ORIGINAL PAPER



Fluoride in Drinking Water in 31 Provinces of Iran

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Received: 16 January 2016/Revised: 24 March 2016/Accepted: 25 March 2016/Published online: 16 April 2016 © Springer Science+Business Media Dordrecht 2016

Abstract The effects of acute fluoride toxicity have been well documented in the literature. Drinking water is an important source of fluoride intake by humans, hence studies need to be carried out to determine the concentration of Fluoride in water. Therefore, this study tends to demonstrate the fluoride concentration in drinking water in thirty-one provinces of Iran during 2014. This cross-sectional study on drinking water was conducted in 2014. SPADNS method was determined for fluoride concentration examination according to instruction of Standard Methods. The minimum concentration of fluoride in provinces, such as Fars, Kermanshah, Kohgiluyeh and Boyer-Ahmad, Markazi, and Hormozgan, was observed to be 0.01 mg/L, while the maximum concentrations were observed to be 3.72 and 3.52 mg/L for Bushehr and Fars, respectively. The minimum and maximum average mean concentrations were 0.193 (SD = 0.11) and 0.889(SD = 0.31) for Kermanshah and Bushehr, respectively. Due to the disadvantages of fluoride and because of the

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existence of different ecological conditions in Iran, there are different concentrations of water fluoride in the country. Therefore, proper policies should be made for water treatment plants based on the regional conditions in order to achieve a desirable fluoride concentration standard.

Keywords Concentration · Drinking waters · Fluoride · Iran

Introduction

Iran is located between $25^{\circ}30'$ and $39^{\circ}47'$ of the northern latitude and 44°14' and 63°20' of the eastern longitude. It has an area of 1,648,195 km², making it the 18th-largest in the world and the second-largest nation in the Middle East. With 78.4 million inhabitants, Iran is the world's 17th most populous nation (World Bank). Fluoride enters the drinking water from atmospheric sources, precipitation (suspended particulates, emissions of volcanic gases, and rainfall contaminated with industrial wastes), human sources (inducing industrial aerosols), minerals (rocks often contain 100-1300 mg/kg of fluorine), and geothermal sources (hot springs) (David and Ozsvath 2009). The entrance of fluoride into the water environment mainly occurred through natural resources (Mesdaghinia et al. 2010). The Earth crust contains fluoride with an average concentration of 0.3 g/kg and the concentration of fluoride in air across the globe is about 3 ng/m^3 (Mesdaghinia et al. 2010). Fluorine has a higher electron affinity than other elements, existing as a negatively charged ion in water. It can be absorbed into the body as soon as it gains access into the human digestive system, thus forming HFo complex with the stomach acid. HFo complex is easily absorbed through the stomach and small intestine by a simple diffusion process and then changes as a free

The annual average of daily maximum temperature (°C)	Minimum average quantity of fluorine (mg/L)	Desirable quantity of fluorine (mg/L)	The maximum amount of fluorine (mg/L)
10–12	1.1	1.2	2.4
12–14.6	1.0	1.1	2.2
14.6–17.7	0.9	1.0	2.0
17.7–21.5	0.8	0.9	1.8
21.5–26.5	0.7	0.8	1.6
26.5–32.5	0.6	0.7	1.4

Table 1 Fluoride variation with temperature changes according to Iran Industrial Standards Institute (Mesdaghinia et al. 2010; Ebrahimi 2015)

