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Health Risk Assessment for Fluoride Content in Public Water Supply and Bottled Waters: A Study of Harsin City, Kermanshah, Iran

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ABSTRACT

Purpose: Continuous monitoring of drinking water is essential to ensure quality and identify potential health risks. Fluoride levels in drinking water (both public and bottled) can be problematic and therefore require regular evaluation. This study aimed to measure fluoride concentrations in the distribution water supply and commonly consumed bottled water in Harsin city, Iran, and to assess the associated health risks.

Methods: Water samples were collected from the public distribution system at 10 locations across the city, as well as from 8 widely consumed bottled water brands. Fluoride concentrations were determined using a spectrophotometric method.

Results: The mean fluoride concentrations in bottled water and the distribution system were 0.56 mg/L and 0.43 mg/L, respectively. The hazard quotient (HQ) for fluoride exposure exceeded 1 for three bottled water brands in the infant group, indicating elevated risk. Monte Carlo simulation results showed that the 95th percentile of HQ values remained below 1 across all groups, suggesting no significant non-carcinogenic risk for 95% of the studied population. Sensitivity analysis identified fluoride concentration as the most influential parameter affecting HQ values in all exposure groups.

Conclusions: The findings indicate that infants are comparatively more vulnerable to potential health effects from fluoride in bottled water. Continuous monitoring of fluoride levels in drinking water sources is therefore recommended to safeguard public health and assess potential future risks.

Keywords: *Drinking water, health risk, fluoride, non-carcinogenic, Iran*

INTRODUCTION

Monitoring of drinking water quality is essential to prevent exposure to hazardous substances such as fluoride and to ensure the safety of water supplies. Fluoride occurs naturally in water and, in some countries, its concentration in drinking water has traditionally been adjusted for public health purposes. In addition to water, fluoride is also found in certain foods and beverages.¹⁻¹⁴ Among the various sources, drinking water remains the primary intake pathway of fluoride.¹⁵ Fluoride is considered as an essential element for human within the limits recommended by World Health Organization (WHO), namely 0.5–1.5 mg/L.⁷ However, concentrations above these limits can cause adverse health effects, including dental and skeletal fluorosis.^{16,17} Excessive intake has also been

associated with negative impacts on the brain, intelligence quotient (IQ), and blood pressure.^{18,19}

Because fluoride levels are strongly influenced by the geochemical characteristics of groundwater, varying concentrations and associated health outcomes have been reported worldwide.

In extreme cases, concentrations as high as 2,800 mg/L have been documented, leading to severe skeletal fluorosis.^{16,20-28} To address such risks, effective defluoridation techniques are required.^{29,30} Although naturally occurring fluoride contamination develops gradually and can be managed with appropriate interventions, bottled water presents unique concerns. In recent decades, bottled water consumption has risen sharply worldwide, largely due to its taste, clarity, convenience, and availability.³¹



Figure 1. Harsin City and sampling points

Global consumption increased by 7% in 2006, reaching a market volume of 89 billion liters.³² Notably, bottled water use grew by 15% in Asia and Pacific countries.³³ This rapid increase has raised concerns about bottled water quality, prompting studies focused on its biological, chemical, and microplastic content.³⁴⁻³⁶ For this, different works have been carried out for Biological, chemical, micro plastics determination in bottled water.³⁴⁻³⁹

Numerous studies have also examined fluoride concentrations in drinking water, reporting a wide range of results.^{3,34-37,40-46}

To evaluate potential health effects of chemical exposures, human health risk assessment (HRA) is commonly employed. HRA integrates scientific data to assess potential risks, providing critical insights for improving public health.^{15,47} It has been applied to various water pollutants, particularly to assess non-carcinogenic risks.^{36,37,41,43-45,48-50}

Given the lack of available data on fluoride concentrations in drinking water sources (both public supply and bottled water) in Harsin city and the potential associated health risks, this study was undertaken.

MATERIAL AND METHODS

Samples collection and study area

Harsin city, with a population of approximately 44,000, is located in the eastern part of Kermanshah Province (figure 1). The city drinking water supply is mainly derived from groundwater sources (wells). According to meteorological data, the region receives an average annual precipitation of about 496 mm, with

a mean annual temperature of 13.5 °C. For this study, water samples were collected from 10 locations across the city distribution system and from 8 commonly consumed bottled water brands. All samples were taken on three separate occasions and analyzed in triplicate to ensure accuracy and reliability of the results.

