) mg/kg/d, respectively. The calculation of CDI values is given in Table 1S.

3.2.2. Hazard quotients

As a developing country, many parts of Iran are faced with serious groundwater contamination. In the study area, groundwater is a key source of drinking water for local residents and high levels of fluoride were reported by health professionals in Larestan County. Therefore, it is important to determine the fluoride concentration and its spatial distribution in the groundwater, and forecast health risk levels to residents in these areas. We considered the fluoride concentration distribution in the groundwater resources of each area and assessed the non-carcinogenic health risk for infants, children, teenagers, and adults. The overall groundwater quality was assessed using the Hazard quotients (HQ) to indicate the suitability for human consumption, and the findings are depicted in Fig. 4.The mean HQ values through drinking water ingestion for infants, children, teenagers, and adults were 0.2180 (0.0467–0.4613) mg/L, 1.5443 (0.3306–3.2678) mg/L, 1.0901 (0.2333–2.3067) mg/L and 0.8735 (0.1870–1.8483) mg/L, respectively, showing that children in the study area are more vulnerable to contaminants compared to other studied age groups. Results showed that calculated non-carcinogenic risk of fluoride for studied groups for each studied area was in the following order:

Children > teenagers > adults > infants.

It can be concluded that approximately 70.6, 48.2 and 34.4% of HQ values for children, teenagers and infants

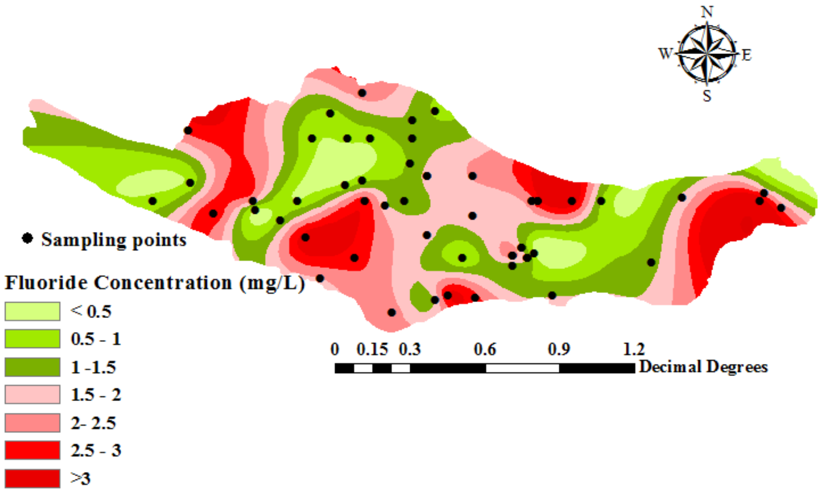

Fig. 3. Map of fluoride concentration in groundwater samples in the study area.

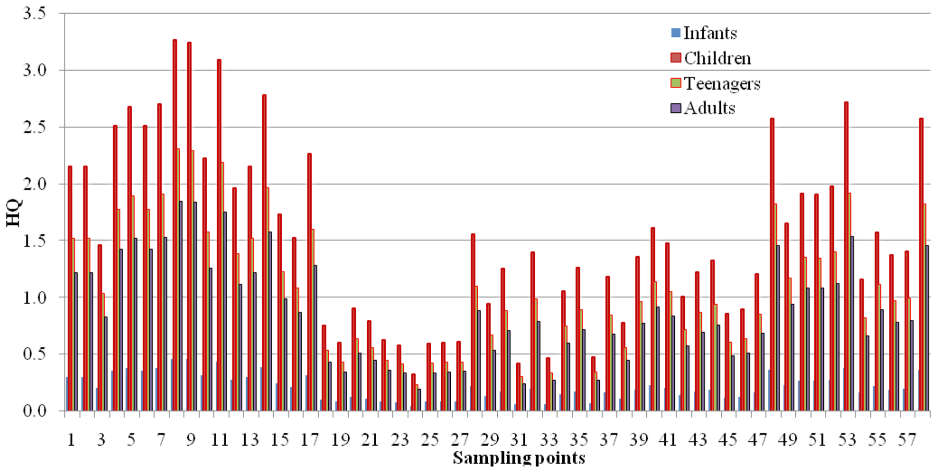

Fig. 4. Human health risk assessment through hazard quotient (HQ) of fluoride ingestion of groundwater in the study area.

in these areas were above the safety level of 1, indicating that these age groups are facing to risk of fluoride through drinking water consumption.