fluoride toxicity are well documented in the literature (Mannina et al. 2013). Some fluoride-related diseases include genetic mutations, birth defects, infants' height and weight disorders (Aghaei et al. 2015), increased allergic reactions, allergic diseases, damage to bones, abnormal skeletal effects, Fluorosis (Azami-Aghdash et al. 2013), blood pressure (Amini et al. 2011), fertility effects, effects on kidney and liver (Xiong et al. 2007), neurological effects, impaired intelligence of children (Seraj et al. 2012; Karimzade et al. 2014; Anna and Guifan 2012; Nagarajappa and Pujara 2013), hyperactivity (Ashley and Till 2015), thyroid hormone effects (Sachdeva et al. 2015; Momeni and Dehghani 2011; Swati and Shashi 2013; Coperchini and Pignatti 2015; Vandana et al. 2015; Susheela 2005; Wolka et al. 2014), carotid atherosclerosis (Liu et al. 2014), cancerous effects, and Alzheimer's disease; in spite of these, more studies are needed in this area (David and Ozsvath 2009; Mannina et al. 2013). Linear regression analyses by Dobaradaran et al. (2008a, b) showed weak increases of the number of decayed permanent teeth and the number of decayed deciduous teeth scores with increasing water F levels (Dobaradaran et al. 2008a, b). The mean fluoride content in the air of Arak city was investigated by Dobaradaran et al. (2008a, b) and it was found to be 73.43 μ g/m³ (Dobaradaran et al. 2009a). The range of fluoride content of 0.53 ± 0.07 tea liquor was reported from to 2.60 ± 0.18 mg/L by Mahvi et al. (2006). The mean fluoride content of bottled drinking water was measured by Dobaradaran et al. (2008a, b) and it was 0.3 mg/L with a range of 0.00–0.59 mg/L in Iran (Dobaradaran et al. 2008b). Fluoride concentrations in the Shush aquifer of Khuzestan had ranged from 0.12 to 2.17 mg/L (mean 0.6 \pm 0.44 mg/ L) (Nouri et al. 2006). Correlation of fluoride with noncarbonated hardness, SO4 2-, Ca2+, and Mg2+ was found to be positive in groundwater of Dashtestan (Dobaradaran et al. 2009b). Fluoride adsorption in aqueous solution by a hybrid resin was studied by Boldaji et al. (2009), where this adsorbent had 61 percent efficiency for water with fluoride less than 4 mg/L (Boldaji et al. 2009). Iran's drinking water standard is based on the guidelines recommended by the

fluoride (David and Ozsvath 2009). The effects of acute

World Health Organization (WHO) on drinking water that contains less than 1.5 mg/L of fluoride. This guideline is based on a water consumption of 21 per day for adults. The Iranian standard for fluoride concentration in drinking water is shown in Table 1. Many studies measured fluoride in drinking water and water wells in different parts of Iran including the daily fluoride intake for residents of Bushehr Province which was estimated by Battaleb-Looie et al. (2013) in this location, of which 44.4 % of the drinking water exceed the guideline value of 1.5 mg/L recommended by WHO (Battaleb-Looie et al. 2013). Fluoride in the main drinking water source was assessed by Derakhshani et al. (2014) in the Zarand in Kerman Province of Iran. The maximum groundwater fluoride concentration was 3.51 mg/L (Derakhshani et al. 2014). Fluoride in drinking water of Muteh was obtained (1.8–2.2 ppm) by Keshavarzi et al. (2010). Fluoride in drinking water of kuhbanan was assessed by Poureslami et al. (2011) and the average content was 2.38 ppm (Poureslami et al. 2011). Fluoride level in Maku was reported to be more than 1.5 mg/L by Ejlali et al. (2015). The maximum of F content in Zarrin Dasht was reported as 2.45 mg/L by Momenifar and Moosavinasab (2013). This study demonstrates the fluoride concentration in drinking water for community water supply in urban areas of 31 provinces of Iran during 2014.

Materials and Methods

Iran, a Middle Eastern country south of the Caspian Sea and north of the Persian Gulf, shares borders with Iraq, Turkey, Azerbaijan, Turkmenistan, Armenia, Afghanistan, and Pakistan. This cross-sectional study was conducted in 2014 on drinking water, and information related to fluoride concentrations in drinking water of Iran's provinces was extracted from the Iranian Wastewater Engineering Company in 2014. Some county with the maximum concentration of fluoride in Drinking Water has been re-sampling and measurement by university that Shown in Table 2. This study was conducted in provinces of Iran by the Iranian

Table 2 The fluoride content in drinking water in the provinces of Iran and location with the maximum fluoride content

