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FLUORIDE CONTENT OF COCONUT WATER AND ITS RISK ASSESSMENT

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ABSTRACT: The coconut water in 15 coconuts bought in 7 cities in Iran was examined for various physicochemical properties (electrical conductivity [EC], total dissolved solids [TDS], turbidity, pH, and fluoride [F] concentration) and the risk associated with the intake of F from coconut water was estimated. The SPADNS Zirconium Lake method was applied for the determination of the F concentration. The results showed that coconut water has a significant F concentration, (mean = 3.401 mg/L, range = 2.680–4.160 mg/L). Significant correlations (Pearson) were found between the EC and TDS (0.980) and between the EC and turbidity (–0.525). The average F intake from the consumption of 350 and 700 mL of coconut water is about 15% and 30%, respectively, of the maximum acceptable daily intake of the fluoride of 0.1 mg/kg body weight/day. The fluoride intake from coconut water should be taken into account when calculating the dietary F intake.

Keywords: Coconut water; Fluoride; Fluoride intake; Risk assessment.

INTRODUCTION

Coconut water, the juice of the endosperm found within the cavity of the coconut, is able to be used as a solution for oral and even intravenous rehydration[1] and to reduce the toxic effects of nicotine and alcohol on reproductive function in men.[2] Coconut water accounts for 25% of the weight of the fruit, and its basic composition is 95.5% water, 4% carbohydrates, 0.1% fat, 0.02% calcium, 0.01% phosphorous, and 0.5% iron, in addition to amino acids, vitamin C, B complex vitamins, and mineral salts.[1] Accurate values of dietary intakes of toxic elements are of importance in correlating epidemiological and laboratory toxicity studies with actual intakes in order to reveal potential problems.[3] There are only a small number of studies on the composition of coconut water[1] and they lack a consideration of toxicity. Nowadays, there are large numbers of studies concerning the fluoride content of foods[3-7] and drinks.[8-12] Fluoride (F) is not essential for human growth and development but is considered, by the Scientific Panel on Dietetic Products, Nutrition and Allergies of the European Food Safety Authority, to be beneficial in the prevention of dental caries (by increasing enamel acid resistance) when ingested in amounts of about 0.05 mg/kg body weight per day and when applied topically with dental products such as toothpaste.[13] However, an excessive intake of fluoride can lead to a reduced mineral content of enamel and to dental fluorosis, which predominantly affects permanent, rather than deciduous, teeth.[13-15] Since water is a major source of dietary fluoride,[7,16] many studies on this subject are related to water quality.[10,17-19] However, foods, drinks, and even food seasonings like table salt also make a significant contribution.[6,7] The upper intake level (UL) for fluoride is considered to be 0.1 mg fluoride/kg body weight (bw)/day in children aged 1–8 years.[13] It has been suggested

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that the incidence and severity of enamel fluoride mottling or fluorosis has become greater in the last decade.[4] Knowledge of fluoride intake is important to optimize its caries-preventive role and while simultaneously preventing adverse effects due to overconsumption. Therefore, the measurement of the fluoride intake usually requires information on the fluoride concentration in foods and drinks.[4,7,12,20] The aim of the present study was to measure the fluoride concentration in the juice of coconuts bought from Iranian markets and to estimate the risk associated with this form of fluoride intake.

MATERIALS AND METHODS

Reagents: All the reagents were prepared by means of analytical grade chemicals (Merck, Germany) and were used while fresh. All the procedures used for the preparation and application of the reagents were in accordance with the relevant reference methods.[21-25]

Preparation and extraction of the coconut juice: Fifteen coconuts were bought from different grocery markets and fruit shops in 7 cities in Iran. All the coconuts were prepared prior to the juice extraction by removing their foliage and purging their shells from dirt by washing them three times in a row using distilled water and letting the shells dry at room temperature. The coconut water was extracted through a drilled hole (using a 5 mm gimlet) and the volume of the juice harvested from each coconut was measured separately and immediately using a graduated cylinder. The samples were then kept refrigerated in PTFE air-tight vessels and examined in less than an hour after the juice extraction.

pH, EC, turbidity and total dissolved solids (TDS) measurement: The TDS of the samples was calculated from their electrical conductivity (EC) which was measured in accordance with the reference methods.[21,25,26] The measurement of pH took place according to the standard methods and procedures.[22,24,26] The turbidity of the samples was measured in compliance with the standard methods[23,26,27] instantly after the volume measurement in order to minimize its alteration by coagulation, oxidation, and other confounders.

