Concentration, distribution and probabilistic health risk assessment of exposure to fluoride in drinking water of Hormozgan Province, Iran

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Research Article

Keywords: Fluoride, Health risk assessment, Drinking water, Monte-Carlo Simulation, Hormozgan

Posted Date: May 7th, 2021

DOI: https://doi.org/10.21203/rs.3.rs-484346/v1

Version of Record: A version of this preprint was published at Stochastic Environmental Research and Risk Assessment on September 15th, 2021. See the published version at https://doi.org/10.1007/s00477-021-02090-1.

Abstract

In the present study, the health risk assessment of exposure to fluoride in drinking water of southern Iran province was performed with a definite and probabilistic approach. Monte Carlo simulation and sensitivity analysis were used for uncertainty in risk estimation. The results showed that fluoride concentration ranged between 0.1–0.9 mg/l with an average of 0.454 ± 0.205 mg/l. The distribution function followed the normal distribution. The highest fluoride concentration was observed in the central and northern regions of the province. Also, the lowest concentration was obtained 0.01 mg/l. In the definitive method, the non-carcinogenic risks associated with fluoride in drinking water were in acceptable range (HQ < 1). Conducting Monte Carlo simulation indicated that the potential non-carcinogenic risk for children in the 95th percentile exceeded the safe limit of 1, which showed that there were a potential non-carcinogenic in this group. Sensitivity analysis showed that fluoride concentration and water consumption had the most considerable effect in the model. Therefore, consumption of water and foods containing fluoride along with excessive consumption of tea may increase human's health risks. Therefore, continuous monitoring of water sources in terms of fluoride concentration along with proper diet training for residents of this province should be done.

Introduction

With increasing public concern about water resources and the environment, the importance of groundwater and surface water conservation as a single resource has become apparent. Issues related to water supply, water quality, and the destruction of aquatic environments have become one of the most attractive issues of the day [1]. Important pollutants in drinking water sources include heavy metals, pesticides, nitrate, fluoride, etc. [2, 3]. Small amounts of fluoride are naturally present in water, air, soil, and living organisms. As a result, humans are exposed to fluoride through food consumption, drinking water, skin contact, and respiration [4]. Fluoride concentration in natural waters differs from 0.1–10 mg/l [5]. Different factors such as geological settings, chemical characteristics of groundwater, porosity of aquifer materials, pH, climate condition and well depth affect fluoride concentration [6]. Various regions in China, India, Pakistan, Iran, Mexico, etc. are involved in the regions with high concentrations of fluoride in groundwater [7, 8].

According to World Health Organization (WHO) guidelines, the maximum concentration level (MCL) of fluoride in drinking water is 1.5 mg/L [9]. Nowadays, the problems of exposure to high fluoride concentrations, especially in children, have caused numerous health problems. Fluoride helps maintain strong bones, but during intake of high contents of fluoride, it can cause discoloration and tooth decay, osteoporosis, and damage to the kidneys, nerves, and muscles [10–12].

One way to assess the potential effects of pollutants on human health is health risk assessment. In the deterministic method of health risk assessment, the risk is usually based on the average value of various parameters. However, if the data standard deviation is large, it may underestimate or overestimate the actual risk. Therefore, the use of probabilistic approaches such as Monte Carlo simulation provides a more accurate risk assessment [13].

Recent studies have reported high levels of fluoride in some parts of Iran and low levels in others. Mirzabeigi et al. performed health risk assessment of fluoride in groundwater resources of 28 villages of Ardakan city, Yazd province. The results showed that the average concentration of fluoride was 2.92 mg/l (range: 0.9-6 mg/l). Also, in half of the villages, F concentration was higher than the standard recommended by the WHO (1.5 mg/l) [14]. Also, Ghaderpour et al. estimated the non-carcinogenic health risk of fluoride in urban and rural areas of Khorasan Razavi province showed that the average fluoride levels in urban and rural areas were 0.74 and 0.59 mg/l, respectively, which were lower than WHO recommended value [15]. These observations have raised concerns about the level of fluoride in drinking water [16]. Therefore, the concentration of fluoride in the water of different regions is one of the important requirements to prevent its health effects. In this study, the concentration and distribution of fluoride in drinking water resources of Hormozgan province were evaluated. Besides, Monte Carlo simulations and sensitivity analysis were applied to estimate the potential health risk of fluoride and quantify the risk-related uncertainty, respectively.