No	Province/county	Min of conc. (mg/L)	Max of conc. (mg/L)	Mean of conc.(mg/L)	Standard deviation SD (mg/L)	Latitude	Longitude
1	Bushehr/Bushkan	0.05	3.52	0.889	0.31	28.923384	50.820314
		_	3.52	-	_	28.827959	51.700211
2	Khorasan-north	0.04	1.52	0.783	0.29	37.471035	57.101319
3	Semnan	0.13	1.49	0.742	0.32	35.578797	53.379811
4	Qazvin	0.18	1.8	0.662	0.45	36.273659	49.998236
5	Qom	0.1	1.5	0.648	0.32	34.639944	50.875942
6	Fars/Zarrin Dash	0.01	3.72	0.617	0.57	29.104381	53.045893
		_	3.72	-	-	31.513357	52.124896
7	Razavi Khorasan-M/ khvaf	0.03	1.5	0.59	0.23	35.102025	59.104176
		_	1.5	_	-	34.571826	60.147409
8	Chaharmahal and Bakhtiari	0.033	1.23	0.532	0.22	29.610683	50.755955
9	Hormozgan/Bandar Lengeh	0.01	3	0.53	0.49	27.138723	55.137583
		_	3	_	_	26.550941	54.886188
10	Khuzestan/Shush	< 0.02	2.38	0.504	0.25	31.436015	49.041312
		_	2.38	_	-	32.195795	48.254283
11	Yazd/Behabad	< 0.02	1.39	0.5	0.27	31.897423	54.356856
		_	1.39	_	_	31.871595	56.019424
12	Hamedan	0.06	1.23	0.472	0.26	34.798857	48.515022
13	Kerman/kuhbanan	0.02	1.74	0.471	0.25	30.283938	57.083363
		_	1.74	_	_	31.412630	56.280060
14	Ilam	0.16	1.18	0.46	0.23	33.634974	46.415281
15	Lorestan	< 0.02	1.2	0.446	0.27	33.581839	48.398819
16	Sistan Baluchestan	< 0.02	1.15	0.439	0.25	27.529991	60.582068
17	Zanjan	0.14	1.56	0.42	0.19	36.683005	48.508721
18	Khorasan -south	0.1	1.1	0.419	0.21	32.517564	59.104176
19	Tehran	0.02	1.58	0.399	0.2	35.689197	51.388974
20	Alborz	0.029	1.548	0.352	0.23	35.996047	50.928925
21	East Azerbaijan	0.19	0.847	0.343	0.12	37.903573	46.268211
22	Mazandaran	< 0.02	1.16	0.335	0.21	36.226239	52.531860
23	Ardebil	0.12	0.58	0.328	0.11	38.253736	48.299990
24	Markazi	0.01	1.52	0.31	0.4	34.612305	49.854727
25	Isfahan	< 0.02	1.5	0.292	0.23	32.654627	51.667283
26	Kurdistan	< 0.02	0.8	0.282	0.16	35.955358	47.136213
27	West Azerbaijan/Maku	< 0.02	2.2	0.281	0.29	37.455006	45.000000
						39.295621	44.514185
28	Kohgiluyeh and Boyer- Ahmad	0.01	0.81	0.264	0.17	30.724586	50.845632
29	Golestan	0.09	0.72	0.264	0.11	37.289812	55.137583
30	Gilan	<0.02	0.82	0.219	0.13	37.280946	49.592413
31	Kermanshah	0.01	0.86	0.193	0.11	34.327692	47.077769
	rennunshull	0.01	0.00	0.175	0.11	54.521072	+1.011109

Water and Wastewater Engineering Company in cooperation with 41 regional water and wastewater companies located in 31 provinces, all across the country. Because this study was done extensively in all 31 provinces of Iran, we should use the standard method for measuring drinking water fluoride content that was present in all 31 provinces; therefore, SPADNS method was used for fluoride concentration examination according to the instructions of standard methods (Lenore et al. 2005). The SPADNS colorimetric method is based on the reaction between

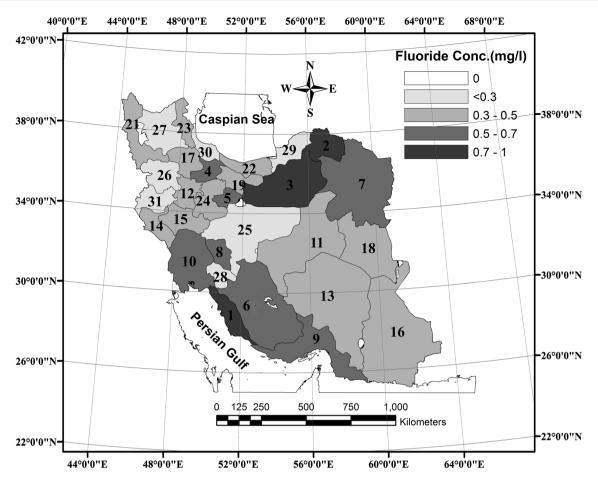


Fig. 1 Provincial mean of fluoride content in the drinking water of Iran

fluoride and a zirconium-dye lake (Merck, Germany). Fluoride reacts with the dye lake, dissociating a portion of it into a colorless complex anion (ZrF_6^{2-}) . As the amount of fluoride increases, the color produced becomes progressively lighter (Lenore et al. 2005). Finally, the resulting solution color is measured in a spectrophotometer (DR 2000 Spectrophotometer, HACH Company, USA) at 570 nm. A calibration curve was prepared using concentrations ranging from 0 to 0.4 mg F⁻/L (Battaleb-Looie et al. 2013; Lenore et al. 2005). All reagents were purchased from Merck. The minimum detectable level of the SPADNS method is 0.02 mg/L, as reviewed, analyzed, and compared by the World Health Organization and Iran national standards. The data obtained were evaluated using SPSS19, GIS 2010, and EXCEL 2007, and the resulted graphs and tables were extracted.

