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## ASSESSING THE NON-CARCINOGENIC RISK DUE TO THE INTAKE OF FLUORIDE FROM FRUIT JUICE AVAILABLE IN THE MARKET IN BUSHEHR

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ABSTRACT: In the present study, the fluoride (F) concentration, estimated daily intake (EDI), and non-carcinogenic risk of different fruit juices were determined. A total of 135 fruit juice samples from different brands and fruit types (cherry, orange, grape, apple, pineapple, and peach) were analyzed for fluoride concentration levels by using an ion selective electrode. The range of the fluoride concentration of the fruit juice samples was 0.00–0.444 mg.L<sup>-1</sup> with a mean value of 0.069 mg.L<sup>-1</sup>. The results indicated that there were statistically significant differences between the fluoride concentration levels of some examined fruit juice brands as well as the fruit types. There was no statistically significant different age groups showed that there was no significant risk for consumers from consuming fruit juice. However, the fluoride intake from other drinks containing a high level of fluoride should be considered when estimating the total daily fluoride intake from drinking, especially for those in warm regions with a high level of drink consumption.

Keywords: Dietary intake; Fluoride; Fruit juice; Risk assessment.

# INTRODUCTION

Fluoride is one of the most important elements in the earth and may enter the environment naturally or as a result of human activities such as volcanic emissions, weathering and dissolution of minerals, marine aerosols, production of phosphate fertilizers, the manufacture and use of hydrofluoric acid, the production of aluminum, steel, and oil, the burning of fluoride-rich coal, and water flow and sediment movement from aluminum production plants.<sup>1</sup> Fluoride may enter the human body via food and beverage consumption and may cause adverse health effects<sup>2-5</sup> such as those affecting the skeleton, teeth, liver, kidney, pancreas, and brain. Many studies have reported on the health effects of various concentration levels of fluoride in

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drinking water, underground water, sea water, milk, foods, fish, and soil as well as on its removal from aqueous solutions.<sup>6-30</sup> Many reports have been published on water containing an elevated fluoride concentration but there are only a limited number of studies on the fluoride content of other drinks such fruit juice. A Mexican study<sup>31</sup> found the fluoride concentration levels of fruit juice ranged from 0.08 to 1.42 mg.L<sup>-1</sup> with a mean value of 0.67 mg.L<sup>-1</sup> and grape juice contained the highest fluoride concentration with a mean value of 1.39 mg.L<sup>-1</sup>. In another study,<sup>32</sup> the fluoride concentration levels of different fruit juices ranged from 0.09 to 0.25 mg.L<sup>-1</sup>. It is necessary to control the consumption of fluoride in child populations<sup>33</sup> and an important key to this is the evaluation of level of fluoride exposure and the source of the exposure, especially during the first five years of life when there is a greater risk of developing dental fluorosis, an aesthetic concern in teeth.<sup>34</sup> Therefore, it is necessary to assess the fluoride content and health risk of fluoride exposure from fruit juice consumption, especially in warm regions such as the Middle East with a high level of drinks consumption, particularly fruit juices. Thus the aims of the present study were to determine: (i) the fluoride concentration of different brands of fruit juice; (ii) the level of fluoride intake from fruit juice consumption and the associated health risk.

#### **MATERIALS & METHODS**

Fruit juice samples, with six different types of fruit (apple, cherry, grape, orange, peach, and pineapple), of 10 different brands were purchased from supermarkets and grocery stores. Three samples of each brand were collected from different areas of Bushehr Port during February to March 2019. A total of 135 samples were obtained. The information on the packaging of the samples was recorded and the samples were kept in a refrigerator in their original closed containers until analysis. For measuring the fluoride concentration of the samples, they were first diluted 1:1 with TISAB II buffer solution to adjust the ionic strength and then the fluoride concentration was measured using an ion selective electrode (model 780, Metrohm). The fluoride content of every sample was measured in triplicate and the mean values were calculated. For evaluation of the health risk assessment, five different age groups were considered as 4–9, 10–17, 18–29, 30–49, and 50–65 years old. The estimated daily intake (EDI) of fluoride was determined by using the following equation:

