Spatial distribution of fluoride and nitrate in groundwater and its associated human health risk assessment in residents living in Western Khorasan Razavi, Iran

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ABSTRACT

Fluoride and nitrate are the important factors that influence the drinking water quality. A health risk assessment was performed for exposure to fluoride and nitrate via drinking water ingestion pathway for the inhabitants living in Bardaskan County, Iran. In the present work, totally 30 drinking water samples were collected from private wells, monitoring wells, and boreholes during June 2018, from different previously unexplored rural and urban areas. The concentration of fluoride and nitrate varied from 0.55 to 1.75 mg/L (mean 0.873 mg/L) and from 5.7 to 25.4 mg/L (mean 12.58 mg/L), respectively. None of the 30 studied areas had fluoride, except one place, and nitrate concentrations above WHO guidelines. Hazard index (HI) values for adults, children and infants varied from 0.4160 to 1.1886 (mean 0.6405), from 1.0921 to 3.1203 (mean 1.6813) and from 1.165 to 3.3283 (mean 1.7934), respectively. HI estimated for groundwater in 3.3%, 100%, and 100% cases were found to be above the safety limit of 1 for adults, children, and infants, respectively. This research provides evidence that local residents in Bardaskan County may be at a high risk of health problems caused from fluoride and nitrate in drinking water. It is, therefore, important to take some remedial measures to prevent any health problem in this county.

Keywords: Fluoride; Nitrate; Human health; Risk assessment; Bardaskan

1. Introduction

Safe water is a key resource necessary for the life of all organisms on the globe [1,2]. It is argued that 80% of the diseases in the world come through the poor quality of drinking water [3]. Groundwater is the main source of

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drinking water among freshwater resources in many regions of the world [4,5] and Iran because it is available in all seasons and is less polluted than surface water and its need is increasing due to fast growth of population, urbanization, industrialization, which has exacerbated the water quality in the recent years [6]. Groundwater is the water beneath the earth's surface that moves freely through pores and cracks in rocks, sediments, and soils [7]. However, there are many

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concerns about the quality of drinking water due to both natural processes (physical and chemical weathering and erosion of rocks, ore, and volcanic deposits) and human activities (agriculture and industries) [8].

Fluoride and nitrate are the critical ions that influence the groundwater quality and have gained considerable attention in recent decades owing to their high concentrations currently detected in drinking water and as such their deleterious impacts on human health [9–12]. Due to natural processes as well as human activities, the concentrations of fluoride in water resources may exceed the recommended levels [13]. Fluoride in water naturally arises from weathering and dissociation of fluoride containing rocks and sediments including fluorite (CaF₂), fluorapatite (Ca₅(PO₄)₃F), and cryolite (Na_3AlF_6) [14]. Generally there exist about 85 million tons of fluoride deposits in the earth crust universally. Weathering of fluoride containing rocks and longterm rock-water reactions increase fluoride concentration in groundwater. Fluoride is a natural element present in varied concentrations globally in water bodies [15]. Fluoride contamination has been a universal environmental problem for years predominantly in mid-latitude regions of the world and has caused great concerns mainly due to its omnipresent nature and risk to human health [1,16,17]. 65% of endemic fluorosis in many regions in the world is due to the consumption of excessive fluoride content in drinking water [18,19]. Worldwide, natural fluoride level in groundwater is found as high as 30 mg/L in more than 30 countries including Mexico, India, China, India, Pakistan, Iran, West Africa, Thailand, China, Sri Lanka, Southern Africa, etc. [9].

Generally, fluoride has both beneficial and harmful impacts on human health [20,21]. Fluoride does not exhibit any colour, taste or smell when dissolved in water and hence, its concentration in range of 0.4-1.0 mg/L has a positive effect on human health, as it protects us from tooth cavity by helping calcification process of dental enamel [22]. However, fluoride level above 1.5 mg/L is detrimental and causes various health problems including dental and skeletal fluorosis, osteoporosis, arthritis, brittle bones, certain types of cancer, infertility, Alzheimer, brain damage, and thyroid disorder [23]. Based on World Health Organization (WHO), the optimum range of fluoride in drinking water is set to be 0.5-1.5 mg/L [24]. Some researchers noticed that the water having high pH values can increase the fluoride dissolution processes and ion-exchange between fluoride ion and hydroxyl. Elevated bicarbonate and sodium may also increase the fluoride level in groundwater [25,26].