Fluoride measurement: Adequate volumes of the homogenized coconut water samples were filtered through individual 0.45 µm pore sized Membrane Filters (MF-Millipore™-HAWP04700). These filtered coconut water samples were analyzed separately for the determination of the fluoride concentration by the Spadns Zirconium Lake method.[26,28] in compliance with the analyzer device (DR 5000™ UV-Vis Spectrophotometer) user manual.[29] Equal volumes (2 mL) of SPANDS solution were used for all the equal sample volumes (10 mL).

Health risk assessment: The human health risk assessment process was used to estimate the probability of adverse effects from an excessive intake of fluoride intake. First, based on our personal daily consumption of one and two 350 mL glasses of coconut water, the daily consumption of coconut water was taken to be either 350 or 700 mL a day. The estimated daily intake (EDI) of fluoride due to drinking of coconut water was then calculated using equation 1.

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

Where:

EDI = Estimated daily intake (mg.kg of body weight^{-1}.day^{-1}
C_f = Concentration of fluoride in coconut juice (mg/L)
C_d = Daily consumption of coconut water (L)
B_w = Body weight (kg)

The daily upper intake level (UL) for fluoride of 0.1 mg/kg body weight/day[30] was considered as its reference dose (Ref. D). The associated hazard quotient (H_Q) was then calculated using equation 2.

$$H_Q = \frac{EDI}{Ref.\ D} \quad (2)$$

Where:

H_Q = Hazard quotient
EDI = Estimated daily intake (mg.kg of body weight^{-1}.day^{-1}
$Ref.\ D$ = Fluoride reference dose (0.1 mg/kg bw/day)

It is noteworthy that the correct calculation of the EDI for fluoride requires the consideration of every available source of fluoride intake including foods,[4-7,12,20] water,[8-10,17,31-33] tea,[11] and the other usual and unusual[34] sources of exposure. When the H_Q value is less than 1, it indicates that experiencing any adverse effect is unlikely for the exposed person, while a value greater than 1 suggests that the imposed health risk from this EDI is more than the acceptable level and there is a considerable chance of adverse consequence. It worth mentioning that when a net EDI for a specific source was used in this estimation, an H_Q value less than 1 showed that source's contribution to the total ADI of fluoride. When the H_Q value is greater than 1, it is necessary to reduce the fluoride intake by substituting, omitting, and treating.[18,19,35,36]

Quality control: The analyzer and measuring probes were calibrated according to their manuals. All the standard solutions were prepared by means of analytical grade chemicals (Merck, Germany) and used freshly. All the experiments were carried out three times in a row to mitigate analyzer measurement bias (by reporting the average of all three measurements). Since the fluoride concentration in all the juice samples were above the analyzer's upper limit of quantification (ULOQ), double distilled water was used for the dilution of the pure juice to the desired concentration and the resulting data were corrected proportionally based on the dilution factor. The distilled water was also checked for its fluoride content and its concentration was under the analyzer's limit of detection (LOD).

RESULTS AND DISCUSSION

The descriptive statistics of the analyzed parameters, of the 15 coconuts bought from different grocery markets in Iran, are shown in Table 1. The smallest range of variation occurred with the pH, followed by the fluoride concentration, and the widest range of variation occurred with the turbidity. The maximum fluoride concentration of coconut juice (range 2.680–4.160 mg/L) is a little more than the enforceable drinking water standard of 4.0 and the mean fluoride concentration of the coconut juice (mean 3.401 mg/L) is greater than the secondary drinking standard of 2.0 mg/L).[32,37]

Table 1. The descriptive statistics of the analyzed parameters

Variable	Minimum	Maximum	Mean	Standard deviation
Sample volume (mL)	35.000	350.000	114.333	89.040
Turbidity (NTU)	16.700	429.000	213.433	118.026
pH	5.200	5.800	5.513	0.188
EC (ms/cm)	8.210	14.290	10.881	1.594
TDS (g/L)	4.110	8.200	5.739	1.188
F (mg/L)	2.680	4.160	3.401	0.479

Figure 1 shows the coconut water fluoride concentration in the 15 coconuts bought from 7 different cities in Iran. The number of coconuts bought in each city was: Tehran = 3, Shemiran = 3, Mashhad = 2, Oromie = 2, Yazd = 2, Bushehr = 2, and Bandar abbas = 1.

