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Assessing fluoride and nitrate contaminants in drinking water resources and their health risk assessment in a semiarid region of southwest Iran

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ABSTRACT

Spatial and temporal variability of fluoride and nitrate in groundwater resources of rural area of Saravan county, Iran were analyzed 2013–2017 using GIS (V10.3). The results were used for human health risk assessment, using probabilistic techniques. The annual mean concentration of fluoride during 2013 to 2017 in villages of Saravan county was 0.42, 0.62, 0.67, 0.57, and 0.55 mg L^{-1}, and the maximum values were 1.1, 1.42, 1.76, 1.46, and 1.3 mg L^{-1}, respectively. Also, the annual mean concentration of nitrate during 2013 to 2017 in Saravan county villages were 12.53, 14.1, 15.43, 14.34, and 14.02, and the maximum values were 66.1, 80, 35.6, 76.12, and 40 mg L^{-1}, respectively. Spatial, temporal, and spatiotemporal variability of fluoride and nitrate in groundwater resources were relatively not constant over the years. The calculated HQ value of fluoride was for groups of infant (0.013–0.235), children (0.035–1.83), teenagers (0.067–1.1), and adults (0.053–0.94). The maximum HQ value was >1 for children and teenagers during the study period. Meanwhile, the HQ values of nitrate were (0.005–0.4) for infants, (0.035–2.69) children, (0.025–2) teenagers, and (0.02–1.6) for adults. Hence, the maximum HQ value was >1 for children, teenagers, and adult during the study period. This study showed that children, teenagers, and adult are vulnerable groups at risk of non-carcinogenic hazards for being exposed to drinking water with high fluoride and nitrate concentration.

Keywords: Drinking water; Nitrate; Fluoride; Human risk assessment; Saravan; GIS

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1. Introduction

About 80% of the diseases around the world are due to poor quality of drinking water. Reduced quality of ground water in arid and semiarid regions of southern Iran is due to geogenic source and human source. Among the many harmful contaminants, nitrate and fluoride are more wide spread. Nitrate, due to its high water solubility, is possibly the most widespread groundwater contaminant in the world, and in recent years, nitrate contamination in drinking water resource has become a worldwide environmental problem. The high concentration of nitrate in drinking water resources can cause serious problem for human health such as cancers, liver damage, and blue baby syndrome, especially in infants [1–7]. Recent epidemiologic researches have shown the relation between high nitrate contamination and the potential risk of specific cancers, adverse reproductive outcomes, and other health issues in the context of current regulatory limit for nitrate in drinking water. Nitrate contaminated water supplies have contributed to the outbreaks of infectious diseases in humans [8,9]. Literature review showed that nitrate contamination can causes diabetes. In addition, nitrates in surface water can stimulate eutrophication, causing water pollution due to heavy algal growth [10]. In this regards many countries have set standard limit as 10 mg L^{-1} of nitrate-nitrogen (NO_3–N) in drinking water. Therefore, the WHO guideline limit of 50 mg L^{-1} nitrate (~10 mg L^{-1} as N) is applied [11–16]. Sources of nitrate contamination in water mainly come from point and non-point sources such as agricultural runoff, landfill leachate, leaking septic tanks, municipal storm water runoff, animal feeding operations and industrial waste [17–19]. Another drinking water contaminant is fluoride. It has been reported that fluoride contamination is responsible for 65% of endemic fluorosis around the world. Also, other studies have suggested that about 200 million people are indirectly exposed to high concentrations of fluoride from groundwater resources (25 countries) [20–23]. It is well-known that low concentration of fluoride is essential for dental protection and beneficial for human health; however, when its amount exceeded the standard levels could adversely lead to dental and skeletal fluorosis [24–26]. The concentration of fluoride in groundwater resources depends on several factors, especially geological structure of the subjected area. Furthermore, it has been shown that long-term adsorption of 5–10 mg L^{-1} excess concentration of fluoride in drinking water could result in non-skeletal fluorosis by adversely affecting non-calcified tissues. Drinking water containing 10 mg of fluoride from birth to adolescence can result in many health problems, such as hypertension, infertility, neurological problems, Alzheimer's diseases, thyroid, cancer, and arthritis. In response to the potentially harmful effects of high concentration-fluoride waters, WHO has set guideline for drinking water quality of 1.5 mg L^{-1} [26–31]. The concentration of fluoride and nitrate in water is one of the most important risk indictor for health outcomes. Hence, regular monitoring of fluoride in drinking water is critical to protect public health. GIS has been widely used as a tool to determine spatial dispersion and relationships between subjected environmental factors and health-related issues, considering their exposure routes. With respect to nitrate and fluoride concentration in drinking water, many studies used GIS risk assessment to categorize the study area from low to high risk [26,31,32]. In this research, human health risk assessment was performed by calculating the chronic daily intake (CDI) and hazard quotient (HQ) of nitrate and fluoride through human oral intake for infants (less than 2 years), children (2 to <6 years), teenagers (6 to <16 years) and adults (≥16 years). The data of this study were collected in five consecutive years 2013–2017. This study aimed to investigate the distributions of fluoride and nitrate concentrations and compare the results with national and international standards as well as its effect on human health in Saravan county in Sistan and Baluchistan province, using GIS to identify areas at risk. The results of this study could be useful to national and regional decision-making organizations, which are involved in safe drinking water supply in Saravan county to prevent long-term potential health risks. Also, this result might be useful for future water resource planning.