Another chemical of concern in water is nitrate. Nitrogen is the main constituent of the air, comprising about 80% of the air we inhale. Many human activities can increase the nitrate concentrations to problematic concentrations. Nitrate can originate from farming activities by application of inorganic fertilizers, or manure from livestock, leaking from the sewage network and septic tanks. Nitrate is one of the important water contaminants in the globe and especially in rural areas [27,28]. High concentrations of nitrate have been determined in the groundwater of many parts of Iran [29–34]. However, based on the studies in many countries, nitrate concentrations in groundwater have been rising due to its stability, high solubility, and mobility and can cause thyroid gland dysfunction, gastric cancer, and methaemoglobinaemia or 'blue baby syndrome' in infants under 6 months of age [35-37]. In methaemoglobinaemia, the oxygen-carrying capability of haemoglobin is remarkably decreased due to its conversion to methaemoglobin. Nitrate itself is not carcinogenic but when transformed into nitrite in the gastrointestinal tract, it combines with secondary and tertiary amines to generate nitrosamines which have been considered as a potent human carcinogen [38]. Literature reviews have shown that infants are exposed to high concentrations of nitrate in drinking water when it is used to mix formula milk or other baby foods [38,39]. Elevated concentrations of nitrate in surface water bodies may lead to various ecological and environmental issues, including eutrophication and seasonal hypoxia [40]. WHO has established the maximum permissible limit of 50 mg/L as nitrate in drinking water to prevent methaemoglobinaemia in infants. Therefore, it is necessary to understand completely the contaminants and the associated health risks, in order to better evaluate the potential health risk of toxic chemicals to consumers.

Risk assessment proposed by U.S. Environmental Protection Agency (USEPA) has proven to be a helpful and frequently applied methodology for groundwater management, environmental planning, and decision making in many studies [41–43]. The aims of this research were to evaluate the fluoride and nitrate contamination in drinking water in Bardaskan County and to assess the associated health risk through consumption in adults, children, and infants using model proposed by the USEPA. The findings of the present work would be useful to health professional and decision makers for protecting the groundwater quality and government in working out strategies for clean drinking water supplies and risk management.

2. Materials and methods

2.1. Study area description

This study was performed in Bardaskan County. Bardaskan is located in Razavi Khorasan Province in East of Iran covering an area of 8,535 km². At the 2017 census, its population was 68,392. The county has three districts including Central District (Bardaskan), Anabad District, and Shahrabad District. This county has three cities including Bardaskan, Anabad, and Shahrabad. The population has considerably dispersed in villages where accessibility of infrastructure for the proper treatment and transportation of surface water are not always possible. The county is located in the north of the Great Salt Desert (Dasht-e Kavir). The climate of Bardaskan County falls under arid climatic zone with extreme temperatures (hot summer and dryness in the non-rainy seasons), relatively low precipitation and high evapotranspiration. The northern area of Bardaskan is cold in winter and in the south and central areas changes from semi dry to hot and dry with a rainfall of 150 mm per year. Dust storm events occur usually during summer months. Monthly rainfall reaches its maximum yearly amounts in the winter and spring, while the minimum yearly levels are reported in the summer and sometimes in autumn. The maximum and minimum temperature is approximately 45°C and 5°C, respectively. Almost all the cities and villages are in

north east, east, and southeast in this county. Furthermore, all the residents in the county rely on groundwater resources for human and livestock drinking as a relatively clean drinking water source.