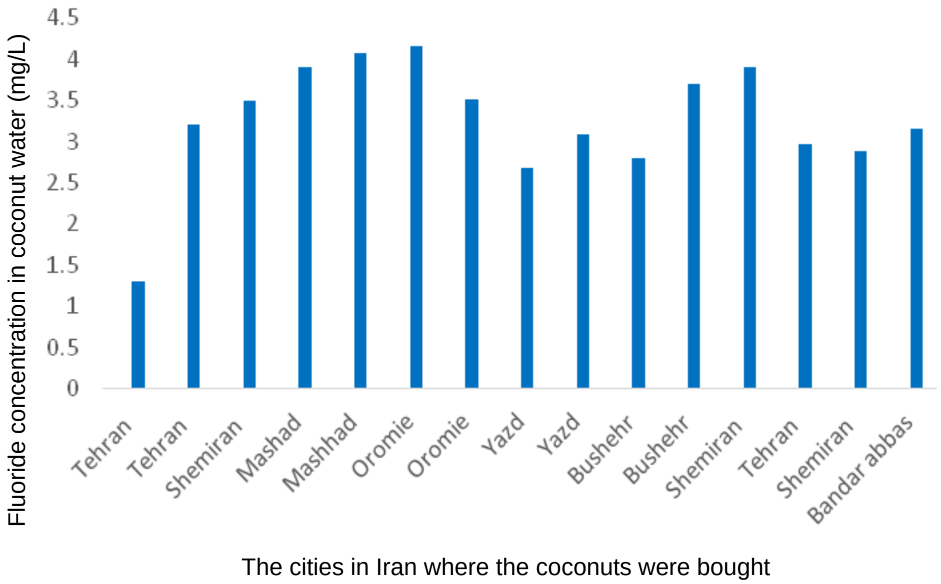

Figure 1. The concentration of the fluoride in the coconut water of the coconuts bought in 7 Iranian cities.

Table 2 shows the results of the different normality tests used for the investigation of this study's experiments results. The values in bold mean that one can reject the null hypothesis H_0 and indicate that the variable extracted from the sample does not follow a normal distribution.

Table 2. The results of the different normality tests used to analyze the experimental results of the study

Variable	Normality test			
	Shapiro-Wilk	Anderson-Darling	Lilliefors	Jarque-Bera
Sample volume	**0.000***	**< 0.0001***	**0.000***	**0.002***
Turbidity (NTU)	0.916	0.910	0.958	0.888
pH	0.204	0.156	0.089	0.830
EC (ms/cm)	0.600	0.446	0.451	0.703
TDS (g/L)	0.222	0.242	0.277	0.525
F (mg/L)	0.511	0.582	0.719	0.607

*The values in bold mean that one can reject the null hypothesis H_0 and indicate that the variable extracted from the sample does not follow a normal distribution.

Table 3 shows the Spearman's correlation matrix for the analyzed parameters of the tested coconuts. By omitting of the variable which did not follow a normal distribution (i.e., the sample volume) from the rest of the results, the Pearson's correlation test became useful to check for relationships among the other variables. There was a significant and strong correlation between the EC and the TDS (which was predictable since the TDS can be estimated based on the EC). There was also a significant but moderate positive linear relationship between the sample volume and its turbidity. The values in bold in Table 3 are different from 0 with a significance level alpha=0.05.

Table 3. The analyzed parameters' correlation matrix (Spearman)

Variable	Sample volume	Turbidity (NTU)	pH	EC (ms/cm)	TDS (g/L)	F (mg/L)
Sample volume	**1***	**–0.600***	0.472	.–0.088	0.000	0.156
Turbidity (NTU)	**–0.600**	**1***	–0.089	0.111	0.043	–0.451
pH	0.472	–0.089	**1***	–0.425	–0.372	–0.049
EC (ms/cm)	–0.088	0.111	–0.425	**1***	**0.949***	–0.245
TDS (g/L)	0.000	0.043	–0.372	**0.949***	**1***	–0.113
F (mg/L)	0.156	–0.451	–0.049	–0.245	–0.113	**1***

The values in bold are different from 0 with a significance level alpha=0.05.

Table 4 shows the Pearson's correlation matrix for the analyzed parameters of the tested coconuts (except for the sample volume). There was a significant but moderate correlation between the EC and the pH of the samples. The values in bold in Table 4 are different from 0 with a significance level alpha=0.05.

