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ORIGINAL ARTICLE

Evaluation of fluoride concentration at inlet and outlet household water treatment systems and bottled water distributive high consumption Ardabil city, Iran

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KEYWORDS

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ABSTRACT: Fluoride is needed for better health, but it can cause adverse health effects if used at higher levels. There are different sources for the uptake of the fluoride, and drinking water is its primary resource. The aim of this study is the evaluation of fluoride concentration at the inlet and outlet of household water treatment systems and highly consumed bottled water distributed in Ardabil city in 2020. This descriptive cross-sectional study was performed on 60 water samples (30 samples of bottled water from 10 distribution brands of Ardabil city and 30 samples of inlet and outlet of household water treatment system). The concentration of fluoride was measured using the SPADNS standard method and spectrophotometer techniques. The SPSS version 22 software was used for analyzing the data. The concentration of fluoride in all samples was obtained to be in the range of 0 to 0.87 $mg.l^{-1}$, with an average of 0.35 $mg.l^{-1}$, which was less than the standard. According to the results, the concentration of fluoride in the studied groups was significantly different (P = 0.001). Moreover, the efficiency of the household water treatment system in the reduction of fluoride was observed to be 67.25%. Although the concentration of fluoride in most samples was lower than standard, further studies on other sources of fluoride, such as vegetables, tea, and so on, are required for accurate comment on fluoride deficiency in drinking water of one area.

INTRODUCTION

One of the indicators of countries' development is the public access to safe drinking water. Drinking water quality contains different aspects, e.g., physical, chemical, microbial, and aesthetic properties. Cations, anions, hardness, and alkalinity are among the chemical properties of water [1]. The important issue in the quality of drinking water is this fact that although some of the features, such as hardness, are vital in terms of consumer

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satisfaction, the presence of some chemicals in drinking water, such as fluoride, at concentrations higher than standard values can threaten the long-term health of consumers [2]. Fluorine is one of the halogen elements, which is naturally found in rock, coal, clay and soil. Although the daily amount of fluoride consumed is dependent on the geographical area and the diet, but the largest source of fluorine needed for the body is the drinking water [3,4]. Fluoride intake can be also increased by air pollution and use of fluoride-containing toothpaste. The level of fluoride intake by drinking water is dependent on the natural conditions of the water. The concentration of fluorine in surface water has been obtained to be less than 1.5 $mg.l^{-1}$, however the concentration of this element in groundwater is higher and may reach several milligrams per liter because of passing through rich areas of fluoride. The soluble fluorine is absorbed after the digestive system enters the body [5]. Water pollution by fluoride generally happens through two pathways, i.e., natural source and human activities. The structure of many minerals contains fluoride and, due to minerals erosion by rainwater, is usually leached out, which consequently contaminates both ground and surface waters [6,7]. In addition, in the case of contamination caused by anthropogenic activities, fluoride was released to drinking water by discharging the wastewater of various industries such as aluminum and steel production, metalworking and electroplating, and glass and semiconductor manufacturing. It could be also occurred by ore beneficiation and fertilization operation [8,9]. It should be noted that even fluoride compounds are widely used as precursors of fluoride release in some industries. For example, some of fluoride compounds employed in high-purity graphite production and nuclear industry have high values of fluoride [10]. Based on reports, more than 200 million people in the world have received fluoride in concentrations higher than the values recommended by WHO (<1.5 $mg.l^{-1}$) [11]. The investigation of health risks due to exposure to high levels of fluoride has been carried out in a variety of studies [12,13].

For the first time, Dean et al. identified that the presence of adequate fluoride in drinking water leads to a decrease in the possibility of decaying the teeth [14]. Fluoride, along with its beneficial effects, in addition to dental and bone diseases, may also lead to decreasing intelligence, altering DNA structure, increasing kidney damage, decreasing thyroid function and osteoporosis, and impairment of the nervous system and muscles, and possibly bladder and lung cancer can also occur, if its high concentrations consumed [15].