2.2. Sampling and analysis

The cross-sectional research (i.e., one sample per location) was performed to measure fluoride and nitrate concentration in drinking water of Bardaskan County, Iran. There are comparatively no data associated with fluoride and nitrate concentrations in groundwater of Bardaskan, which is normally used for domestic and industrial purposes. Herein, totally 30 drinking water samples were collected from private wells, monitoring wells, and boreholes during June 2018, from previously unexplored rural and urban areas. The details of sampling locations are coded as S1 to S30. The sampling locations and their codes are given in Table 1. Location map of water sampling sites in Bardaskan County are shown in Fig. 1. The study area has both deep and shallow wells. The pumped groundwater was collected and stored in big concrete water reservoirs and then immediately transformed to the water distribution system. All the wells selected for sampling were routinely used for domestic and industrial needs. Samples were gathered by using 1,000 mL washed and dry screw-capped high-density polyethylene sampling bottles. All pumps/wells were flushed for at least 5 min to obtain fresh water prior to collecting groundwater samples. Note that the bottles had been rinsed three times at sampling site with well water before samples were collected. Samples were kept in ice box and immediately after collection were brought to the chemistry laboratory of Gonabad University of Medical Sciences for analyses of fluoride and nitrate. All of the samples were tested within 24 h of collection. Sampling, transportation, and conservation procedures followed the Standard Methods for the Examination of Water and Wastewater published by the American Public Health Association. Quantitative estimation of fluoride and nitrate was made by using a spectrophotometer UNICO-2100.

2.3. Non-carcinogenic health risk assessment model

Risk assessment is an important element that ensures the utilization of scientific knowledge to set standards, guidelines, and other recommendations associated with water safety to help better protection of the people health. Based on the United States Environmental Protection Agency (USEPA), a human health risk methodology is the method applied to assess the nature and likely of detrimental health effects in residents who may be exposed to elements or compounds in polluted environmental media now or in the future [44]. Groundwater health risk assessment is mainly focused on toxic materials, which can be classified into carcinogenic and non-carcinogenic contaminants based on their toxicological impacts on the humans. Ingestion was reported to be the most important route for exposure to many contaminants in water and the health risk through the dermal contact exposure pathway is usually very low [45-47]. Therefore, among three major pathways of exposure (ingestion, inhalation, and dermal contact), only ingestion route was considered in this study. The exposure magnitude was estimated based on the chronic daily intake (CDI), which was computed using the daily water intake rate and fluoride and nitrate concentrations in drinking water. The oral CDI for fluoride and nitrate detected in the various water samples was computed for each age category according to the following formula [48]:

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

where *C* is the concentration of fluoride and nitrate in the water in mg/L; DI is the daily water intake in L/d; *F* is the exposure frequency in days/year; ED is the exposure duration in years; BW is the body weight of studied age group in kg; and AT is the average timing in days.

Values and units of *C*, DI, *F*, ED, BW, and AT for adults, children, and infants are described in Table 1S.

The hazard quotient (HQ) was used to evaluate the probable chronic non-carcinogenic health hazard associated with

Table 1

Sampling sites, population, and concentrations of fluoride and nitrate in different rural and urban areas of Bardaskan County

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Code	Name	Population	Fluoride	Nitrate	Code	Name	Population	Fluoride	Nitrate
S1	Shamsabad	241	0.55	25.4	S16	Hatiteh	930	1.15	10.6
S2	Khanqah	259	0.68	18.8	S17	Islamabad	1,099	1.09	10.4
S3	Ahubam	122	0.82	18	S18	Seyfabad	884	0.74	8.4
S4	Kabudan	841	0.80	14.6	S19	Mohammadabad	885	0.81	11
S5	Hodk	499	0.81	16.2	S20	Khorramabad	676	0.86	7.9
S6	Sar Borj	122	0.84	16	S21	Kusheh	1,768	0.72	6.4
S7	Nezamabad	222	0.75	14.4	S22	Zaheerabad	1,606	0.62	7.6
S8	Khommi	241	0.85	17.9	S23	Quzhdabad	627	0.66	5.7
S9	Aliabad	140	0.75	6.2	S24	Roknabad	1,963	0.79	6.7
S10	Ebrahimabad	948	1.06	20.7	S25	Azimabad	1,075	0.92	6.9
S11	Anabad	1,480	1.28	21.2	S26	Firuzabad	661	0.83	8.7
S12	Mozaffarabad	1,437	1.75	19.9	S27	Shahrabad	2,185	0.67	7.9
S13	Marandiz	651	1.14	16.8	S28	Shafiabad	2,035	0.76	10.1
S14	Kalateh-ye Now	821	0.78	7.4	S29	Abnow	498	0.97	11
S15	Zirakabad	1,026	0.92	10.6	S30	Bardaskan	28,233	0.83	14