Results

The data for mean, minimum, and maximum concentrations of fluoride in drinking waters for each province across the country in 2014 were measured in milligrams per liter as shown in Table 2. As shown in Table 2, the minimum concentration of fluoride in provinces, such as Fars, Kermanshah, Kohgiluyeh and Boyer-Ahmad, Markazi, and Hormozgan, was observed to be 0.01 mg/L.

However, the maximum concentrations of fluoride were observed as 3.72 and 3.52 mg/L for Bushehr and Fars, respectively, while the minimum and maximum average mean concentrations were 0.193(SD = 0.11) and 0.889 (SD = 0.31) standard deviation for Kermanshah and Bushehr, respectively.

Discussion

In this study, the fluoride concentrations in drinking water of Iran's provinces were studied. As shown in Table 2, Figs. 1 and 2, Bushehr, north of khorasan and Semnan, had a higher mean concentration of fluoride than other provinces, but all provinces had an average concentration of fluoride lower than the WHO guideline value of 1.5 mg/L. The high-fluoride concentration of drinking water in some districts of Bushehr province was higher than the WHO guideline value, which was also reported by Mesdaghinia



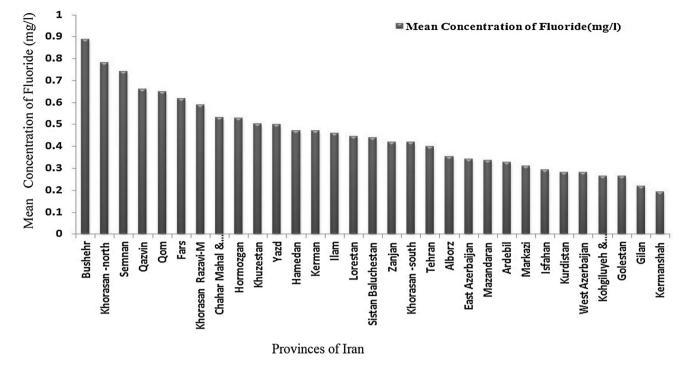


Fig. 2 Mean concentration of fluoride in drinking water in Iran

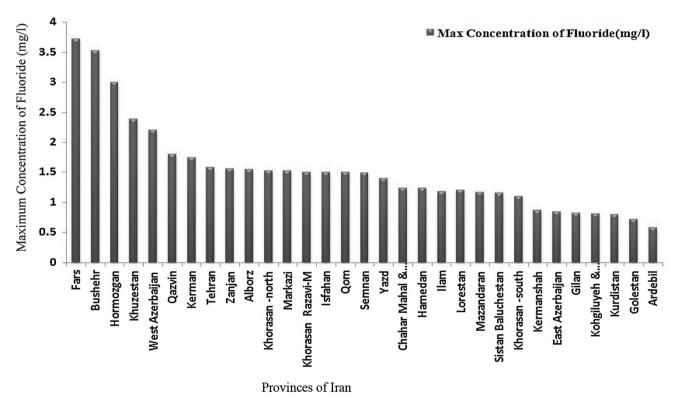
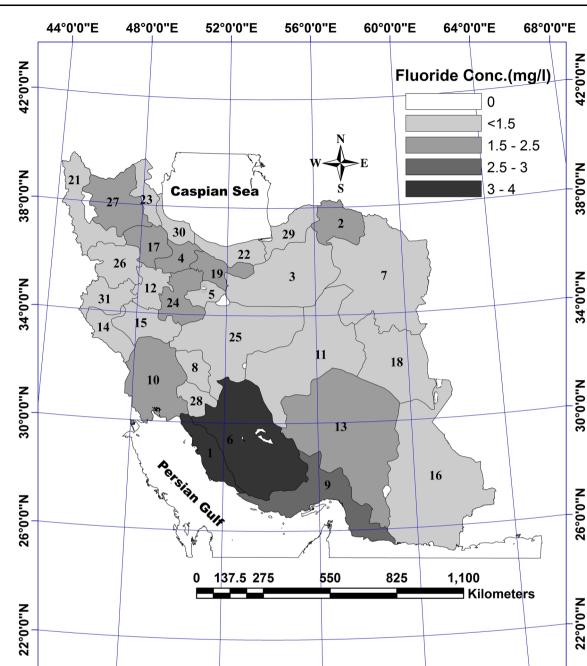


Fig. 3 Maximum concentration of fluoride in drinking water in Iran

et al. (2010). Some districts of Fars also had higher fluoride concentration in drinking water than the WHO guideline value, but was not reported in the study of Mesdaghinia et al. (2010).