2. Materials and methods

2.1. Study areas

Saravan county is located in Sistan and Baluchistan province, Iran encompassing an area of about 9,739 km^2 (Fig. 1) and its aquifers are located in South-East Iran between the latitudes 27°22′15″ N and longitudes 62°20′03″ E. The subjected area is a semi-flat plain region with a gentle slope toward the south with a warm climate and an annual average of 22.1°C in which the highest and lowest temperatures are 45.3°C and –7°C, respectively [33–35].

2.2. Sample analysis

A total of 520 drinking water samples were taken from water well resources in the rural areas of Saravan county in Sistan and Baluchistan province, within the 5-year-monitoring period (2013–2017). Samples were collected in the polythene bottles (1 L) and then transferred to the central laboratory of water and wastewater company. All analysis methods were taken from "Standard Methods for Examination of Water and Wastewater". Fluoride and nitrate content were determined through Spectrophotometer model DR-5000 (Hach Company, Loveland, CO, USA) and compared with WHO guidelines [36–39].

2.3. Human health risk assessments

Human health risk assessment is defined as the probability of harmful impacts on human health including residents, workers, recreational visitors as a result of contaminants and other stressors that might be present in the environment now or in the future [26,40].

Health risks caused by various contaminants entering the human body via different exposure routes are categorized into carcinogenic and non-carcinogenic risks. Carcinogenic risk is the incremental possibility of a person developing any type of cancer during lifetime due to exposure to specified carcinogens [26,40,41].

Non-cancer risk is estimated by considering an exposure level during a specified time period, with a reference dosage obtained for a similar exposure period [26,42]. Consequently,