dietary fluoride and nitrate exposure. The HQ is expressed as the ratio of exposure magnitude to a single element in relation to an RfD or to the magnitude at which no deleterious impacts are expected [49,50].

The value of non-carcinogenic hazard quotient (HQ) of a contaminant can be computed by Eq. (2) [51]:

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

where RfD is the oral reference dose. The RfD values for fluoride and nitrate are set to be 0.06 and 1.6 mg/kg/d, respectively [52].

To estimate the overall non-carcinogenic impacts of exposure to multiple contaminants in water via different routes, the sum of the HQs of contaminants via all pathways is considered as the hazard index (HI). The equation to estimate this index is as follows [53,54]:

$$HI = \sum_{k=1}^{n} HQ = HQ_{\text{fluoride}} + HQ_{\text{nitrate}}$$
(3)

If the values of HQ or HI be ≥ 1 , it was considered as an unacceptable hazard of deleterious non-carcinogenic impacts on health, and if HQ or HI was found to be <1, then it was considered as within the acceptable level [55–57].



Fig. 1. Location map of water sampling in Bardaskan County, Iran.

3. Results and discussion

3.1. Occurrence of fluoride and nitrate in water samples

Water contamination is one of the greatest environmental issues and fluoride and nitrate are among the most common and prevalent contaminants in many groundwater resources. There has been a rising trend for clean water demand in the last 20 years due to the rapid growth of population [58,59]. Furthermore, communities face severe water shortages, because groundwater has been used faster than it is naturally recharged by rain or snow. Drinking water is the main contributor to fluoride intake, among all other sources in human body. Concentrations of fluoride and nitrate were studied in the water samples of Bardaskan County to evaluate the drinking suitability of the water. Fluoride in water is harmful in both high and low concentrations [60]. For example, children who consume water containing fluoride concentrations lower than 0.5 mg/L have a low occurrence of moderate or severe fluorosis [61]. Excessive nitrate levels found in drinking water can cause many health issues worldwide such as gastrointestinal cancers, methaemoglobinaemia, Alzheimer's disease, vascular dementia, and multiple sclerosis in human [62]. In this study, the concentration of fluoride and nitrate varied from 0.55 to 1.75 mg/L (mean 0.873 mg/L) and from 5.7 to 25.4 mg/L (mean 12.58 mg/L), respectively. The results of the analysis are given in Table 1. The lowest and highest fluoride content in water samples was observed in Shamshabad and Mozaffarabad, respectively. For nitrate, the lowest and highest content in the samples was found in Quzhdabad and Shamshabad, respectively (Table 1).

Fluoride concentration in 3% (1 out of 30) of groundwater samples collected from different rural and urban areas of Bardaskan County was above the WHO permissible limit (1.5 mg/L). The levels of fluoride and nitrate are depicted in Figs. 2 and 3, respectively. The red column in Fig. 2 means that the area had fluoride level above WHO guideline. All the sampled wells had nitrate concentrations within the suggested WHO guideline (50 mg/L) [63]. Therefore there is no concern regarding nitrate in the study area in 2018.

Based on the results, levels of fluoride and nitrate were detected in all the collected water samples (S1–S30) in this work. Though the concentrations of fluoride and nitrate are low, long-term exposure can induce various health risks.



Fig. 2. Fluoride concentrations (mg/L) in groundwater of Bardaskan County.



Fig. 3. Nitrate concentrations (mg/L) in groundwater of Bardaskan County.