All water samples were coded and analyzed for fluoride concentration. The mean values ($\pm$ SD) were calculated and used for subsequent health risk assessment. Fluoride determination was performed using the colorimetric SPADNS method,⁵¹ which has been widely applied in previous studies.^{3,26,34,52-54}

The SPADNS method is suitable for fluoride concentrations in the range of 0–1.4 mg/L.⁵¹ Upon addition of the SPADNS reagent, color development occurs rapidly, and measurements are taken with a spectrophotometer at a wavelength of 570 nm. Fluoride concentrations in the samples were quantified using a calibration curve prepared from standard fluoride solutions.⁵¹

Health Risk assessment

Human health risk assessment (HRA) is a suitable approach for evaluating potential risks associated with various elements in drinking water, providing scientific evidence to safeguard consumers. In Iran, increasing public awareness has heightened concern about the possible health effects of fluoride across different population groups. In this study, HRA was applied to assess fluoride-related risks for distinct demographic groups.^{34,55,56} For conducting the health risk assessment (HRA), key parameters such as the Estimated Daily Intake (EDI) of fluoride and the Hazard Quotient (HQ) were calculated using Equations (1) and (2).^{36,37,55-57}

$$HQ = \frac{EDI}{RfD} \quad (1)$$

$$EDI = \frac{C \times IR \times EF \times ED}{BW \times AT} \quad (2)$$

Table 1. Fluoride levels and the labeled value in different bottled waters

Symbol	Type	Fluoride		
		Labeled value (mg/L) ***	Mean Measured (mg/L)	±SD
S1	BW*	0.5	0.81	±0.10
S2	BW	>1	1.2	±0.01
S3	BW	0.2	0.71	±0.01
S4	BW	0.41	0.29	±0.01
S5	BW	0.14	0.45	±0.01
S6	BW	0.5	0.49	±0.03
S7	BW	0.09	0.20	±0.00
S8	BW	0.25	0.35	±0.01
DW	DW**	-	0.43	±0.03

* Bottled Water, **Distribution Water, *** Fluoride concentration reported by company on the bottled

From the equation, C represents the fluoride concentration (mg/L), IR represents the ingestion rate for HQ calculation for different groups (Infants <2 years old, children 2<and <6 years old, teenagers 6<and <16 years old and adults >16 years old), EF defines exposure frequency (365 days/year) for all groups, ED indicates exposure duration (year), 1.5, 4, 13, and 40 years for infants, Children, teenagers and adults, respectively.³⁴ BW is body weight of the groups in kg, for infants (7.5 kg), Children (15 kg), teenagers (50 kg), and adults (72 kg). AT indicate of average exposure time for the groups during life time, multiply of EF×ED.³⁴ The reference dose for fluoride (RfD-F) was set at 0.06 mg/kg-day for all population groups.⁵⁸

Monte Carlo Simulation

Risk assessment involves inherent uncertainties that must be taken into account to ensure reliable outcomes and to guide appropriate risk management actions.^{58,59} Uncertainty is an integral component of Health Risk Assessment, and addressing it is essential for making well-informed decisions and effectively managing potential outcomes. In this context, uncertainty refers to the lack of precise knowledge regarding specific variables, parameters, or models.⁶⁰ Monte Carlo Simulation (MCS) is widely applied to reduce such uncertainties by generating numerous random iterations to represent variability in system input variables.^{34,35} Monte Carlo Simulation (MCS) operates by generating a large number of random repetitions to represent the uncertainty associated with various input variables of a system. A mathematical model is then applied to each repetition to calculate potential outcomes. By performing this process repeatedly, a distribution is obtained that characterizes the uncertainty of possible outputs⁵⁹. In this study, fluoride concentration (C, lognormal distribution), exposure duration (ED), body weight (BW, normal distribution), and intake rate (IR, lognormal

distribution) were used as input variables for the simulation.^{34,61} A total of 10,000 iterations were conducted within a confidence interval of 1–99%.⁶¹

Sensitivity Analysis

In general, sensitivity refers to the degree of variation in a model's output resulting from changes in its input values. Sensitivity analysis (SA) is therefore used to evaluate the extent to which model input assumptions contribute to variability or uncertainty in the output.^{58–60} In this study, Crystal Ball software (version 11.1.2.4, Oracle, Inc., USA) was employed to perform both Monte Carlo Simulation (MCS) and SA using 10,000 trials.^{56,61,62} The variables considered for SA were the same as those used in the MCS and are presented in Eq. 2, consistent with previous studies.^{34,56,62,63}

RESULTS

Fluoride concentrations in the analyzed samples are presented in table 1 and figure 2. As previously noted, S1 to S8 represent bottled water samples, while DW refers to the average value for the city public distribution water. According to table 1 and figure 2, fluoride concentrations in bottled waters ranged from 0.20 ± 0.01 mg/L to 1.07 ± 0.06 mg/L, whereas the concentration in public water was 0.43 ± 0.03 mg/L. The overall mean concentration across all samples was 0.55 mg/L. Among the bottled waters, the highest fluoride level was observed in sample S2.