According to Figs. 3 and 4, the sensitive regions in which the fluoride concentration in drinking water was higher than the WHO guideline value are located in Fars > Bushehr > Hormozgan > Khuzestan > East Azer-



56°0'0"E

Fig. 4 Provincial maximum fluoride content in drinking water of Iran

52°0'0"E

48°0'0"E

baijan > Qazvin > Kerman > Tehran > Zanjan > Alborz > Khorasan -north > Markazi, in the order of the highest to lowest fluoride concentration. In these provinces, the maximum values of fluoride were 3.72, 3.52, 3, 2.38, 2.2, 1.8, 1.74, 1.58, 1.56, 1.55, 1.52, and 1.52 mg/L, respectively. Therefore, further studies could be conducted in areas associated with the prevalence of common diseases and more attention is needed in the case of treatment of drinking water. In this study, fluoride concentration was

observed to be in the range of 0.05-3.52 with a mean of 0.889 mg/L in Bushehr Province, which was higher than that reported in the study of Dobaradaran et al. (2008a, b) in which fluoride concentration in the range of 0.99–2.5 was obtained with a mean of 0.270 mg/L (Dobaradaran et al. 2008b). The aforementioned results were lower than that reported by Mesdaghinia et al. (2010) which investigated the fluoride concentration in groundwater resources, and a concentration in the range of 1.11–4.06 was obtained

60°0'0"E

64°0'0"E

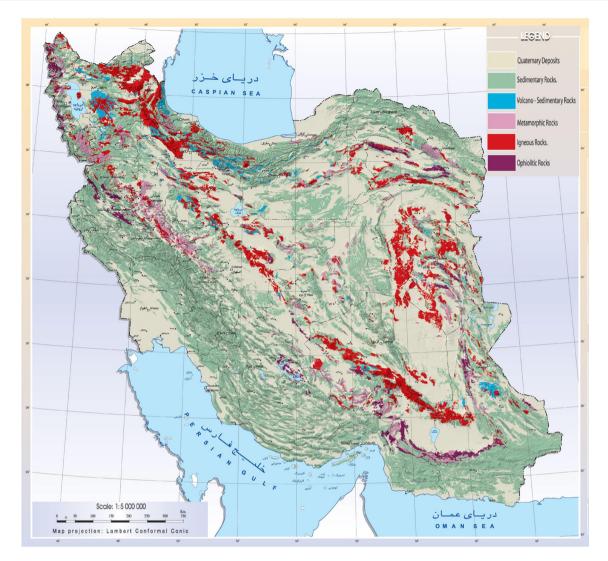


Fig. 5 Simplified Geological Map of Iran Based on Rock type (http://www.gsi.ir/) (Geological Survey of Iran)

with a mean value of 1.86 mg/L. Thus, this could be attributed to the portion of groundwater in water supply (Mesdaghinia et al. 2010). The range of F concentration in Fars Province was (0.01–3.72) with a mean value of 0.617 mg/L and this could be attributed to its ecological conditions which are more than that reported in the study of Mesdaghinia et al. (2010).

The high concentration of fluoride in Fars Province shows that the use of underground water resources is on the increase. Recently, sinkhole has been created with a depth of 100 m in Estahban county of Fars Province. Figure 5 represents the distribution map of three major groups of rocks such as sedimentary, igneous, and metamorphic rocks that constituent the continental crust. In addition to these three major groups of lithologies, rocks comprised sedimentary and volcanic units and quaternary unconsolidated deposits. It is considered that the sedimentary rocks have a wider distribution relative to other groups. Because water resources for drinking water in rural areas of Maku such as Takhteh Duz, Kharmanyeri, Rend, and Ali Qandu are from basaltic aquifers, which are the main source of fluoride, it increases fluoride content in ground water in these regions. Last Ararat volcano flows just to Takhteh Duz and Danalu village (North Maku) in this place, then continues along the river until coverage with sedimentary rocks. Izadkhvast is located in Zarrin dash County of Fars Province, because of gypsum and salt tissue of their soils and correlation between fluoride and calcium, sodium, sulfate ions. It seems that weathering and dissolution of sulfate and evaporated minerals are the reason for increasing fluoride in ground water in these regions. Bushkan is a village in Dashtestan County, Bushehr Province, Iran. Bandar Lengeh is a harbor city in, and capital of Bandar Lengeh County, in Hormozgan Province of Iran