Since there is great potential for contamination of urban water resources, the consumption of bottled water, and the application of household water treatment systems are a popular alternative. Having information on the quality of drinking water is valuable to prevent the harmful effects of contaminated water. Due to the difference in fluoride content in different waters, its health importance to human health, the high rate of tooth decay in Ardabil city, which shortage in fluoride is one of its reasons and lack of comprehensive study in this area, this study was conducted to evaluate the amount of fluoride in the inlet and outlet of the household water treatment system and distributed bottled water in Ardabil city in 2020.

MATERIALS AND METHODS

This was a descriptive-cross-sectional study. The total population of the study included 60 drinking water samples consisted of 30 samples from 10 mostly consumed brands distributed (Vata, Atash, Aquaina, Pana, Didi, Parmin, Piorlife, Damavand, Oxab and Dasani) in Ardabil city (3 samples were selected from each brand). Out of these 10 brands, 5 samples were bottled mineral water and 5 samples were of bottled drinking water) and 30 samples of water inlet and outlet of household water treatment systems, which were randomly collected and tested. For sampling water of household water treatment systems, 6 samples were taken from each of the 5 districts of Ardabil city (including 3 samples from inlet and 3 samples from outlet). Samples of bottled water were also randomly selected and purchased from each of the 5 districts of Ardebil, 6 stores and one package from each store and one sample from each package. The samples were transferred to the laboratory and tested.

The household water treatment systems used in the homes were three-step to six-step reverse osmosis, which included 5-micron polypropylene fiber pre-filters, activated carbon, 1-micron microfilter, reverse osmosis, final activated carbon, and in some cases, ion exchange resins and mineral filters. Samples were taken from the nearest water tap to the household water treatment systems (faucet of dishwashing sink) and the outlet faucet.

Fluoride measurement was performed using SPADNS standard method according to standard method for examination of water and wastewater. In this method, 50 ml of the water sample was prepared and 10 ml of spadens and zirconium acid mixture were added and the color intensities were measured by spectrophotometer at 570 nm. The unknown concentrations were determined by plotting the calibration curve by preparing standard solutions in the range of 0 to 1.4 $mg.l^{-1}$. In this study, composite sampling method was used.

Data were analyzed by Excel and SPSS version 22 using one way ANOVA and T-test.

RESULTS AND DISCUSSION

The results of the study related to mean fluoride ion concentration in 30 samples of distributed bottled water in Ardabil city are presented in Table 1. According to Table 1, Atash mineral water had the highest mean fluoride concentration (0.53 $mg.l^{-1}$1) and mineral water with the Watta and Parmin brands had the lowest mean fluoride concentration (0.29 $mg.l^{-1}$). Among the bottled drinking water, the highest and lowest mean concentrations of fluoride were in the Damavand brand (0.76 $mg.l^{-1}$) and in the Purelife brand (0.18 $mg.l^{-1}$), respectively. These values are in agreement with the fluoride found in bottled waters used in other countries of the world [16-19].

In general, the concentration of fluoride for high consumed bottled water distributed in Ardabil city was between 0.15 to 0.87 $mg.l^{-1}$ and an average of 0.41 $mg.l^{-1}$ (less than the WHO standard) (Tables 2 and 3).

Table 1. Comparison of the mean fluoride measured with the amount of fluoride reported on the bottle label in Distributed Bottled Water in Ardabil City

Brands types	Fluoride concentration $mg.L^{-1}$	
	Distributed Bottled Water	fluoride reported on the bottle label
Vata	0.29	0.11
Atash	0.53	0.4
Aquafina	0.46	0
pana	0.35	0
DiDi	0.33	0.1
Pamin	0.29	0
Piorlif	0.18	0.07
Damavand	0.76	0
Oxab	0.44	0.1
Dasani	0.45	0.1

Table 2. Results of Standard deviation, mean, maximum and minimum fluoride values of the studied waters

Number of waters studied	Minimum	Maximum	mean	Standard deviation
30 samples (bottled water)	0.15	0.87	0.4073	0.9943
15 samples (water inlet to household water treatment systems)	0.0	0.78	0.4113	0.25509
15 samples (Outlet water from household water treatment systems)	0.0	0.36	0.1613	0.13495
60 samples (Total waters studied)	0.0	0.87	0.2473	0.22623

Table 3. International and regional fluoride set standard limits and optimal level.