Health risk assessment

Health risk assessment (HRA) for the consumers was performed using HQ.³⁴ In this study, the estimated daily intake (EDI) of fluoride was calculated for different age groups, including infants, children, teenagers, and adults. The resulting EDI values were 0.05, 0.03, 0.02, and 0.02 mg/kg-day, respectively, for these groups.

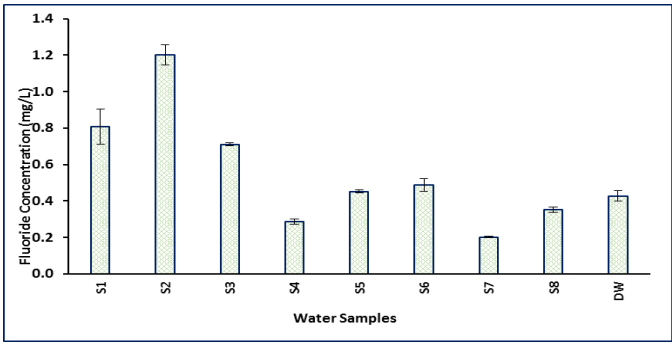


Figure 2. Fluoride concentration in different water sources

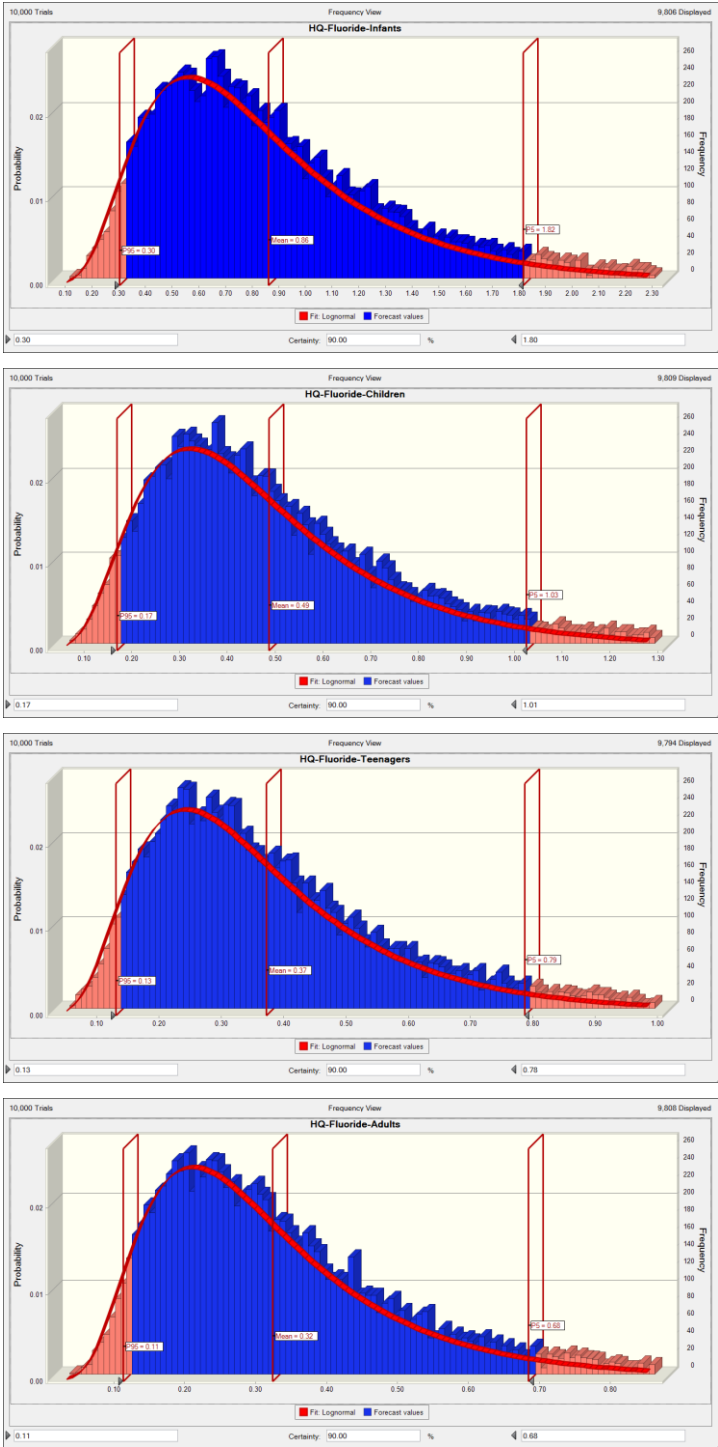


Figure 3. HQ value related to fluoride contents for different groups



Figure 4. Sensitivity analysis of fluoride exposure for different groups

Monte Carlo simulation and sensitivity analysis

Figure 3 depicts the results for MCS. From figure 3, 95th percentile in infants, children, teenagers, and adults were 0.30, 0.17, 0.13, and 0.11, respectively.

Sensitivity analysis

Sensitivity analysis (SA) of the Hazard Quotient (HQ) for different age groups is presented in Figure 4. Among the influencing parameters—drinking water intake rate (IR), fluoride concentration (C), and exposure frequency (EF)—fluoride concentration exhibited the greatest impact across all groups. Specifically, the contribution of fluoride concentration to HQ variability was 93.7%, 93.4%, 93.6%, and 94.3% for infants, children, teenagers, and adults, respectively.

DISCUSSION

Fluoride content in the samples

From the results (table 1 and figure 2), fluoride values of the analyzed bottled waters (S1 to S8) represents a maximum value of 1.07 mg/L. This value is at the range of recommend guideline (0.5–1.5 mg/L).⁷ Fluoride contents of bottled water are consistent with other reports.^{34,38,39,42,43,62,64,65} In comparison to the labeled values (table 1), the measured values are not match and mainly higher than labels. Such an inconsistency have been reveled and reported widely.^{3,34,62} From the consumers point of view, labeled

value is the available tool for water safety insurance. This study and other works revealed that consumers cannot rely only on the labels. Although the values are mainly in the standard range.^{3,34,62}

From the findings, the mechanism of labeling and continuous quality control is of concern. Differences between the values on the label and the values measured in laboratories may be due to inaccuracy in labeling, slight changes in the composition of the water after bottling, or non-compliance with scientific standard methods; although the values measured in research laboratories are more accurate and reliable.