Materials And Method

Study area

Hormozgan province is one of the southern provinces of Iran with an area of 70,000 Km^2 and a population of 1.7 million. Hormozgan province is located between the geographical coordinates of 25° 24' to 28° 57' N latitudes and 53° 41' to 59° 15' E longitudes (Fig. 1). The province includes 11 cities of Jask, Bashagard, Sirik, Rudan, Minab, Bandar Abbas, Hajiabad, Khamir, Bastak, Bandar Lengeh and Parsian. According to the De Martone climate classification, the climate of the study area is very hot and humid [17, 18]. The average rainfall, temperature, and humidity are 215.8 mm, 27 °C, and 19-100%, respectively. This province has a complex geological structure and structurally can be divided into Zagros, Makran, and Central Iran zones [19, 20].

Sample collection and analysis

In 2019, 56 drinking water samples, including tap water, well water, and springs, were collected from 11 cities of Hormozgan province (Fig. 1). Pre-rinsed low-density polyethylene bottles were used for sampling. Firstly, they were soaked in nitric acid solution (20%) for 24 hours and then washed with tap water and distilled water. Before sampling, the bottles were washed three times with sample water. The samples were then stored at 4 °C until chemical analysis. The collected water samples were transferred to the chemistry laboratory for fluoride analysis using the SPADNS colorimetric method. After the reaction of fluoride with zirconium, the produced color in the solution was measured by a UV visible spectrophotometer at a maximal absorption wavelength of 570 nm [21].

Exposure and health risk assessment

Health risk assessment is a method to assess the potential harmful effects on human health from exposure to specific chemical agents for a specific period [22]. This method has been widely used and validated in various cases of health risk estimation in different environments [23]. The optimal fluoride concentration is assigned by the WHO for temperate regions and is unsuitable for tropical, arid, and semi-arid regions with higher air temperatures because it is created as a function of the average temperature of 16 °C [24]. The optimal level of fluoride concentration in drinking water is calculated based on the average annual maximal temperature (Tm) and varies from 0.5 to 1.5 mg/L depending on the temperature and climate of the region.

The regional optimal concentration of fluoride in drinking water was calculated by Eq. (1) [24]:

$$D(mg/l) = \frac{0.34}{-0.038 + (0.0062 \times T_m)} \qquad \text{Eq. (1)}$$

Where D is the optimal dose of fluoride in drinking water (mg/L) and Tm is the average annual maximal daily temperature for the last 5 years (°F). Due to the existence of different regions and climatic conditions in Iran, the temperature range across the country may vary from -20 °C to +50 °C. The average calculated Tm in Hormozgan province is 25.2 °C [25].

People may expose to fluoride through oral consumption (drinking water), skin absorption, and inhalation. However, the routes of inhalation and skin contact have not been included in this study due to lack of toxicological data, such as inhalation reference dose for fluoride, water-to-air transfer efficiency, and low skin exposure. The intake dose of fluoride through water consumption was estimated for three age groups (C:children; T: teens; A: adults) according to the USEPA method, according to Eq. 2 [26]:

$$EDI_{ing} = \frac{c_w \times IR_w}{BW} \qquad \text{Eq. (2)}$$

Also, HQ, which represents the non-carcinogenicity risk through different exposure pathways, was calculated using estimated daily intake (EDI) and oral reference dose (RfD) (Eq. 3) [27]:

$$HQ = \frac{CDI}{RfD} \qquad \text{Eq. (3)}$$

A HQ value of >1 implies a significant risk level where can affects the teeth and bones and or can lead to potentially skeletal problems.

Table 1. Input parameters for risk assessment.