$$\mathsf{EDI} = \frac{\mathsf{Cf} \times \mathsf{Cd}}{\mathsf{bw}}$$

Where: (mg.kg bw<sup>-1</sup>.day<sup>-1</sup>) Estimated daily intake EDI =  $(mq.L^{-1})$ Cf Fluoride concentration in fruit juice = Average daily consumption of fruit juice (L.day<sup>-1</sup>) Cd = Body weight bw (kg)=

The EDI values (mg.kg bw<sup>-1</sup>.day<sup>-1</sup>) of fluoride were calculated based on the average daily consumption of juice (Cd) in L.day<sup>-1</sup>, the fluoride concentration in the juice (Cf) in mg.L<sup>-1</sup>, and the body weight (bw) in kg. The body weights of the different age groups, of 4–9, 10–17, 18–29, 30–49, and 50–65 years old, were considered to be 25.2, 44.3, 65.76, 70.34, and 69.17 kg, respectively.<sup>35,36</sup> The average juice consumption rates, in the target groups of 4–9, 10–17, 18–29, 30–49,

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and 50–65 years old, were considered to be 75, 55.5, 84, 65, and 42 mL, respectively.<sup>37,38</sup> The hazard quotient (HQ) was determined to assess non-carcinogenic risk of fluoride by the following equation:

Where:

= EDI RFD

HQ=Hazard quotientEDI=Estimated daily intake $(mg.kg bw^{-1}.day^{-1})$ RFD=Oral reference dose $(mg.kg^{-1}.day^{-1})$ 

HQ

The oral reference dose (RFD) of fluoride (0.06 mg.kg<sup>-1</sup>.day<sup>-1</sup>) was obtained from the data-base of Integrated Risk Information System, USEPA.<sup>39</sup> An HQ value less than 1 indicates a low probability of adverse health effects in a population, while an HQ value greater than 1 shows an increment in the probability of adverse effects on humans.<sup>40</sup>

### **RESULTS AND DISCUSSION**

The fluoride concentration levels of the fruit juice samples ranged from 0.00 to 0.444 mg.L<sup>-1</sup>, with a mean concentration of 0.069 mg.L<sup>-1</sup> (Tables 1A–1C). The H brand contained the lowest concentration of fluoride, with a mean value of 0.004 mg.L<sup>-1</sup>, while the B brand had the highest fluoride concentration, with a mean value of 0.229 mg.L<sup>-1</sup>. The grape juice sample contained the highest fluoride concentration, with a mean value of 0.165 mg.L<sup>-1</sup>, while the peach juice samples, with a mean value of 0.045 mg.L<sup>-1</sup>, had the lowest fluoride concentration.

Brand (type of fruit)	Range	Mean ± SD	рН
A (cherry)	0.046-0.058	0.053±0.004	4.51
A (orange)	0.001–0.015	0.005±0.005	4.79
A (grape)	0.065–0.093	0.080±0.011	4.85
A (pineapple)	0.000-0.002	$0.002 \pm 0.0005$	4.95
A (peach)	0.00-0.026	0.016±0.004	5.2
Sub total	0.00–0.093	0.030±0.029	4.92

 Table 1 A. Fluoride concentration level (range and mean±SD, mg.L<sup>-1</sup>) and pH value of the fruit juice samples

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 Table 1 B. Fluoride concentration level (range and mean±SD, mg.L<sup>-1</sup>) and pH value of the fruit juice samples