3.2. Evaluation of human health risk due to fluoride and nitrate in groundwater samples

3.2.1. Chronic daily intake (CDI) calculation of fluoride and nitrate

The chronic daily intake (CDI) of two chemicals (fluoride and nitrate) was computed according to the mean concentration of each chemical in each water sample and the respective consumption rates. The CDI of fluoride and nitrate from consumption of water for three studied age groups are given in Table 2S. The CDI values of fluoride ranged from 0.0157 to 0.05 mg/kg d (mean 0.0249 mg/kg d) and from 0.0412 to 0.1312 mg/kg d (mean 0.0655 mg/kg d), from 0.044 to 0.14 mg/kg d (mean 0.0698 mg/kg d) for adults, children, and infants, respectively. For nitrate, the CDI values ranged from 0.1628 to 0.7257 mg/kg d (mean 0.3594 mg/kg d), from 0.456 to 2.032 mg/kg d (mean 1.0064 mg/kg d) for adults, children, and infants, respectively.

3.2.2. Non-cancer health risk assessment

Health risk assessment is helpful to understand the probability of harmful health impacts in residents who are exposed to chemicals in contaminated environment. It is often a key step in guaranteeing safety and health [59,64]. The human health risks from consumption of water containing fluoride and nitrate by adults, children, and infants populations were assessed based on HQ (the ratio of measured dose of a contaminant to a reference dose level) and HI. The HI value expresses the combined non-carcinogenic impacts of multiple chemicals. Health risk assessment model proposed by the USEPA was applied to assess the health risks that fluoride and nitrate could pose on human via direct ingestion of groundwater in Bardaskan. Although adults are also considered in the present work special focus is concentrated on the children and infants because these age groups are in a window of danger by many biological and social agents including growth stage, their central nervous, reproductive, and immune systems are rapidly developing; they could be exposed to higher levels due to their size and body weight. HQ values of fluoride and nitrate in drinking water of studied rural and urban areas are given in Table 3S. The HQ values of fluoride ranged from 0.2619 to 0.8333 (mean 0.4158) and from 0.6875 to 2.1875 (mean 1.0916), from 0.7333 to 2.3333 (mean 1.1644) for adults, children, and infants, respectively. For nitrate, the HQ values ranged from 0.1017 to 0.4535 (mean 0.2246) and from 0.2671 to 1.1906 (mean 0.5896), from 0.285 to 1.27 (mean 0.629) for adults, children, and infants, respectively. HQ values for groundwater consumption containing fluoride in 60% and 76% of studied locations were above the acceptable USEPA limit of 1 for children and infants, respectively. Whereas for nitrate, HQ values in 3.3% and 10% of studied locations were above the acceptable USEPA limit of 1 for children and infants, respectively.

HI values for adults, children, and infants varied from 0.4160 to 1.1886 (mean 0.6405), from 1.0921 to 3.1203 (mean 1.6813), and from 1.165 to 3.3283 (mean 1.7934), respectively. HI estimated for groundwater in 3.3%, 100%, and 100% cases were found to be above the safety limit of 1 for adults, children, and infants, respectively. This means there is a potential risk of occurrence of non-cancer health risk in infants, children, and adults via water consumption in most sites of study area. The HI values are depicted in Fig. 4.

The HQ levels were calculated for ingestion of water for adults, children, and infants along with HI, based on the fluoride and nitrate concentrations. HQ and HI evidenced that the consumption of the water may result in deleterious health risks to the residents. The concentration of fluoride above the guideline value water resources can undoubtedly harm plants, animals, and induce serious health problems in humans [65-67]. Many scientists studied human health risk estimation in different areas; for example, Ahada and Suthar [9] measured fluoride levels and associated health risks in groundwater of the southern districts of Punjab in India and found that children were more vulnerable to fluorosis. Munoth et al. [62] evaluated contamination status of fluoride and nitrate in groundwater of Rajasthan, India. They found fluoride concentrations up to 8.70 mg/L in Bharatpur district. A maximum level of nitrate was also observed in Chittaurgarh district as 1,392 mg/L. Thus, the water in villages of Rajasthan was not potable to drinking [62].