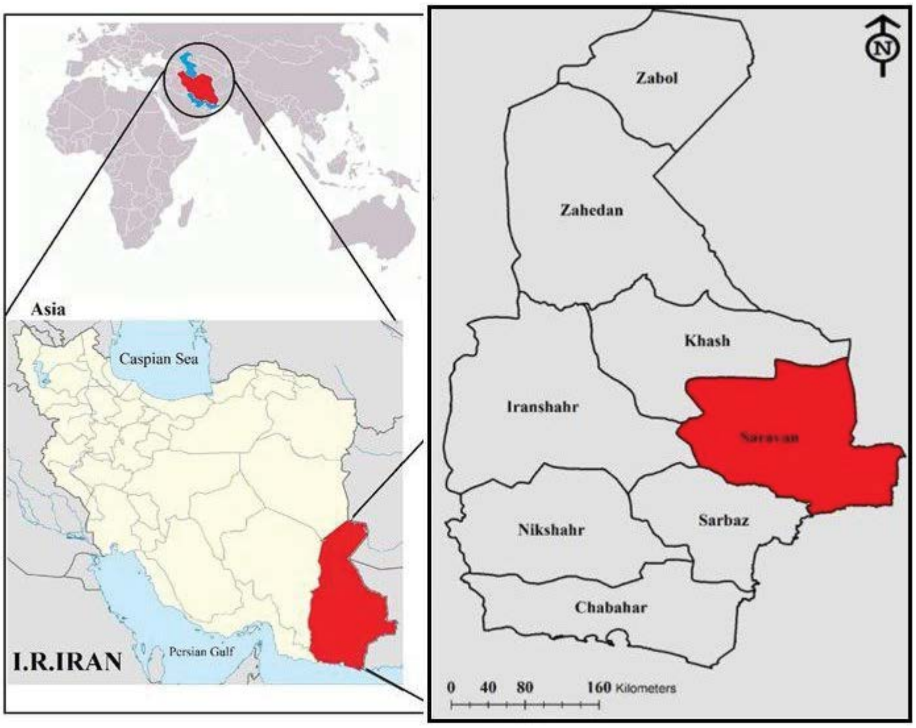

Fig. 1. Location of the study areas, Saravan county in Sistan and Baluchistan province, Iran.

the quantitative health risk assessment of nitrate and fluoride through consumption of drinking water resource was evaluated in rural area population of Saravan county, Sistan and Baluchistan province. For this purpose, drinking water samples were taken from different villages in the study period.

In this study, people were categorized into four age groups based on physiological and behavioral differences same as Yousefi et al. [26] study as follows: infants (less than 2 years), children (2 to <6 years), teenagers (6 to <16 years) and adults (≥16 years). Exposure to nitrate and fluoride were calculated in these groups using Eq. (1):

$$\text{EDI} = \frac{C_f - C_d}{B_w} \quad (1)$$

Estimated daily intake (EDI) of nitrate and fluoride was calculated based on the daily average consumption of drinking water (C_d), concentration of nitrate and fluoride in drinking water (C_f), and body weight (B_w).

In this study, default body weights of 10, 15, 50, and 78 kg were considered for infants, children, teenagers, and adults, respectively.

EDI is expressed in unit of milligrams per kilogram of body weight per day. The water consumption data and body weight were estimated based on a questionnaire asked of the target groups (infants and children parents, teenagers, and adults). The average water consumption rates in infants (0–2 years old), children (2–6 years old), teenagers (6–16 years old), and adults (≥16 years old) were 0.08, 0.85, 2, and 2.5 L d^{-1}, respectively (Table 1).

Table 1
Parameters used in the present study for health exposure assessment in drinking water [43]

Parameter	Risk exposure factors	Values for groups			
		Infants	Children	Teenagers	Adults
Fluoride	C_f, mg L^{-1}	–	–	–	–
	C_d, L d^{-1}	0.08	0.85	2	2.5
	B_w, kg	10	15	50	78
	RfD, mg kg^{-1} d^{-1}	0.06	0.06	0.06	0.06
Nitrate	C_f, mg L^{-1}	–	–	–	–
	C_d, L d^{-1}	0.08	0.85	2	2.5
	B_w, kg	10	15	50	78
	RfD, mg kg^{-1} d^{-1}	1.6	1.6	1.6	1.6

The non-carcinogenic risk of nitrate and fluoride to human health risk assessments is expressed as HQ. It was estimated to evaluate nitrate and fluoride risks using the following formula:

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

In the above formula, RfD is the reference dosage of fluoride in mg kg^{-1} d^{-1}. The reference dose is used in human health risk assessments and it is an estimate of a daily exposure of human population. The oral reference doses of nitrate and fluoride (1.6 and 0.06 mg kg^{-1} d^{-1}) were obtained from the database of Integrated Risk Information System, USEPA [43]. The HQ is the ratio between the EDI and RfD; an HQ value less than one indicates that it is unlikely even for sensitive populations to experience adverse health effects. When the HQ value is higher than 1, it indicates that the non-carcinogenic risk exceeds the acceptable level and adverse health effects are possible.