Table 4. The analyzed parameters' correlation matrix (Pearson)

Variables	Turbidity (NTU)	pH	EC (ms/cm)	TDS (g/L)	Fluoride (mg/L)
Turbidity (NTU)	**1***	–0.083	0.284	0.263	–0.456
pH	–0.083	**1***	**–0.525***	–0.462	–0.021
EC (ms/cm)	0.284	**–0.525***	**1***	**0.980***	–0.217
TDS (g/L)	0.263	–0.462	**0.980***	**1***	–0.163
Fluoride (mg/L)	–0.456	–0.021	–0.217	–0.163	**1***

*The values in bold are different from 0 with a significance level alpha=0.05.

Figure 2 shows the cumulative distributions diagram for the sample volumes versus their turbidity.

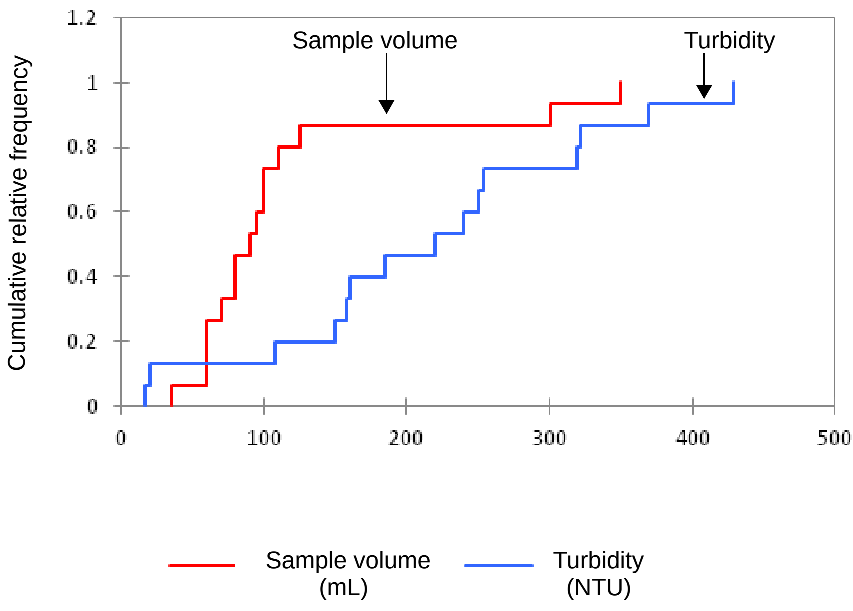

Figure 2. Cumulative distributions (sample volume / turbidity [NTU]) diagram.

Table 5 shows the two-sample Kolmogorov-Smirnov test results between the sample volume and the turbidity variables. As the computed p-value is lower than the significance level alpha=0.05, one should reject the null hypothesis H_0, and accept the alternative hypothesis H_a. This means that the distributions of the two samples are different.

Table 5. Two-sample Kolmogorov-Smirnov test results

D	0.667
p-value	0.003
alpha	0.05

Table 6 shows the minimum, the average, and the maximum H_Q values for fluoride intake from the consumption of 350 and 700 mL/day of coconut water with a mean F content of 3.401 mg/L and a F range of 2.680–4.160 mg/L. The HQ values were calculated based on the net fluoride intake from the coconut juice with no consideration of fluoride from other sources. The EDI was calculated based on a standard body weight of 76 kg for men.[38] The average daily fluoride intakes from the daily consumption of 350 and 700 mL of coconut water (1.19 and 2.38 mg/day, respectively) were approximately 15% and 30%, respectively, of the maximum acceptable intake of the fluoride (7.6 mg/day for a 76 kg man with a maximum acceptable intake of 0.1 mg/kg bw/day).[30]

Table 6. The minimum, average, and maximum H_Q values* for the fluoride intake from the consumption of coconut water

Parameter	Daily coconut water consumption	
	350 mL	700 mL
Minimum H_Q	0.123	0.247
Average H_Q	0.157	0.313
Maximum H_Q	0.192	0.383

*The minimum, average, and maximum H_Q values were calculated using coconut water fluoride concentrations of 2.680, 3.401, and 4.160 mg/L corresponding to the minimum, mean, and maximum fluoride concentrations, respectively.