Fig. 4. Non-carcinogenic risks induced by fluoride and nitrate in drinking water.

In a study, nitrate and fluoride levels in groundwater of Davanagere Taluka in Karnataka were investigated. Totally 61 wells were sampled for the purpose of the study. The levels of fluoride and nitrate measured in different samples ranged from 0.19 to 2.06 mg/L and 0.08 to 308 mg/L, respectively [68].

Mirzabeygi et al. [69] studied the level of fluoride and health risk assessment in drinking water in 28 villages of the Ardakan city in Yazd, Iran. Based on their results, the range of fluoride was 0.9-6 mg/L (mean 2.92 mg/L), also in 50% of the villages, the fluoride level was above WHO guideline. They also reported that 46.6% of their samples had HQ value above the safety level of 1 [69]. Nikbakht et al. [70] assessed the health risk of fluoride and nitrate in more than 17 wells in two wet seasons (May) and the dry season (September) in Lar area, South Iran. The mean concentration of fluoride and nitrate in the wet season was 2.9 and 19.7 mg/L and the dry season 1.8 and 16.3 mg/L, respectively. The order of HQ values were as children > women > men [70]. Qasemi et al. estimated non-carcinogenic risk to human health due to intake of fluoride in the groundwater in 39 villages of Gonabad and Bajestan, Iran. A total of 55% and 4.7% of the studied villages in Gonabad and Bajestan, respectively, had fluoride concentrations below the lowest recommended value of WHO for fluoride (0.5 mg/L). Health risk index values for fluoride contamination for 44% and 90% of children and infants in villages of Gonabad and Bajestan, respectively, were more than unity (HQ \geq 1) [41]. Yousefi et al. assessed health risk to fluoride in 112 drinking water samples collected from 28 rural areas in Poldasht city, Northwest of Iran. The fluoride concentration in drinking water was in range 0.27-10.3 mg/L (mean 1.70 mg/L). Totally, 57% of samples exceeded the WHO limit. The computed HQs were above 1 for all groups of residents (infants, children, teenagers, and adults) in Agh otlogh and Sari soo areas [71].

Rezaei et al. [64] studied the health risk related to the fluoride and nitrate in drinking water of 106 rural and urban areas in the Sanandaj, Kurdistan County, Iran. The mean level of fluoride in urban and rural drinking water was 0.22 and 0.27 mg/L, respectively. The level of nitrate in urban and rural water samples ranged from 0.28 to 27.77 mg/L and from 1.28 to 80 mg/L, respectively. Their study also showed that all three groups studied (men, women, and children) were exposed to non-cancer health problems associated with nitrate (HQ \geq 1) [64].

Chen et al. [10] assessed the nitrate and fluoride contamination of drinking water and their related health risk to rural residents in a semiarid area of Northwest China. They reported a range from 2.66 to 103 and from 0.11 to 6.33 mg/L for nitrate and fluoride, respectively, in groundwater resources. Totally, 60% and 8% of the analyzed samples had nitrate and fluoride levels exceeding the recommended limits for drinking purpose set by the WHO (50 and 1.5 mg/L), respectively. The calculated hazard index showed that majority of the samples (72% and 60%) may pose deleterious impacts on infants and children, respectively [10]. In another study in China, Wu and Sun [72] worked on groundwater contamination and related health risk in an alluvial plain in Mid-west, China. They reported that nitrate concentration was high mainly because of industrial and agricultural activities. They also argued that children in the

area were at higher health risk than adults, and oral ingestion was the main exposure route of health risk [72]. Wongsanit et al. [73] surveyed nitrate contamination in groundwater and its potential human health in lower Mae Klong river basin in Thailand. They found the risks induced by groundwater nitrate were in range 0.04-4.58 and 0.02-2.29 for children and adults, respectively, in the study area [73]. The relative higher fluoride concentration in some areas in this study is due to the geology of the area not the industrial activities. Although levels of nitrate were lower than the standard, the health risk calculated was higher than the standards. Therefore proper management options should be considered to reduce even the existing levels of nitrate in drinking water resources and since nitrate mostly enter water due to improper management of sewage and application of fertilizers in farmlands, therefore, a special management of these options should be considered or alternative water resources with lower amount of nitrate or fluoride should be used.