To describe data in the present research, we calculated mean, standard deviation, median, and range. Then, the values of CDI and HQ were estimated to evaluate the non-carcinogenic health impacts due to fluoride and nitrate exposure via oral exposures to water in urban population of rural area in Saravan county in Sistan and Baluchistan province, Iran. Finally, all the data for the related parameters were estimated, using Microsoft Excel 2016 software. After entering the concentrations of nitrate and fluoride, layers were prepared in ArcGIS (V10.3) based on field information. Distribution map is illustrated in Figs. 4 and 5.

3. Results and discussion

This study was conducted to evaluate fluoride and nitrate concentration of drinking water from groundwater resources of rural area of Saravan county and also to estimate the non-carcinogenic health risks assessment of fluoride and nitrate associated with drinking water in four categories of people resident including infants, children, teenagers, and adults. The annual mean concentrations of nitrate and fluoride during study period from 520 different points of drinking water resource in rural areas of Saravan county are summarized in Table 2. Figs. 2 and 3 compare fluoride and nitrate concentration with WHO guideline. Also, geological distribution of nitrate and fluoride in the study area is shown in Figs. 4 and 5. Fluoride and nitrate exposure levels for different rural population HQ and EDI were observed in four age groups shown in Tables 3 and 4.

3.1. Fluoride contamination in groundwater

The annual mean concentration of fluoride during the years 2013 to 2017 in villages of Saravan county were 0.42 (0.1–1.1) mg L^{-1}, 0.62 (0.13–1.42) mg L^{-1}, 0.67 (0.11–1.76) mg L^{-1}, 0.57 (0.1–1.46) mg L^{-1}, and 0.55 (0.1–1.39) mg L^{-1}, respectively. By comparing annual fluoride concentration in the studied area with WHO guideline during 2013–2017, 72%, 35.2%, 45.7%, 42%, and 48.7% of sample were below

Fig. 2. Comparison of annual mean fluoride concentrations in the studied area with WHO guideline of fluoride in drinking water.

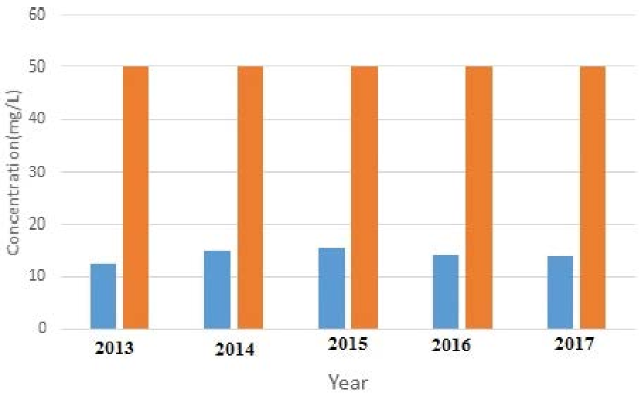

Fig. 3. Comparison of annual mean nitrate concentrations in the studied area with WHO guideline of nitrate in drinking water.