From the results (table1 and figure 2) the average of analyzed sampled distribution water (DW) was 0.43 mg/L. Fluoride content of drinking water of the city is lower than some parts of Iran. In Sothern parts of the country such as Bushehr the fluoride of drinking water are as high as 3.64 mg/L.⁵² Although the average fluoride concentration of bottled waters was slightly higher than that of public water, statistical analysis using the Mann–Whitney test indicated no significant difference between the two groups ($P > 0.05$). Fluoride concentration in ground waters are mainly higher than surface waters.⁵² In Poldasht, in the northeast of Iran, fluoride concentration as high as 10.3 mg/L has been reported, along with signs of dental fluorosis.¹⁷

Thought Harsin city is supplied from ground waters, fluoride content in distribution system is at standard range.

Table 2. HQ Fluoride for different age groups

Sample code	HQ-Fluoride			
	Infants	Children	Teenage	Adults
S1	1.26	0.70	0.54	0.47
S2	1.87	1.04	0.80	0.70
S3	1.11	0.62	0.47	0.41
S4	0.45	0.25	0.19	0.17
S5	0.70	0.39	0.30	0.26
S6	0.76	0.42	0.33	0.28
S7	0.31	0.17	0.13	0.12
S8	0.55	0.30	0.23	0.20
DW	0.66	0.37	0.28	0.25

Health risk assessment

Health risk assessment (HRA) for the consumers was performed via HQ³⁴. In this work, the daily exposure (EDI) of fluoride was calculated for the mentioned groups (infants, children, teenagers and adults). From the results, the EDI values for the group categories namely infants, children, teenagers and adults were 0.05, 0.03, 0.02, and 0.02 mg/kg.day, respectively.

As revealed, infants and children showed comparatively higher EDI values. Consequently, these groups are likely at a higher risk compared to teenagers and adults. Similar findings have also been reported in previous studies.^{34,66}

It has been proposed that fluoride exposure in children exceeding the reference dose (RfD) of 0.06 mg/kg.bw.day may lead to dental problems, particularly dental fluorosis.⁵⁸ In this study, three bottled water brands (S1, S2, and S3) were found to have EDI values higher than the RfD for infants, with brand S2 showing the highest value (0.11 mg/kg.day).