	Fluoride (mgl^{-1}) standard limit	References
International		
U.S. EPA, U.S. FDA/CFR	2.0 and 4.0	22,39
No fluoride added[a]	1.4–2.4[b]	
Fluoride added[a]	0.8–1.7[b]	
No fluoride added[c]	1.4	
Fluoride added[c]	0.8	
EC/DWD	1.5	23
WHO	1.5	25
HHS	0.7	40
Regional		24
GCC	0.6–1.7	

[a] Packaged in the United States, [b] At retail store air temperature in the range of "79.3–90.5 °F" to''53.7 °F and below", respectively. [c] Imported to the United States.

To control the effects of fluoride, different international organizations have announced regulation and limits for its concentration. Fluoride is one of the minerals that has been classified under the National Drinking Water Regulations (NDWR) by the United States Environmental Protection Agency (U.S. EPA) [20]. Under the NDWR, fluoride is classified as the Primary standard, where the levels may lead to health effects, and as the secondary standards, where it may lead to aesthetic and cosmetic effects, depending on the level of the established standard [20]. Hence, the U.S. EPA and United States Food and Drug Administration/Code of Federal Regulations [21] have introduced standard levels for fluoride levels in drinking water [20,22]. Likewise, other standard limits for fluoride has been established by organizations such as the World Health Organization (WHO), the European Commission/Drinking Water Directive (EC/DWD), and the regional Gulf Cooperation Council (GCC) (Table 3) [23-25].

Based on the guidelines recommended by WHO, the highest level of fluoride concentration in drinking water is 1.5 $mg.l^{-1}$ $mg.l^{-1}$. The mentioned value is not a constant amount but can be modified based on local conditions, e.g., volume of water consumed and any additional sources of fluoride in the diet. To prevent excessive fluoride ingestion and dental caries, an appropriate fluoride in drinking water is recommended as 0.5-1 $mg.l^{-1}$ [26, 27]. Table 1 shows that, among 10 studied brands of bottled waters, 3 brands contain an insufficient amount of fluorine (<0.3 $mg.l^{-1}$). This rate leads to increasing the risk of tooth decay. In 50.0% of bottled waters, the fluoride concentrations were observed to be below to 0.5 $mg.l^{-1}$; the Ministry of Health and the Ministry of National Education has established this prophylactic dose in their strategy against dental decay in schools [18]. In these brands of bottled waters, the adequate dose of fluoride is not existed for children to guarantee their better oral health. Only 20.0% of bottled waters observed the standards (0.5 to 1.5 $mg.l^{-1}$) and were an important source of fluoride for young children to protecting against tooth decay.

Szmagara et al. (2019) conducted a study to survey the fluoride concentration in 24 samples of bottled mineral water; the results revealed that the fluoride concentration was lower than the standard values in all samples studied [28]. Abouleish et al. (2016) also conducted a study to examining fluoride levels in 23 brands of bottled water in United Arab Emirates. The results showed that fluoride concentration in all samples was lower than WHO standards [29]. The results of study conducted for evaluation of fluoride concentration in 9 different brands of bottled water in Brazil showed that fluoride concentrations were in the range of 0.07 and 0.63 $mg.l^{-1}$ and there was a significant difference between the values reported on the labels and the obtained results [30], which is in agreement with the results of the present study. Also, in this study, according to ANOVA analysis, there was a significant difference ($P = 0.014$) between the groups of bottled water with fluoride ion concentration.