Table 2
Statistically analyzed concentrations of nitrate and fluoride during the study periods (2013–2017) in rural area of Saravan county, Sistan and Baluchistan province, Iran

Statistical	Year-Fluoride concentration					Year-Nitrate concentration				
	2013	2014	2015	2016	2017	2013	2014	2015	2016	2017
Mean	0.42	0.62	0.67	0.57	0.55	12.53	14.81	15.43	14.34	14.02
Max	1.1	1.42	1.76	1.46	1.39	66.1	80	35.6	76.12	40
Min	0.1	0.13	0.11	0.1	0.1	1	1	1.32	1	1
SD	0.25	0.24	0.33	0.23	0.21	10.75	13.78	8.37	10.5	6.81
N	75	108	46	116	175	75	108	46	116	174

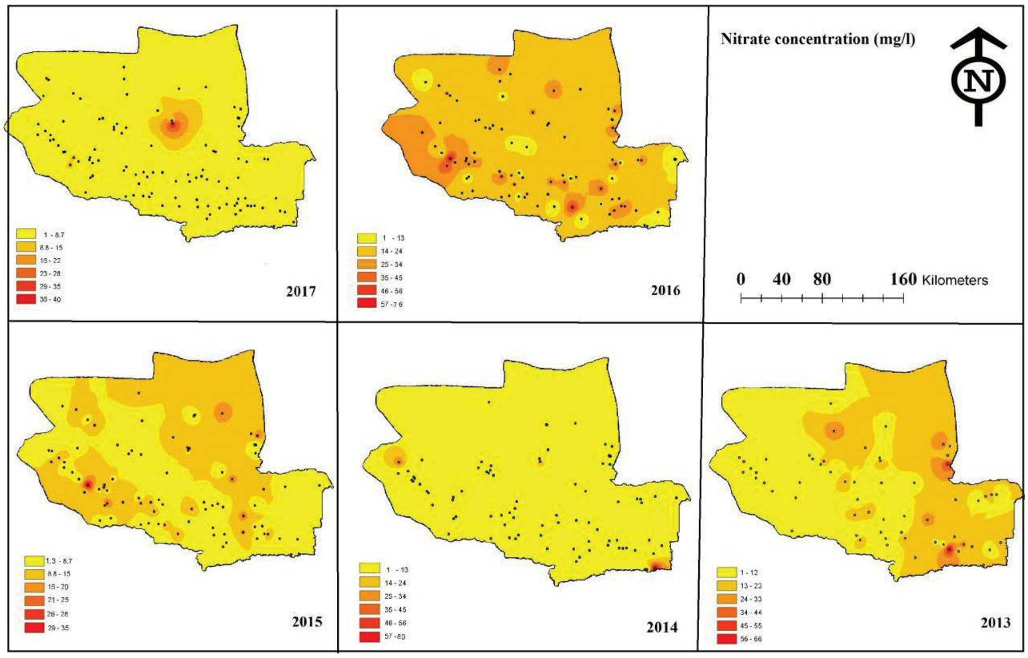

Fig. 4. Spatial distribution of nitrate contamination in the groundwater within the studied area (2013–2017).