	-	-		
Brand (type of fruit)	Range	Mean ± SD	рН	
B (cherry) 0.283–0.346		0.323±0.030	4.53	
B (orange)	0.089–0.199	0.166±0.019	4.51	
B (grape)	0.162–0.262	0.248±0.017	4.7	
B (apple)	0.234–0.288	0.266±0.025	4.8	
B (pineapple)	0.135–0.214	0.175±0.039	4.89	
B (peach)	0.150-0.444	0.197±0.103	5	
Sub total	0.089-0.444	0.229±0.083	4.77	
C (cherry)	0.084–0.130	0.102±0.022	4.72	
C (orange)	0.000-0.000	0.000±0.000	4.56	
C (grape)	0.049–0.130	$0.051 \pm 0.0005$	4.8	
C (apple)	0.46-0.124	$0.095 \pm 0.024$	5.23	
C (pineapple)	0.043–0.069	0.051±0.008	5.1	
C (peach)	0.000-0.000	0.000±0.000	4.91	
Sub total	0.043–0.130	0.05±0.043	4.90	
D (cherry)	0.09–0.159	0.128±0.024	4.66	
D (orange)	0.046–0.079	0.058±0.012	4.76	
D (apple)	0.159–0.179	0.17±0.006	5.22	
D (pineapple)	0.097–0.129	0.115±0.011	4.95	
D (peach)	0.006–0.019	0.055±0.039	5.19	
Sub total	0.006-0.179	0.092±0.049	4.95	
E (cherry)	0.013-0.037	0.024±0.009	4.39	
E (orange)	0.000-0.000	$0.000 \pm 0.000$	4.27	
E (grape)	0.043–0.049	0.029±0.002	4.7	
E (pineapple)	0.000-0.000	0.000 ±0.000	4.55	
E (peach)	0.006–0.012	0.009±0.002	4.85	
Sub total	0.006–0.049	0.025±0.017	4.55	

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Brand (type of fruit) Range Mean ± SD pН F (cherry) 0.159-0.213 0.196±0.016 4.58 F (grape) 0.384-0.429 0.406±0.018 4.76 F (apple) 0.009-0.012 0.011±0.001 5.19 0.000-0.000  $0.000 \pm 0.000$ F (pineapple) 4.87 F (peach) 0.001-0.008 0.005±0.003 5.07 Sub total 0.001-0.429 0.123-0.162 4.89 G (cherry) 0.141±0.012 4.64 0.124-0.158 G (apple) 0.093-0.119 0.107±0.010 4.87 G (pineapple) 0.000-0.000  $0.000 \pm 0.000$ 4.85 G (peach) 0.024-0.034 0.03±0.002 5.05 Sub total 0.024-0.158 0.07±0.058 4.85 0.000-0.008 0.003±0.002 4.25 H (cherry) H (orange) 0.000-0.000  $0.000 \pm 0.000$ 4.35  $0.000 \pm 0.00$ 4.51 H (pineapple) 0.000-0.000 H (peach) 0.001-0.011 0.007±0.003 4.88 Sub total 0.000-0.011 4.50 0.004±0.005 4.69 I (orange) 0.037-0.041 0.039±0.001 I (peach) 0.036-0.036 0.038±0.0002 4.89 Sub total 0.036-0.041 4.79 0.039±0.001 0.034-0.042 0.037±0.003 4.87 J (grape) 0.017-0.018 J (pineapple) 0.017±0.0005 4.54 Sub total 0.017-0.042 0.027±0.01 4.7

# Table 1 C. Fluoride concentration level (range and mean±SD, mg.L<sup>-1</sup>) and pH value of the fruit juice samples

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Some of the fluoride contents in different fruit juices that have been reported in the literature are also presented in Table 2.