The results of this study and the values reported on the labels of the bottled waters indicate that bottled waters have a wide range of parameter values and have different characters. A comparison of the study results with the reported values on the labels of bottled water for fluoride have been carried out in Table 1. As observed, four bottled water brands (Aquafina, Pana, Parmin, and Damavand) did not report the fluoride concentration on the label of their products. Therefore, the comparison cannot be carried out for this parameter. For other waters, the relationship between mean concentrations of analytical and labeled values was observed to be different. For the majority of these waters, the measured values were lower than the declared value.

The comparison of the results of the analysis of the present study did not correspond to the values reported on the bottles labels; therefore, continuous monitoring of these bottled water supply and production sites is necessary, and the information on the actual quality of the water and the values reported on the bottles labels should be consistent. The results of the present study on the mean fluoride content of bottled water showed that only two brands of bottled water called Thirsty mineral water with average fluoride mean of 0.53 $mg.l^{-1}$ and Damavand drinking water with mean fluoride of 0.76 $mg.l^{-1}$ was in the standard level, and the rest of the bottled waters had the fluoride concentrations below the standard limit. In the study conducted by Cochrane et al. (2006), the concentration of fluoride levels in 10 samples of bottled water in Australia was evaluated, and the results showed that 5 samples of these waters had fluoride content of 0.03 $mg.l^{-1}$ and less than the standard levels [31]. In another study, Ali Mohammadi et al. studied the probability of non-carcinogenic risk of fluoride due to the consumption of bottled water in different age groups and reported that the average concentration of fluoride in bottled water was 0.227 mg/ml, which is less than the WHO minimum guideline; thus, non-carcinogenic effects of fluoride due to the consumption of bottled water are very low [32].

The results related to mean fluoride concentration in the urban distribution network of 5 districts of Ardabil city from inlet and outlet of household water treatment systems showed that the fluoride range was 0 to 0.78 $mg.l^{-1}$ and the mean value was 0.41 $mg.l^{-1}$ (below WHO standard limit) (Tables **2** and **3**). Out of the 15 samples of inlet household water treatment systems, 7 samples had fluoride in the standard range, and the rest had fluoride below the standard level. Also, the examination of 15 samples of outlet water of household water treatment systems showed that fluoride ranged from zero to 0.36 $mg.l^{-1}$, and its mean value was 0.16 $mg.l^{-1}$, which was much lower than the standard recommended for drinking water. Therefore, household water treatment systems have an effect on the reduction of fluoride ion in drinking water. In total, the mean fluoride concentration in the 30 samples collected from the inlet and outlet of household water treatment systems was 0.228 $mg.l^{-1}$. In the present study, according to ANOVA analysis, there was a significant difference ($p = 0.002$) between water groups of household water treatment systems and fluoride ion concentration. According to the results of the T-test, there is no homogeneity of variance. Therefore, the assumption of the equality of two variances is rejected (Table 4). The results of the T-test show that there is a significant difference in the mean fluoride content of the studied waters. Also, according to Table 5, the concentration of fluoride was lower than the acceptable levels (0.5 mg.l-1) in all the studied samples (except for the input sample of district No.2).

Table 4. Comparison of mean fluoride in the studied waters

Comparison of mean fluoride	P	t	Homogeneity of variance
Bottled water and water inlet to household water treatment system	0	0	**There is no homogeneity of variance.**
Bottled water and A water outlet from the household water treatment system	0	0	**There is no homogeneity of variance.**
Inlet and outlet water from household water treatment system	0.003	3.355	**There is no homogeneity of variance.**