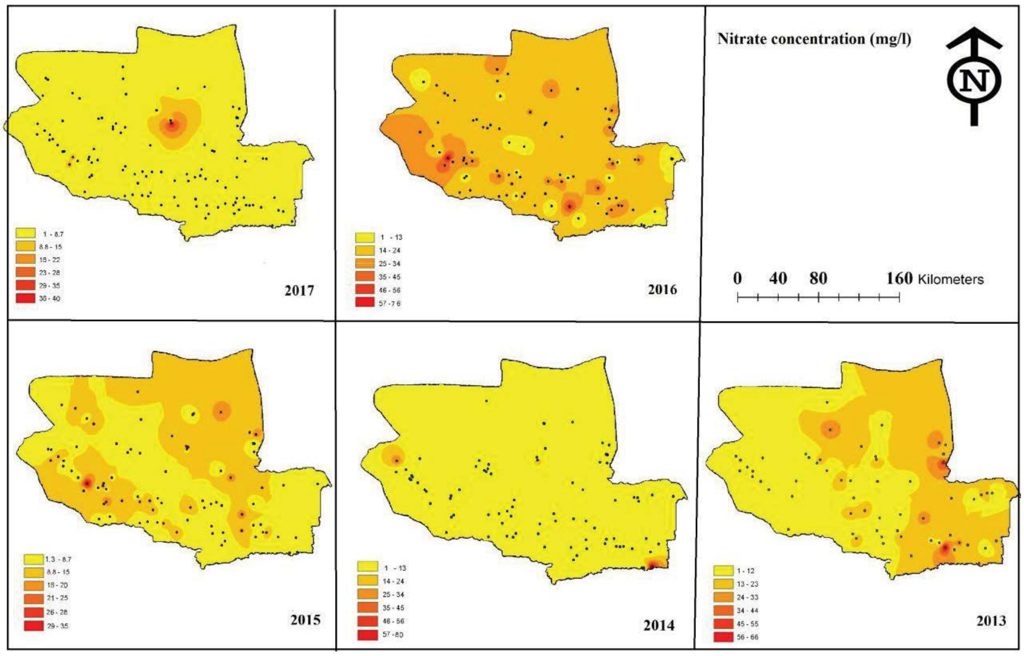

Fig. 5. Spatial distribution of fluoride contamination in the groundwater within the studied area (2013–2017).

0.5 mg L^{-1}, which was the minimum limit by WHO. The results indicates that only 28%, 64.8%, 50%, 57.8%, and 51.7% of water samples were in the range of WHO guideline.

These results are similar to the findings by other researchers, such as Amouei et al. [20] that found the fluoride contamination in villages of Khaf county was 0.11 to 3.59 mg L^{-1}. This research reported that the fluoride level in 31% of the samples was less than the standard, and 4% above the standard and 64% within permissible standard range [20].

A study conducted by Mirzabeygi et al. [31] showed that the average concentration of fluoride was 2.92 mg L^{-1} (range: 0.9–6 mg L^{-1}), also in half of the villages, the concentration range of this element was over the standard levels (1.5 mg L^{-1}) given by WHO.

The results of the study by Chen et al. [44] showed that fluoride content ranging from 0.11 to 6.33 mg L^{-1} with a mean of 0.85 mg L^{-1}. It was founded that 4 and 11 samples had high concentrations exceeding the permissible limits for F^- based

Table 3
Statistically analyzed estimated daily intake (EDI) and hazard quotient (HQ) of fluoride concentration during the study period 2013–2017 for infants, children, teenagers, and adults

Year	Statistical	EDI				HQ			
		Infants	Children	Teenagers	Adults	Infants	Children	Teenagers	Adults
2013	Mean	0.003	0.024	0.017	0.013	0.056	0.398	0.281	0.225
	Max	0.009	0.062	0.044	0.035	0.147	1.039	0.733	0.588
	Min	0.001	0.006	0.004	0.003	0.013	0.094	0.067	0.053
2014	Mean	0.005	0.035	0.025	0.020	0.083	0.589	0.416	0.333
	Max	0.011	0.080	0.057	0.046	0.189	1.341	0.947	0.759
	Min	0.001	0.007	0.005	0.004	0.017	0.123	0.087	0.069
2015	Mean	0.005	0.038	0.027	0.021	0.089	0.629	0.444	0.356
	Max	0.014	0.100	0.070	0.056	0.235	1.662	1.173	0.940
	Min	0.001	0.006	0.004	0.004	0.015	0.104	0.073	0.059
2016	Mean	0.005	0.032	0.023	0.018	0.076	0.539	0.381	0.305
	Max	0.012	0.083	0.058	0.047	0.195	1.379	0.973	0.780
	Min	0.001	0.006	0.004	0.003	0.013	0.094	0.067	0.053
2017	Mean	0.004	0.031	0.022	0.018	0.073	0.517	0.365	0.293
	Max	0.011	0.079	0.056	0.045	0.185	1.313	0.927	0.743
	Min	0.001	0.006	0.004	0.003	0.013	0.094	0.067	0.053

Table 4
Statistically analyzed water estimated daily intake (EDI) and hazard quotient (HQ) of nitrate concentration during the study period (2013–2017) for infants, children, teenagers, and adults