CONCLUSIONS

It became apparent that the fluoride levels in coconut water of the coconuts tested in our study were significant and that taking this intake into account is a prerequisite for correctly calculating total dietary intake of fluoride, since its coconut water fluoride concentration is much more than of that reported for some other foods and drinks.[20] It is necessary to reduce coconut water consumption in some areas like Koohbanan[12] where the fluoride concentration of local foods and drinks is high. Coconut water is not a suitable drink for infants since its fluoride concentration is much more than that in common infant foods and drinks.[4] Further studies are suggested to assess the effect on the coconut pulp and the coconut water fluoride concentrations of the storage conditions for the fruit and the elapsed shelf time. As there were significant differences in the coconut water fluoride concentrations among the different fruits examined in our study, which is consistent with the literature, it is recommended that investigations be done on imported fruits in order to determine the best sources for each product.[5]

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REFERENCES

1 Vigliar R, Sdepanian VL, Fagundes-Neto U. Biochemical profile of coconut water from coconut palms planted in an inland region. Jornal de Pediatria 2006; 82:308-12.
2 Gopalakrishnan Nair SV, Rajamohan T. The role of coconut water on nicotine-induced reproductive dysfunction in experimental male rat model. Food and Nutrition Sciences 2014;5:1121-30.
3 Daberka RW, McKenzie AD, Lacroix GMA. Dietary intakes of lead, cadmium, arsenic and fluoride by Canadian adults: a 24-hour duplicate diet study. Food Additives and Contaminants 1987;4:89-102.
4 Vlachou A, Drummond BK, Curzon MEl. Fluoride concentrations of infant foods and drinks in the United Kingdom. Caries Res 1992;26:29-32.
5 Waldbott GL. Fluoride in food. American Journal of Clinical Nutrition 1963;12:455-62.
6 Yousefian F, Hosseini SS, Mahvi AH. Investigation of the fluoride content of edible salt and the fluoride intake from edible salt consumption in Iran. Fluoride 2016;49:495-502.
7 Zohouri FV, Rugg-Gunn AJ. Fluoride concentration in foods from Iran. International Journal of Food Sciences and Nutrition 1999;50:265-74.
8 Dobaradaran S, Mahvi AH, Dehdashti S. Fluoride content of bottled drinking water available in Iran. Fluoride 2008;41:93-4.
9 Dobaradaran S, Mahvi AH, Dehdashti S, Abadi DRV. Drinking water fluoride and child dental caries in Dashtestan, Iran. Fluoride 2008;41:220-6.
10 Fouladi Fard R, Mahvi AH, Hosseini SS, Khazaei M. Fluoride concentrations in bottled drinking water available in Najaf and Karbala, Iraq. Fluoride 2014;47:249-52.
11 Mahvi AH, Zazoli MA, Younecian M, Esfandiari Y. Fluoride content of Iranian black tea and tea liquor. Fluoride 2006;39:266-8.
12 Poureslami HR, Khazaeli P, Noori GR. Fluoride in food and water consumed in Koohbanan (Kuh-e Banan), Iran. Fluoride 2008;41:216-9.
13 European Food Safety Authority (EFSA). Opinion of the Scientific Panel on Dietetic Products, Nutrition and Allergies on a request from the Commission related to the tolerable upper intake level of fluoride. The EFSA Journal 2005;192:1-65. Available from: http://www.efsa.eu.int/science/nda/nda_opinions/catindex_en.html
14 Hosseini SS, Mahvi AH. Removal of fluoride from drinking water by freezing technology. Fluoride 2019;52(3 Pt 1):231-47.
15 Hosseini SS, Pasalari H, Yousefi N, Mahvi AH. Eggshell modified with alum as low-cost sorbent for the removal of fluoride from aquatic environments: isotherm and kinetic studies. Desalination and Water Treatment 2019;146:326-32.
16 Yousefi M, Ghoochani M, Mahvi AH. Health risk assessment to fluoride in drinking water of rural residents living in the Poldasht city, Northwest of Iran. Ecotoxicology and Environmental Safety 2018;148:426-30.
17 Nouri J, Mahvi AH, Babaei A, Ahmadpour E. Regional pattern distribution of groundwater fluoride in the Shush aquifer of Khuzestan County, Iran. Fluoride 2006;39:321-5.
18 Yousefi N, Fatehizedeh A, Ghadiri K, Mirzaei N, Ashrafi SD, Mahvi AH. Application of nanofilter in removal of phosphate, fluoride and nitrite from groundwater. Desalination and Water Treatment 2016;57:11782-8.

19 Zazouli MA, Mahvi AH, Dobaradaran S, Barafrashtehpour M, Mahdavi Y, Balarak D. Adsorption of fluoride from aqueous solution by modified *Azolla filiculoides*. Fluoride 2014;47 349-58.