In the present study, the status of fluoride levels in drinking water sources (including inlet and outlet water from household water treatment systems and bottled water consumed in Ardabil city) was investigated. Based on the results of this study, the amount of fluoride in household water treatment systems is significantly lower than the desired level in urban water, and the efficiency of these systems for reducing the fluoride in the present study was obtained to be 63.60%. Decreasing the fluoride by household water treatment systems is one of the major disadvantages of these devices and may have human health implications. Of course, other similar studies of fluoride removal by these devices have approved the reduction of its level to values less than the drinking water standard. Our result is in agreement with the results of studies conducted by Tavangar et al. [33], Miranzadeh and Rabbani [34], Dehghani et al. [35], and Matloob [36]. The results of the study also showed that the fluoride content of the waters studied was less than the WHO standard and National Drinking Water Standard No.1053 (fluoride level of 0.5 to 1.5 mg). This has been consistent with our results regarding the fluoride content of the waters surveyed in Ardabil. The reason for this event is the fact that, in the reverse osmosis (RO) process, all ions are removed irrespective of their usefulness, and there is no selective removal mode, thus one of the major disadvantages of these devices is the reduction of fluoride concentration to values lower than standard levels.

The results of fluoride removal efficiency by household water treatment systems are presented in Table 6. According to Table 6, household water treatment systems No.3, No.12, and No.15, with 100% removal efficiency, had the highest fluoride removal efficiency and water treatment system No. 7, with a removal efficiency of 36.96%, had the least role in decreasing water fluoride. Overall, the reduction efficiency of the studied water treatment system was 67.25%. This is consistent with the results of the study by [37]. In another study conducted by Eftekhar et al. (2015), the performance of six types of household water treatment systems to remove fluoride examined; the results other study showed that in all 6 brands of household water treatment systems, the amount of fluoride in the output was significantly reduced [38], which was in agreement with the results of the present study.

Table 5. Average Fluoride Concentration in Inlet and Outlet of Household Water treatment system in 5 Areas of Ardabil and Comparison with Drinking Water Standard 1053

Brands types	**Fluoride concentration mg.L^{-1}**		
	Average fluoride	**Desired limit**	**Maximum allowed**
Vata	1.5	0.5	0.07
Atash	1.5	0.5	0.023
Aquafina	1.5	0.5	0.64
pana	1.5	0.5	0.29
DiDi	1.5	0.5	0.47
Pamin	1.5	0.5	0.24
Piorlif	1.5	0.5	0.38
Damavand	1.5	0.5	0.05
Oxab	1.5	0.5	0.48
Dasani	1.5	0.5	0.19

Table 6. Fluoride removal efficiency by household water treatment system

Fluoride removal (%)														
HW1	**HW2**	**HW3**	**HW4**	**HW5**	**HW6**	**HW7**	**HW8**	**HW9**	**HW10**	**HW11**	**HW12**	**HW13**	**HW14**	**HW15**
58.82	0	100	64.52	50	52.38	36.96	52.83	55.81	62.5	88.23	100	65.62	53.85	100

CONCLUSIONS

Based on results obtained from studying on 60 water samples, the minimum fluoride concentration, maximum fluoride concentration, mean fluoride concentration, and standard deviation were 0, 0.87 mg.l^{-1}, 0.35, and 0.226, respectively. The statistical analysis of the achieved data using ANOVA indicated that there are significant differences between the groups in terms of fluoride concentration (P = 0.001). This may be due to the reduction of fluoride by the household water treatment system and the difference in water resources as the quality of drinking water in each area is often influenced by the geological structure of that area and varies from region to region. Based on the results of the present study, the concentration of fluoride in most samples was lower than the standard. Since drinking water is considered as an important pathway for fluoride intake, and the fluoride concentration from 0.5 to 1.5 mg/day is advantageous for the growth of teeth and bones, the amount of fluoride in Ardebil drinking water network and bottled water should be increased. However, it should be mentioned that the other sources, such as processed foods and drinks, toothpaste, etc. play a role in the intake of fluoride. Therefore, monitoring fluoride concentration in different foods is recommenced to calculate the total amount of fluoride absorption by the human body through all sources of fluoride.

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Conflict of interests

The authors declare that there is no conflict of interest.

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