Type of drink	Mean concentration (mg.L <sup>-1</sup> )	Range (mg.L <sup>-1</sup> )	Country	Reference
Peach juice	0.18	_		
Orange juice	0.46	_	Spain	33
Pineapplejuice	0.55	-		
Fruitjuice	0.197	0.09–0.25	India	32
Orana iulaa	0.75	0.00.4.00		
	0.75	0.02 1.39		
Appre juice	0.57	0.08-1.32	Mexico	31
Pineappie juice	0.53	0.13-1.18		
Orange juice	0.20	0.11–1.24		
Grape (white) juice	1.45	0.15–2.80		
Grape (red) juice	0.74	0.05–2.45		
Cherry iuice	0.74	0.12-1.71		
Appleiuice	0.54	0.03-2.64	USA	41
Orange juice	0.37	0.02–1.85		
Pineannle juice	0.16	0.03-0.88		
	0.10	0.00 0.00		
Children's juices	0.75	0.11–1.81	USA	43
Appleiuice	1 09	_		
Grane juice	0.33	_		44
Orange juice	0.53	_	004	
	0.07			
Orange juice	0.37	_	Turkov	40
Peach juice	0.16	-	TURKEY	42
Appleiuice	0.00			
Apple Juice	0.03	_		
Grape (Reu) juice	0.02	—		
Grape (vvnile) juice	0.00	—	17	45
Peach juice	0.07	-	Kuwalt	45
	0.08	-		
Orange juice	0.09	_		
Pineapple juice	0.09	_		
Coconut juice	3.401	2.68-4.16	Iran	46

Table 2. The fluoride content in different fruit juices (mg.L<sup>-1</sup>) found by other authors

In a study in the USA,<sup>41</sup> the fluoride concentration of fruit juice ranged from 0.02 to 2.80 mg.L<sup>-1</sup> with a mean value of 0.56 mg.L<sup>-1</sup>. In agreement with our study, the products containing white grape or red grape juice generally had a higher fluoride

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concentration than the other fruit juices. In another study in Spain,<sup>33</sup> the fluoride concentration level of natural fruit juices ranged from 0.06 to 1.14 mg.L<sup>-1</sup> with a mean value of 0.4 mg.L<sup>-1</sup>. The highest fluoride concentration were found in the grape  $(1.14 \text{ mg.L}^{-1})$  and guava  $(0.66 \text{ mg.L}^{-1})$  fruit juices while the lowest concentration of fluoride was recorded in the apricot  $(0.06 \text{ mg.L}^{-1})$  and carrot  $(0.07 \text{ mg.L}^{-1})$  fruit juices. Also, in a further study, the fluoride level of peach and orange fruit juice in Turkey<sup>42</sup> ranged from 0.05 to 0.5 mg.L<sup>-1</sup>.

As seen in Figure 1, there were some significant differences between the fluoride concentration levels of some examined brands.





Figure 1. Comparison of the mean fluoride concentration levels of different fruit juice brands.

The fluoride content of the H brand was significantly lower than all other fruit juice samples and the fluoride content of B brand was significantly higher than other fruit juices. Some of fruit juices examined had fluoride concentrations that were not significantly different from one another, for example as with the D and F brands (Figure 1).

The results also showed that there were some significant differences in the fluoride content of the fruit juices made with different fruits. The mean fluoride concentrations of all brands of the cherry, apple, and grape fruit juices were significantly higher than those for the pineapple, orange, and peach juices. (Figure 2).





Figure 2. Comparison of the mean fluoride concentration levels of the fruit juices made from different fruits.

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The order of fluoride content, from highest to lowest, in the different types of fruit juices were: cherry > grape > apple >pineapple > peach > orange. There were no significant differences between the fluoride contents of the cherry and grape juices, between the apple and grape juices, and between the pineapple, orange and peach juices. The fluoride contents of the different fruit juices in each brand (A-J) are presented in Figures 3A-3J.





**Figures 3A and 3B.** Comparison of the mean fluoride concentration levels in the fruit juices made from different types of fruit in brands A and B.



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Fruit juices in brand C



Figures 3C and 3D. Comparison of the mean fluoride concentration levels in the fruit juices made from different types of fruit in brands C and D.

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Fruit juices in brand E



**Figures 3E and 3F.** Comparison of the mean fluoride concentration levels in the fruit juices made from different types of fruit in brands E and F.

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Figures 3G and 3H. Comparison of the mean fluoride concentration levels in the fruit juices made from different types of fruit in brands G and H.