4. Conclusions

The current research was performed to estimate fluoride and nitrate levels in drinking water and their associated human health risks in Bardaskan County for assessing the drinking suitability. Generally, the results indicated that levels of fluoride, except in one location, and nitrate were less than those recommended by the USEPA and WHO. This is an important finding as human health is directly affected by consumption of water. The estimated HQ and HI values for the children and infants exceeded the safe levels, meaning that the intake of fluoride and nitrate associated with the consumption of water is hazardous to residents. According to the results obtained in the present work, it is clear that the infants and children have a higher risk of presenting health impacts induced by the consumption of water containing fluoride and nitrate content compared with adults. The main contribution of this study is to provide a quick and cheap decision-making tool related to environmental health problems and to protect the health of the people that live in the areas with natural or anthropogenic contamination in the Bardaskan County. The current research could also help to provide existing baseline data regarding water quality of Bardaskan County, which will bring alertness to health professionals, inhabitants, and water supply organizations about its purity and quality importance.

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Supplementary Information

Table 1S

Parameters applied for health exposure assessment through drinking water

Risk exposure		Values for groups				
factors	Adults (age > 19)	Children (6 > age > 12)	Infants (age < 1)			
С				mg/L		
DI	2	1.5	0.8	L/d		
F	365	365	365	d/year		
ED	40	10	1	years		
BW	70	20	10	kg		
AT	14,600	3,650	365	d		

Table 2S Input parameters to characterize the CDI (mg/kg d) values for three age groups in studied locations

Code		Fluoride		Nitrate				
	CDI (Adults)	CDI (Children)	CDI (Infants)	CDI (Adults)	CDI (Children)	CDI (Infants)		
S1	0.0157	0.0412	0.044	0.7257	1.905	2.032		
S2	0.0194	0.051	0.0544	0.5371	1.41	1.504		
S3	0.0234	0.0615	0.0656	0.5142	1.35	1.44		
S4	0.0228	0.06	0.064	0.4171	1.095	1.168		
S5	0.0231	0.06075	0.0648	0.4628	1.215	1.296		
S6	0.0240	0.063	0.0672	0.4571	1.2	1.28		
S7	0.0214	0.0562	0.06	0.4114	1.08	1.152		
S8	0.0242	0.0637	0.068	0.5114	1.3425	1.432		
S9	0.0214	0.0562	0.06	0.1771	0.465	0.496		
S10	0.0302	0.0795	0.0848	0.5914	1.5525	1.656		
S11	0.0365	0.096	0.1024	0.6057	1.59	1.696		
S12	0.05	0.1312	0.14	0.5685	1.4925	1.592		
S13	0.0325	0.0855	0.0912	0.48	1.26	1.344		
S14	0.0222	0.0585	0.0624	0.2114	0.555	0.592		
S15	0.0262	0.069	0.0736	0.3028	0.795	0.848		
S16	0.0328	0.0862	0.092	0.3028	0.795	0.848		
S17	0.0311	0.0817	0.0872	0.2971	0.78	0.832		
S18	0.0211	0.0555	0.0592	0.24	0.63	0.672		
S19	0.0231	0.0607	0.0648	0.3142	0.825	0.88		
S20	0.0245	0.0645	0.0688	0.2257	0.5925	0.632		
S21	0.0205	0.054	0.0576	0.1828	0.48	0.512		
S22	0.0177	0.0465	0.0496	0.2171	0.57	0.608		
S23	0.0188	0.0495	0.0528	0.1628	0.4275	0.456		
S24	0.0225	0.0592	0.0632	0.1914	0.5025	0.536		
S25	0.0262	0.069	0.0736	0.1971	0.5175	0.552		
S26	0.0237	0.0622	0.0664	0.2485	0.6525	0.696		
S27	0.0191	0.0502	0.0536	0.2257	0.5925	0.632		
S28	0.0217	0.057	0.0608	0.2885	0.7575	0.808		
S29	0.0277	0.0727	0.0776	0.3142	0.825	0.88		
S30	0.0237	0.0622	0.0664	0.4	1.05	1.12		