Year	Statistical	EDI				HQ			
		Infants	Children	Teenagers	Adults	Infants	Children	Teenagers	Adults
2013	Mean	0.100	0.710	0.501	0.402	0.063	0.444	0.313	0.251
	Max	0.529	3.746	2.644	2.119	0.331	2.341	1.653	1.324
	Min	0.008	0.057	0.040	0.032	0.005	0.035	0.025	0.020
2014	Mean	0.118	0.839	0.592	0.475	0.074	0.524	0.370	0.297
	Max	0.640	4.533	3.200	2.564	0.400	2.833	2.000	1.603
	Min	0.008	0.057	0.040	0.032	0.005	0.035	0.025	0.020
2015	Mean	0.123	0.874	0.617	0.495	0.077	0.546	0.386	0.309
	Max	0.285	2.017	1.424	1.141	0.178	1.261	0.890	0.713
	Min	0.011	0.075	0.053	0.042	0.007	0.047	0.033	0.026
2016	Mean	0.115	0.812	0.573	0.460	0.072	0.508	0.358	0.287
	Max	0.609	4.313	3.045	2.440	0.381	2.696	1.903	1.525
	Min	0.008	0.057	0.040	0.032	0.005	0.035	0.025	0.020
2017	Mean	0.112	0.795	0.561	0.449	0.070	0.497	0.351	0.281
	Max	0.320	2.267	1.600	1.282	0.200	1.417	1.000	0.801
	Min	0.008	0.057	0.040	0.032	0.005	0.035	0.025	0.020

on the WHO and Chinese standards (1.5 and 1.0 mg L^{-1}), respectively [44].

3.2. Nitrate contamination in groundwater

The annual mean concentration of nitrate during 2013–2017 in villages of Saravan county was 12.53 (1–66.1) mg L^{-1}, 14.1 (1–80.1) mg L^{-1}, 15.43 (1.32–35.6) mg L^{-1}, 14.34 (1–76.12) mg L^{-1}, and 14.02 (1–40) mg L^{-1}, respectively. By comparing the range of annual nitrate concentration in the studied area with WHO guideline of nitrate in drinking water during 2013–2017 in rural area of Saravan county were 98.3%, 97.7%, 100%, 98.3%, and 100% sample were below 50 mg L^{-1}, which was the maximum limit by WHO guideline.

The results indicate that only 1.7%, 2.3%, 0%, 1.7%, and 0% of water samples were higher than the range set by WHO guideline.

Chen et al. [44] study on nitrate and fluoride contaminants in drinking water and their health risk to rural residents living in a semiarid region of northwest China indicated that the nitrate concentrations in the groundwater samples varied from 2.66 to 103 mg L^{-1}. Thirty samples had high nitrate level exceeding the acceptable limits of WHO (10 mg L^{-1}), and the nitrate values in 14 samples were above the Chinese standard (20 mg L^{-1}) for drinking purpose [44].

The study by Mohammadi et al. [45] showed that the nitrate concentration in water samples of Bandar-e Gaz city was lower than the permissible limit by national standard of Iran and WHO.

3.3. Human Health Risk Assessments

The minimum, maximum, and mean EDI and HQ of fluoride and nitrate during 2013–2017 infants, children, teenagers, and adults are shown in Tables 3 and 4.

The HQ values for fluoride were calculated for infants during the study period (2013–2017) 0.056 (0.013–0.147), 0.083 (0.017–0.189), 0.089 (0.015–0.235), 0.076 (0.013–0.195), 0.073 (0.013–0.185), respectively (Table 3). Also, the HQ values for children during the study period (2013–2017) were 0.398 (0.094–1.039), 0.589 (0.123–1.341), 0.629 (0.104–1.662), 0.539 (0.094–1.379), 0.517 (0.094–1.313), respectively. Also the HQ values for teenagers during the study period (2013–2017) were 0.281 (0.067–0.733), 0.416 (0.087–0.947), 0.444 (0.073–1.173), 0.381 (0.067–0.973), 0.365 (0.067–0.927), respectively. Also, HQ values for adults during the study period (2013–2017) were 0.225 (0.053–0.588), 0.333 (0.069–0.759), 0.356 (0.059–0.940), 0.305 (0.053–0.780), 0.293 (0.053–0.743), respectively.