To compare the findings obtained in the present study with those in literature, results of some studies are described. For example, in a work, fluoride in drinking water in 31 provinces of Iran was studied. The range of fluoride was 0.01–3.72 mg/L [42]. Fluoride concentration in drinking water of Qom city, Iran was measured. The results indicated the mean level of fluoride in groundwater was 0.41 mg/L [16]. In another study in Mianeh city, Iran, 14 groundwater sources during four seasons of 2009 were studied for fluoride levels. The range of source fluoride in spring, summer, autumn, and winter seasons were 0.039–0.295 mg/L, 0.17–0.47 mg/L, 0.18–0.48 mg/L, and 0.06–0.4 mg/L, respectively [14]. In another study, 14 villages in Dashtestan with only a groundwater source were analyzed for fluoride. Based on this study, fluoride ranged from 0.99 to 2.50 mg/L in groundwater samples [68]. Health risk assessment in young consumers, children and teenager's groups due to fluoride in drinking water of 28 villages in the Poldasht city, Northwest of Iran was estimated. The findings showed a range of 0.27–10.3 mg/L (mean 1.7 mg/L). The estimated HQ value was above the safe level of 1 for all groups of residents in Aghotlogh and Sari soo areas [65]. In another study, distribution and health risk assessment of natural fluoride in 573 drinking groundwater resources including 473 tube wells, 62 springs, and 38 qanats in the region of Isfahan, center Iran, was investigated, during 2016–2017. The fluoride concentrations were below 0.50 mg/L in 63% of the drinking groundwater samples, 0.51–1.5 mg/L in 33.15%, and above 1.5 mg/L in 3.85% of the samples. The HQ index for children, teens, male and female adults had health hazards (HQ>1) in 51, 17, 28, and 18 of samples, respectively, and children have the highest risk [69].

In another study, groundwater fluoride contamination and its related health risks to human health was performed in 39 rural areas of Gonabad and Bajestan, Iran. The findings showed that fluoride concentrations in 2 rural areas exceeded the WHO guideline. A total of 55 and 4.7% of the studied rural areas in Gonabad and Bajestan, respectively, had fluoride concentrations lower than the minimum WHO recommended value 0.5 mg/L. Health risk quotient (HQ) values for fluoride contamination for 44% and 90% of children and infants in rural areas of Gonabad and Bajestan, respectively, were above the safety level (HQ > 1) [41].

Health risk assessment of fluoride in water distribution network of Mashhad, northeast of Iran, over five consecutive years (from 2012 to 2016) was assessed. The mean annual levels of fluoride in all stations during this period were lower than the maximum permissible limits (1.5 mg/L). Health risk estimation in this study showed that the risk associated with fluoride exposure through drinking water in the area was acceptable because the mean HQ values of fluoride were less than the safety level [67]. In a case study contamination status of fluoride in 104 groundwater samples in hard rock aquifers of Siddipet, Telangana State, India, was evaluated. The study indicated that the concentrations of fluoride in groundwater samples were in range 0.2–2.2 mg/L with a mean of 1.1 mg/L. Nearly 22% of groundwater has more than the permissible limit of fluoride (1.5 mg/L). Based on this study, the majority of the residents living in these areas suffer from dental fluorosis [70].

In another work, groundwater contamination for fluoride in a semi-arid region of Nirmal Province, South India was evaluated. Based on this study, 20.59% of groundwater samples had high fluoride levels, exceeding the maximum permissible limits (1.5 mg/L) set by WHO. The maximum value of HQ-fluoride was 2.78, 3.28 and 3.75 for men, women, and children, respectively, indicating that fluoride may cause deleterious health impacts in the order of:

Children > women > men [71].

Generally fluoride in water originates from the geological formation of the soil that water flows [72,73]. High levels of fluoride in this study was due to the presence of fluoride containing minerals in the area including clay minerals (illite and montmorillonite), mica (biotite and muscovite), fluorapophyllite [53], faulted carbonatic (limestone and dolomite), evaporatic (gypsiferrous) and conglomeratic formations. When considering the risks calculated in the present research, proper actions for many of these high-concentration areas are necessary. Therefore, the relevant organizations should take actions about these areas, installing warning notice boards, and forbid the continuation of groundwater use, or closed sites and finding alternative water resources.

4. Conclusions

In arid and semi-arid regions of Iran, many residents primarily rely on groundwater resources for drinking and other purposes. However, groundwater is the main resource in the current study majorly for drinking purposes and the heavy groundwater exploitation has decreased the groundwater level and water quality in these areas. Health risk analysis indicated that the hazard quotients were higher than the safe limit; therefore, the residents are susceptible to developing different health-related problems. Children were at the greatest health risk among all in the studied areas. The mean levels of fluoride in 48.27% of the groundwater samples were higher than the recommended WHO guideline of 1.5 mg/L for fluoride, and large variances between sampling locations due to different geological formations were observed.