Table 3S	
Risk assessment for Fluoride and Nitrate in drinking water	

Code	Fluoride			Nitrate			Hazard Index (HI)		
	HQ	HQ	HQ	HQ	HQ	HQ	HI	HI	HI
	(Adults)	(Children)	(Infants)	(Adults)	(Children)	(Infants)	(Adults)	(Children)	(Infants)
S1	0.2619	0.6875	0.7333	0.4535	1.1906	1.27	0.7154	1.8781	2.0033
S2	0.3238	0.85	0.9066	0.3357	0.8812	0.94	0.6595	1.7312	1.8466
S3	0.3904	1.025	1.0933	0.3214	0.8437	0.9	0.7119	1.8687	1.9933
S4	0.3809	1	1.0666	0.2607	0.6843	0.73	0.6416	1.6843	1.7966
S5	0.3857	1.0125	1.08	0.2892	0.7593	0.81	0.675	1.7718	1.89
S6	0.4	1.05	1.12	0.2857	0.75	0.8	0.6857	1.8	1.92
S7	0.3571	0.9375	1	0.2571	0.675	0.72	0.6142	1.6125	1.72
S8	0.4047	1.0625	1.1333	0.3196	0.8390	0.895	0.7244	1.9015	2.0283
S9	0.3571	0.9375	1	0.1107	0.2906	0.31	0.4678	1.2281	1.31
S10	0.5047	1.325	1.4133	0.3696	0.9703	1.035	0.8744	2.2953	2.4483
S11	0.6095	1.6	1.7066	0.3785	0.9937	1.06	0.9880	2.5937	2.7666
S12	0.8333	2.1875	2.3333	0.3553	0.9328	0.995	1.1886	3.1203	3.3283
S13	0.5428	1.425	1.52	0.3	0.7875	0.84	0.8428	2.2125	2.36
S14	0.3714	0.975	1.04	0.1321	0.3468	0.37	0.5035	1.3218	1.41
S15	0.4380	1.15	1.2266	0.1892	0.4968	0.53	0.6273	1.6468	1.7566
S16	0.5476	1.4375	1.5333	0.1892	0.4968	0.53	0.7369	1.9343	2.0633
S17	0.5190	1.3625	1.4533	0.1857	0.4875	0.52	0.7047	1.85	1.9733
S18	0.3523	0.925	0.9866	0.15	0.3937	0.42	0.5023	1.3187	1.4066
S19	0.3857	1.0125	1.08	0.1964	0.5156	0.55	0.5821	1.5281	1.63
S20	0.4095	1.075	1.1466	0.1410	0.3703	0.395	0.5505	1.4453	1.5416
S21	0.3428	0.9	0.96	0.1142	0.3	0.32	0.4571	1.2	1.28
S22	0.2952	0.775	0.826	0.1357	0.3562	0.38	0.4309	1.1312	1.2066
S23	0.3142	0.825	0.88	0.1017	0.2671	0.285	0.4160	1.0921	1.165
S24	0.3761	0.9875	1.0533	0.1196	0.3140	0.335	0.4958	1.3015	1.3883
S25	0.4380	1.15	1.2266	0.1232	0.3234	0.345	0.5613	1.4734	1.5716
S26	0.3952	1.0375	1.1066	0.1553	0.4078	0.435	0.5505	1.4453	1.5416
S27	0.3190	0.8375	0.8933	0.1410	0.3703	0.395	0.4601	1.2078	1.2883
S28	0.3619	0.95	1.0133	0.1803	0.4734	0.505	0.5422	1.4234	1.5183
S29	0.4619	1.2125	1.2933	0.1964	0.5156	0.55	0.6583	1.7281	1.8433
S30	0.3952	1.0375	1.1066	0.25	0.6562	0.7	0.6452	1.6937	1.8066