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As seen in Figures 3A–3J for brands A-J, there were some significant differences between the fluoride concentration levels of fruit juices made from different types of fruit in each brand. In brands D, E, F, and G there were more significant differences between the fluoride contents of the different types of fruit juice compared to the other brands examined. The fluoride content of the grape juice of brands A, E, F, and J was significantly higher than the juices made with other types of fruit while in the case of brands B and G, the cherry juice contained a significantly higher concentration level of fluoride compared to other fruit types. For brand C (Figure

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3C), the cherry and apple juices contained significantly higher content of fluoride compared to the other fruit types while the apple juice had the highest fluoride content between all fruit types of brand D (Figure 3D). Pearson's correlation coefficient showed that there is no statistically significant difference (r=0.01, p=0.904) between the pH and the F concentrations in the juice samples. In contrast to the present study, a study in India reported that there was no significant difference between the fluoride levels of different brands of fruit juice.<sup>32</sup> The variation in the fluoride content of fruit juice samples can be due to differences in the fluoride concentration in water supply used to manufacture the juice, the recipes used, the amount and type of ingredients, the storage time, and the type of cooking vessel used.<sup>31,47</sup>

The EDI values for the different age groups through the ingestion of different fruit juices are summarized in Table 3.

Brand		Concentration of F (mg.L <sup>-1</sup> )	EDI for various age groups (years) (µg.kg <sup>-1</sup> bw.day <sup>-1</sup> )				
			4–9 (yr)	10–17 (yr)	18–29 (yr)	30–49 (yr)	50–65 (yr)
А	Maximum	0.093	0.277	0.117	0.119	0.086	0.056
	Mean	0.03	0.094	0.039	0.04	0.029	0.019
P	Maximum	0.44	1.321	0.556	0.567	0.410	0.270
В	Mean	0.23	0.682	0.287	0.293	0.212	0.139
0	Maximum	0.134	0.399	0.168	0.171	0.124	0.081
C	Mean	0.050	0.149	0.063	0.064	0.046	0.030
П	Maximum	0.179	0.533	0.224	0.229	0.165	0.109
D	Mean	0.10	0.306	0.129	0.131	0.095	0.062
Е	Maximum	0.049	0.146	0.061	0.063	0.045	0.030
_	Mean	0.02	0.06	0.025	0.026	0.019	0.012
F	Maximum Mean	0.429	1.277 0.367	0.537	0.548	0.396	0.260
		0.12	0.007	0.100	0.100	0.114	0.075
G	Maximum Mean	0.158 0.07	0.470 0.208	0.198 0.088	0.202 0.089	0.146 0.065	0.096 0.043
	Maximum	0.011	0.033	0.014	0.014	0.010	0.007
Н	Mean	0.004	0.033	0.004	0.014	0.004	0.003
	Maximum	0.041	0 122	0.051	0 052	0.038	0 025
I	Mean	0.04	0.115	0.048	0.049	0.036	0.023
	Maximum	0.042	0.125	0.053	0.054	0.039	0.026
J	Mean	0.02	0.064	0.027	0.027	0.02	0.013
	Maximum	0.16	0.470	0 108	0 202	0146	0.006
Mean	Mean	0.10	0.470	0.196	0.202	0.140	0.090

 Table 3. Estimated daily intake (EDI) values for fluoride from fruit juice consumption

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The mean EDI values of the fruit juices, for age groups of 4–9, 10–17, 18–29, 30–49, and 50–65 years old, were 0.206, 0.087, 0.088, 0.64, and 0.042  $\mu$ g.kg bw<sup>-1</sup>.day<sup>-1</sup>, respectively. The highest EDI value was present in the age group of 4–9 years old (0.206  $\mu$ g.kg bw<sup>-1</sup>.day<sup>-1</sup>), while the lowest EDI value was found in the age group of 50–65 years old (0.042  $\mu$ g.kg bw<sup>-1</sup>.day<sup>-1</sup>). The lower EDI value in the age group of 50–65 years old, compared to other age groups, is because of a lower daily consumption of fruit juice. In a study in China, the EDI values for fluoride through fruit drink consumption were 0.015 mg.kg bw<sup>-1</sup>.day<sup>-1</sup> for 15 kg children and 0.005 mg.kg bw<sup>-1</sup>.day<sup>-1</sup> for 60 kg adults.<sup>34</sup>