Based on the results, HQ values for fluoride were as follows: infants (0.013–0.235), children (0.035–1.83), teenagers (0.067–1.1), and adults (0.053–0.94). The HQ index for children and teenagers had health hazard (HQ > 1) in 2013–2017 samples. Drinking water resources having a risk more than one were located in the villages of Saravan county. Hence, in this rural area, there are potential risks of fluorosis. In the study by Chen et al. [44] infants and children are the most vulnerable groups, exposed to health risks of fluorosis. Result of research reported by Martínez-Acuña et al. [46] showed that children were at higher risk of health effects through exposure to a high fluoride level in Zacatecas, Mexico. The research results of Guissouma et al. [47] reported that young consumers including infants and children were more exposed to health risk of fluoride in Tunisia. Similar report by Huang et al. [48] showed that highest risk of fluoride was in children group.

Mirzabeygi et al. assessed the concentration data of fluoride and health risk assessment in drinking water in the city of Ardakan, Yazd province, Iran, and showed that HQ value was higher than 1 in 46.4% of the groundwater resource samples amongst infants, children, teenagers and adults. Therefore, it is necessary to take measures in reducing fluoride concentration in drinking water to prevent fluorosis [31].

Their results were in line with our study. HQ index of this study indicated that health risks assessment in relation to fluoride concentration for children and adults are significant through drinking water consumption and there is a potential risk for dental and skeletal fluorosis. The HQ index for children and teenagers had health hazard (HQ > 1) in 2013–2017 of samples. Thus, in this rural area, there is a potential risk of fluorosis.

The HQ values of nitrate were analyzed in infant (0.005–0.4), children (0.035–2.69), teenagers (0.025– 2), and adults (0.02–1.6). The HQ index for children, teenagers, and adults had health hazard (HQ > 1) in 2013–2017 of the water samples. Drinking water resources with a risk of more than one were located in the villages in Saravan county. Therefore, in these rural areas, there are potential health risks. In the study by Arumi [49] in a rural area of Chile the mean HQ for adults in the study area was 0.12, indicating an absence of risk for this population group. For infants, HQ values had a maximum value of 3.1 in some locations, but the average was 0.69 (still below 1.0), indicating that the well water in the study area was generally not hazardous for infants [49]. In the study conducted by Sadler in drinking water from village wells in Semarang, Indonesia, result showed the HQ values as evaluated against WHO guideline values at 50%–95% points were HQ50 at 0.42 and HQ95 at 1.2, respectively. Based this research a low risk of infant methaemoglobinaemia for the whole population, but some risk for the sensitive portion of the population [32]. Similar study was carried out by Zhai et al. [50] in NE China, indicating that potential health risks of adult males and females within 60% of the area were at an acceptable level, while those within 40% were beyond the acceptable level. The NO_3^- concentration in southeast and northeast of the study area was the highest; hence, that residents in these regions were at the highest health risk [50].

4. Conclusion

Accordingly, the following conclusions can be drawn:

First, these findings can provide scientific evidence to policymakers, water resources management in the rural area of Saravan county in Sistan and Baluchistan province, Iran. Also, this is the first study to show spatial, temporal, and spatiotemporal variability of fluoride, and nitrate concentrations in this area.

Second, this paper quantitatively describes the hazardous degree of environmental pollution to human health that could help to solve core issues regarding the quality of groundwater in this area.

Third, based on the results, it is imperative to take measures in reducing fluoride and nitrate concentration level in drinking water and to control it. Actions should be implemented to enhance monitoring of fluoride and nitrate concentration in order to prevent the potential risk to people residing in these areas.

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Conflict of interest

The authors of this article declare that they have no conflict of interests.

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