20 Jackson RD, Brizendine EJ, Kelly SA, Stookey GK, Hinesley R, Dunipace AJ. The fluoride content of foods and beverages from negligibly and optimally fluoridated communities. Community Dent Oral Epidemiol 2002;30:382-91.

21 American Society for Testing and Materials. ASTM D1125-95(2005) Standard test methods for electrical conductivity and resistivity of water. West Conshohocken, Pennsylvania, USA: ASTM International (ASTM); 2005. pp. 1-7.

22 Buck RP, Rondinini S, Covington AK, Baucke FGK, Brett CMA, Camoes MF, et al. Measurement of pH. Definition, standards, and procedures. Pure Appl Chem 2002;74:2169-200.

23 Gregorio D, Burres E. Turbidity using a nephelometer (“Turbidimeter”). Standard Operating Procedure (SOP) 3.1.5.4. The Clean Water Team Guidance Compendium for Watershed Monitoring and Assessment; 2010. pp. 1-3.

24 Katznelson R. Measurement of pH with a pocket meter. Standard Operating Procedure (SOP) 3.1.4.3. The Clean Water Team Guidance Compendium for Watershed Monitoring and Assessment; 2002. pp. 1-3.

25 Katznelson R. Measurement of electrical conductivity using a pocket meter DQM Standard Operating Procedure (SOP) 3131 (V3). The Clean Water Team Guidance Compendium for Watershed Monitoring and Assessment 2004, p. 1-9.

26 Eaton AD, Franson MAH. Standard Methods for the Examination of Water & Wastewater. 21st ed. Washington, D.C: American Public Health Association, American Water Works Association, Water Environment Federation; 2005.

27 O’Dell JW, editor. Method 180.1 Determination of turbidity by nephelometry. Revision 2.0. Cincinnati, Ohio, USA: Environmental Monitoring Systems Laboratory, Office of Research and Development, US Environmental Protection Agency; 1993.

28 US EPA. Method 13A - Determination of total fluoride emissions from stationary sources (Spadns Zirconium Lake Method). US EPA, 2013, pp. 1-14.

29 Hach C. DR5000™ Spectrophotometer procedures manual. 2nd ed. Germany: Hach; 2005, pp. 1-864.

30 European Food Safety Authority (EFSA). Nitrate in vegetables. Scientific Opinion of the Panel on Contaminants in the Food Chain. The EFSA Journal 2008; 689:1-79.

31 Mahvi AH, Zazoli MA, Younecian M, Nicpour B, Babapour A. Survey of fluoride concentration in drinking water sources and prevalence of DMFT in the 12 years old students in Behshar City. Journal of Medical Sciences 2006;6:658-61.

32 Rahmani A, Rahmani K, Dobaradaran S, Mahvi AH, Mhamadjani R, Rahmani H. Child dental caries in relation to fluoride and some inorganic constituents in drinking water in Arsanjan, Iran. Fluoride 2010;43:179-86.

33 Rahmani A, Rahmani K, Mahvi AH, Usefie M. Drinking water fluoride and child dental caries in Noorabademamasani, Iran. Fluoride 2010;43:187-93.

34 Dobaradaran S, Fazelinia F, Mahvi AH, Hosseini SS. Particulate airborne fluoride from an aluminium production plant in Arak, Iran. Fluoride 2009;42:228-32.

35 Bazrafshan E, Ownagh KA, Mahvi AH. Application of electrocoagulation process using iron and aluminum electrodes for fluoride removal from aqueous environment. E-Journal of Chemistry 2012;9:2297-308.

36 Boldaji MR, Mahvi AH, Dobaradaran S, Hosseini SS. Evaluating the effectiveness of a hybrid sorbent resin in removing fluoride from water. International Journal of Environmental Science and Technology 2009;6:629-32.

37 US EPA. Questions and answers on fluoride. Office of Water (4606M), 2011, pp. 1-10. Available from: https://www.epa.gov/sites/production/files/2014-12/documents/2011_fluoride_questionsanswers.pdf

38 Australian Government National Health and Medical Research Council (NHMRC). Fluoride (updated 20170. In: Nutrient reference values for Australia and New Zealand. Commonwealth of Australia; 2017. pp. 165-70. Available from: https://www.nrv.gov.au/sites/default/files/content/Fluoride%20section.pdf