Based on the total health risk assessment of fluoride, 70.6, 48.2 and 34.4%of the samples had a high non-carcinogenic risk, due to exceeding the maximum permissible limit of HQ = 1 approved by the USEPA for children, teenagers and infants, respectively. Results indicated that an estimated non-carcinogenic risk for studied groups were in the following order: children > teenagers > adults > infants. Considering the results of the present study, it is recommended that the responsible organizations should use socioeconomically feasible de-fluoridation techniques with the specific aim of lowering the levels of the water contaminant to meet WHO drinking water range for water supplied to the residents and that alternative sources of water should be developed for the people depending on the sites with high fluoride water. Public health awareness projects should also be performed by relevant agencies and health professionals to increase consciousness regrading deleterious health impacts of consuming fluoride contaminated water above the recommended range. In addition, regular clinical exploration is required to identify individuals with fluorosis for proper medical referral and treatment.

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Supplemantary

Table 1S
CDI (mg/kg/day) values for studied groups in Larestan County

Code	CDI				Code	CDI			
	Infants	Children	Teenagers	Adults		Infants	Children	Teenagers	Adults
1	0.0182	0.1292	0.0912	0.0731	30	0.0106	0.0754	0.0532	0.0426
2	0.0182	0.1292	0.0912	0.0731	31	0.0036	0.0255	0.0180	0.0144
3	0.0124	0.0878	0.0620	0.0497	32	0.0118	0.0839	0.0592	0.0474
4	0.0213	0.1507	0.1064	0.0853	33	0.0040	0.0283	0.0200	0.0160
5	0.0227	0.1609	0.1136	0.0910	34	0.0090	0.0635	0.0448	0.0359
6	0.0213	0.1507	0.1064	0.0853	35	0.0107	0.0759	0.0536	0.0429
7	0.0229	0.1621	0.1144	0.0917	36	0.0041	0.0289	0.0204	0.0163
8	0.0277	0.1961	0.1384	0.1109	37	0.0101	0.0714	0.0504	0.0404
9	0.0275	0.1949	0.1376	0.1103	38	0.0066	0.0470	0.0332	0.0266
10	0.0189	0.1337	0.0944	0.0756	39	0.0115	0.0816	0.0576	0.0462
11	0.0262	0.1859	0.1312	0.1051	40	0.0137	0.0969	0.0684	0.0548
12	0.0166	0.1179	0.0832	0.0667	41	0.0126	0.0890	0.0628	0.0503
13	0.0182	0.1292	0.0912	0.0731	42	0.0086	0.0606	0.0428	0.0343
14	0.0236	0.1672	0.1180	0.0946	43	0.0104	0.0737	0.0520	0.0417
15	0.0147	0.1043	0.0736	0.0590	44	0.0113	0.0799	0.0564	0.0452
16	0.0130	0.0918	0.0648	0.0519	45	0.0073	0.0516	0.0364	0.0292
17	0.0192	0.1360	0.0960	0.0769	46	0.0076	0.0538	0.0380	0.0304
18	0.0064	0.0453	0.0320	0.0256	47	0.0102	0.0725	0.0512	0.0410
19	0.0051	0.0363	0.0256	0.0205	48	0.0218	0.1547	0.1092	0.0875
20	0.0077	0.0544	0.0384	0.0308	49	0.0140	0.0992	0.0700	0.0561
21	0.0067	0.0476	0.0336	0.0269	50	0.0162	0.1150	0.0812	0.0651
22	0.0054	0.0380	0.0268	0.0215	51	0.0162	0.1145	0.0808	0.0647
23	0.0050	0.0351	0.0248	0.0199	52	0.0168	0.1190	0.0840	0.0673
24	0.0028	0.0198	0.0140	0.0112	53	0.0230	0.1632	0.1152	0.0923
25	0.0050	0.0357	0.0252	0.0202	54	0.0098	0.0697	0.0492	0.0394
26	0.0051	0.0363	0.0256	0.0205	55	0.0134	0.0946	0.0668	0.0535
27	0.0052	0.0368	0.0260	0.0208	56	0.0117	0.0827	0.0584	0.0468
28	0.0132	0.0935	0.0660	0.0529	57	0.0119	0.0844	0.0596	0.0478
29	0.0080	0.0567	0.0400	0.0321	58	0.0218	0.1547	0.1092	0.0875