The risk assessment of toxic chemicals is becoming a serious concern due to the effects toxic chemicals on human health. In order to calculate the non-carcinogenic risk to human health due to the fluoride intake through fruit juice consumption, HQ values for the aforementioned different age groups were determined and are presented in Table 4.

Brand			HQ for various age groups (years)					
		4–9 (yr)	4–9 10–17 18–29 30–49 50–65 (yr) (yr) (yr) (yr) (yr)					
A	Maximum	0.0046	0.0019	0.0020	0.0014	0.0009		
	Mean	0.0016	0.0007	0.0007	0.0005	0.0003		
В	Maximum	0.0220	0.0093	0.0095	0.0068	0.0045		
	Mean	0.0114	0.0048	0.0049	0.0035	0.0023		
С	Maximum	0.0066	0.0028	0.0029	0.0021	0.0014		
	Mean	0.0025	0.001	0.0011	0.0008	0.0005		
D	Maximum	0.0089	0.0037	0.0038	0.0028	0.0018		
	Mean	0.0051	0.0021	0.0022	0.0016	0.001		
Е	Maximum	0.0024	0.0010	0.0010	0.0008	0.0005		
	Mean	0.001	0.0004	0.0004	0.0003	0.0002		
F	Maximum	0.0213	0.0090	0.0091	0.0066	0.0043		
	Mean	0.0061	0.0026	0.0026	0.0019	0.0012		
G	Maximum	0.0078	0.0033	0.0034	0.0024	0.0016		
	Mean	0.0035	0.0015	0.0015	0.0011	0.0007		
н	Maximum	0.0005	0.0002	0.0002	0.0002	0.0001		
	Mean	0.0002	0.0001	0.0001	0.0001	0.0000		
I	Maximum	0.0020	0.0009	0.0009	0.0006	0.0004		
	Mean	0.0019	0.0008	0.0008	0.0006	0.0004		
J	Maximum	0.0021	0.0009	0.0009	0.0006	0.0004		
	Mean	0.0011	0.0004	0.0005	0.0003	0.0002		
Mean	Maximum	0.0078	0.0033	0.0034	0.0024	0.0016		
	Mean	0.0034	0.0014	0.0015	0.0011	0.0007		

Table 4. Calculated hazard quotient (HQ) of fluoride from fruit juice consumption

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The mean values of HQ for the different age groups, of 4–9, 10–17, 18–29, 30–49, and 50–65 years old, were 0.0034, 0.0014, 0.0015, 0.0012, and 0.0008, respectively. The non-carcinogenic risk was in the following order: 4–9 years > 18–29 years > 10–17 years > 30–49 years > 50–65 years. The HQ values for all the age groups were less than unity (<1), which indicates that the different age groups were not at a non-carcinogenic health risk due to the fluoride intake from fruit juice consumption.

### CONCLUSIONS

Fruit juice is a common source of fluoride and can increase the fluoride intake in humans, especially in tropical areas such as the Middle East region which has a high drink consumption. In this study, in 10 different brands of fruit juice with six different types of fruit, the range of the fluoride concentrations, was 0.00 to 0.444 mg.L<sup>-1</sup>. The calculated HQ values for all age groups, 4-9, 10-17, 18-29, 30-49, and 50-65 years old, were less than unity which indicates that none of the age groups were at a non-carcinogenic health risk due to the intake of fluoride from fruit juice consumption. Finally, the fluoride intake from other drinks containing a high level of fluoride should be considered when estimating the total daily fluoride intake from drinking, especially for those in warm regions with a high level of drink consumption.

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