The hazard quotient (HQ) for fluoride exposure was calculated across four age groups. As shown in Table 2, HQ values for three bottled water brands (S1, S2, and S3) exceeded 1 in infants, while S2 also exceeded 1 in children. This indicates that these groups may be exposed to elevated non-carcinogenic risks. For the remaining samples, HQ values were below 1, suggesting no significant health concerns.

When considering the average fluoride content across all water sources, the HQ values ranked as follows: infants (0.86) > children (0.48) > teenagers (0.37) > adults (0.32). These results demonstrate that infants and children are at greater health risk from fluoride ingestion via drinking water. Their higher susceptibility is mainly due to lower body weight and higher intake relative to body size.^{34,36,67,68}

In this study, dental fluorosis was not identified as a health concern based on the HQ values for the target groups. It should be noted, however, that these findings are limited to fluoride intake from drinking water (both bottled and distributed). In reality, fluoride exposure can also occur through other sources such as foods, beverages, tea, meat, and dietary supplements, which may substantially increase the overall health risk.^{2,4}

Monte Carlo simulation and sensitivity analysis

In addition to the point estimation of health risk assessment (HRA) in terms of HQ (calculated using Equations (1) and (2)), Monte Carlo simulation (MCS) was applied to estimate uncertainties.⁶⁹ In this study, MCS with 10,000 trials was performed using Oracle Crystal Ball software to evaluate the variance of HQ values. A probabilistic approach was applied to assess fluoride exposure across the four age groups. The distributions considered in the simulation included lognormal for fluoride concentration and ingestion rate (IR), and normal for body weight (BW), as reported for infants, children, teenagers, and adults.³⁴ Figure 3 depicts the regarding results. As shown in Figure 3, the 95th percentile HQ values for infants, children, teenagers, and adults were 0.30, 0.17, 0.13, and 0.11, respectively. Although these values are all below 1, indicating no significant non-carcinogenic risk, the comparatively higher value in infants suggests that this group may be more vulnerable to fluoride-related health risks. This increased susceptibility is likely due to their lower body weight compared with the other three groups.^{34,56,70}

Sensitivity analysis

Sensitivity analysis (SA) was conducted to evaluate the contribution of different variables to the hazard quotient (HQ) across the four age groups^{36,67}. The SA results are presented in Figure 4. Among the influencing parameters—drinking water intake rate

(IR), fluoride concentration (C), and exposure frequency (EF)—fluoride concentration (C-F) had the greatest impact in all groups. The effect proportions of IR were 93.7%, 93.4%, 93.6%, and 94.3% for infants, children, teenagers, and adults, respectively, indicating its substantial influence on risk estimates for all exposed groups. Similar findings regarding the dominant role of fluoride concentration have been reported in previous studies.^{34,56,62}

In addition, body weight (BW) was identified as the second most influential variable for children, accounting for 48.4% of the effect after fluoride concentration. For the other groups, body weight (BW) and intake rate (IR) showed a much lower influence on exposure risk. In contrast, previous studies have reported a more significant role of IR compared to other variables.^{36,61} Overall, since fluoride concentration emerged as the most critical factor in the sensitivity analysis, controlling its level through regular monitoring or removal methods can effectively reduce the associated health risks.

CONCLUSIONS

Fluoride levels in both bottled and municipal distribution water in Harsin city were measured to assess potential non-carcinogenic health risks. Fluoride concentrations were analyzed in eight brands of bottled water and at ten locations throughout the city distribution network. The average fluoride concentrations in the collected bottled and distribution water showed no statistically significant differences. However, discrepancies were observed between the labeled fluoride values on the bottled water and the measured concentrations. Despite these differences, all samples were within the guideline values recommended by the WHO and the Iranian national standard.

From a risk perspective, infants were found to be comparatively more vulnerable to fluoride exposure from both bottled water and the distribution system. The hazard quotient (HQ) values for three bottled water brands exceeded 1 for the infant group. Monte Carlo simulations (MCS) indicated that the 95th percentile of fluoride exposure was below 1 for all age groups, suggesting a low non-carcinogenic risk for 95% of the studied population. Sensitivity analysis (SA) further revealed that fluoride concentration had the greatest influence on HQ compared to other variables across all age groups.

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DISCLOSURE OF FINANCIAL AND NON-FINANCIAL RELATIONSHIPS AND ACTIVITIES AND CONFLICTS OF INTEREST

There is no conflict of interest regarding to this article.

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