on the coast of the Persian Gulf. Their rocks composes of sedimentary and quaternary unconsolidated deposits, and as mentioned, it can be the cause high-fluoride content of drinking water in these areas (Derakhshani et al. 2014; Keshavarzi et al. 2010; Poureslami et al. 2011; Ejlali et al. 2015; Momenifar and moosavinasab 2013).

Conclusion

The results of this research showed that although the average fluoride concentration in drinking waters of Iran's provinces is less than the WHO guideline value, the fluoride concentration in some other regions is still higher than the WHO guideline value. The relationship between fluoride and human health was first discovered in the late nineteenth century, when several chemists recognized the contents of fluoride in bones and teeth of humans (David and Ozsvath 2009). Despite some of the benefits of fluoride in the body, the effects of acute fluoride toxicity have been well documented in several literatures. Some researchers, such as Anna Augustsson from Sweden, have studied the risk assessment of fluoride intake from various sources, including food and beverage (tea, water, and other drinks), toothpaste, soil particles, and certain medications in 2014 and found that fluoride intake through drinking water is 77 % in comparison with other factors (Augustsson and Berger 2014). The minimum risk level of 0.05 mg/kg/d calculated by Agency for Toxic Substances and Disease Registry, and Battaleb-Looie et al. (2013) estimated the maximum fluoride intakes (from dates and drinking water) for children and adults were 3.4 and 1.6 times higher than the minimum risk level, respectively (Battaleb-Looie et al. 2013). Due to the disadvantages of fluoride and as a result of the existence of different ecological conditions in Iran, there are different concentrations of fluoride in drinking water in this country. Therefore, proper policies should be made for water treatment plants based on regional conditions, in order to achieve the desirable fluoride concentration standard. Fluoride standards in drinking water should be based on human health concerns, socio-economic conditions, culture, and environment of that region. As a result, further studies should be carried out in Iran to assess the prevalence of some diseases in areas with fluoride concentrations higher than the standard. There are many methods to remove extra fluoride from drinking water that have been reported by researchers. These include application of home-based water purifier on fluoride concentration of drinking water by Jaafari-Ashkavandi and Kheirmand (2013) which gained the removal efficiency of 85.6 % (Jaafari-Ashkavandi and Kheirmand 2013). To reduce the amount of fluoride to acceptable drinking water standard, Rahmani Boldaji et al. (2009) evaluated the effectiveness of a hybrid sorbent resin. They found that adsorbent with 61 % efficiency is suitable for the regions where F concentration is less than 4 mg/L (Boldaji et al. 2009).

Fresh and modified manganese oxide bone char can be useful for the removal of fluoride from water (Yazdanbakhsh et al. 2015). Other study for remove extra fluoride from drinking water are such as: Absorption in the bauxite containing Titanium (Das et al. 2005), reverse osmosis (Gopal et al. 2004), bone absorption, coagulation with alum, absorbed in the activated Alumina (Kagne et al. 2008; Srimurali et al. 1999; Tripathy and Raichur 2008), absorbed in granular ferric iron (Kumar et al. 2009), activated carbon absorption in conventional and modified (Daifullah et al. 2007) and absorbed in Kaolinite (Meenakshi et al. 2008), soften with lime in the presence of high levels of magnesium, by the use of Tricalcium phosphate granules (Maliyekkal et al. 2006; Sarkar et al. 2006). These researches show that most of these methods cannot be used in cross-country condition and faced with limitations of technical and economical conditions (Ghanizadedh 2002). Due to the most of places with high concentrations of fluoride in drinking water in Iran are in areas with low population like Behabad in Yazd Province, Kuhbanan in Kerman Province, Bandar Lengeh in Hormozgan Province, Maku in West Azerbaijan province, Zarrin Dasht in Fars Province, Shush in Khuzestan, Khvaf in Razavi Khorasan, and most of them use supply wells as drinking water, it is better to apply home-based water purifier or reverse osmosis in these areas. The government should provide the cost of buying those devices for the residents of these areas by bank loans.

Acknowledgments The authors are grateful to the Iranian Wastewater Engineering Company for their cooperation.

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