Parameters	Units	Distribution type and values[a]	Reference
Fluoride concentration	*mg/l*	-	
Ingestion rate (IR_w)	L/day	C: N [1.25, 0.57]	[28]
		T: N [1.58, 0.69]	
		A: N [1.95, 0.64]	
Body weight (BW)	kg	C: LN [20, 1.48]	[29]
		T: LN [46.25, 1.18]	
		A: LN [70, 1.10]	
Oral reference dose (RfDo)	mg/kg/day	P [0.06]	[30]

[a] *C: children; T: teens; A: adults (N = normal [arithmetic mean, standard deviation]; LN = lognormal [geometric mean, geometric standard deviation]; P = point [fixed value]).*

Monte Carlo simulations and sensitivity analyses

Monte Carlo simulations investigate uncertainties in a probabilistic approach constructing possible resulted models by substituting a wide ranges of values for the probability distribution in each parameter [31]. During the simulation paths, the values of the selected parameters are randomly sampled based on the input probability distribution. The input probability distribution can take many forms, including uniform, normal, and triangular.

The output of the simulation process is a function of the model output distribution. One of the main advantages of Monte Carlo simulation is the ability to reflect the overall uncertainty of input variables in risk assessment models. Sensitivity analysis, which is another part of Monte Carlo simulation, allows the researcher to identify the parameter that has the greatest impact on risk assessment [32]. In this study, R6.32 software and mc2d Package were used for simulation and sensitivity analysis with 10,000 replications.

All data are expressed as triplicate with standard error. The Fitdistrplus package was used to find the best fluoride concentration distribution in drinking water. Analysis of variance (ANOVA) was used by Tukey's multiple groups to determine significant differences between water supply sources.

Results And Discussion

Fluoride concentration

The distribution map and Box plot of fluoride concentration in drinking water in Hormozgan province are presented in Figs 1 and 2.

The mean level of fluoride in drinking water was 0.454 ± 0.205 mg/l and the range of changes was 0.01-0.9 mg/l. The highest fluoride concentration was observed in the central and northern regions of the province, namely the Hassan Langi region in Bandar Abbas county, and the Sargaz Ahmadi region in

Hajiabad country (0.9 mg/l). Also, the lowest concentration was obtained in Rudan-Posht Banan with a concentration of 0.01 mg/l. Based on Kruskal-Wallis analysis, there was a significant difference between fluoride concentrations in different sampling points in Hormozgan province (P-value = 0.046).

The findings of this study are similar to the results conducted in Mashhad, Bardaskan and East Azerbaijan [33-35]. However, a conducted study by Aslani et al. in rural areas of Maku -Poldasht showed that the fluoride concentration in drinking water in these areas was higher than WHO's recommended value [36]. Generally, in the most cases in Iran fluoride in groundwater and drinking water resources has a geogenic sources related to geological and geochemical characteristics of studied regions [37].

Using the probability distribution input data, various parameters in the model are estimated, and then based on this probability distribution, the probable risk assessment is performed [38]. The density plots along with the empirical distribution function related to fluoride concentration is shown in Fig. 4. The Fitdistrplus package in R software was used to adapt the fluoride data probability distributions. Fig. 4 shows the degree of proximity of experimental data to a variety of known probability distribution functions. Different distribution functions are labeled with their specific mark. In this condition, the more observation point (blue dot) was closer to any mark (sign), the more probable that the data show that theorical distribution. As can be seen, the Cullen and Frey diagram predicts that the data distribution was close to the normal distribution function. Moreover, the theoretical distributions and diagrams of the cumulative distribution function on the actual data are plotted in Fig. 5. It is indicated that Weibull, and Gamma functions represented the best distributions to describe fluoride concentration data in drinking water.

The statistical results summary of each used distribution function is presented in Table 2. One of the evaluation criteria of statistical models is the Akaike Information Criterion (AIC), which is calculated in terms of Likelihood Function [39]. Usually, the model with the lowest AIC and BIC is selected as the best model. The AIC coefficient is ordinarily positive, but what is important is the difference between the two AIC values (or better AICc), which indicates the suitability of the two models [40]. According to Goodness-of-fit statistics and Kolmogorov-Smirnov and Cramer-von Mises statistical tests, the best distribution for the data were the Weibull distribution, but the AIC and BIC coefficients prefer the normal distribution. Under these conditions, the p-value for the K-S test separately for the normal and Weibull distributions was 0.936 and 0.967, respectively. Therefore, both distributions were suitable for this data. because the normal distribution is simply a distribution with a certain shape, it will be the basis of subsequent calculations.

Table 2. Results of maximal likelihood parameter estimation for normal, Weibull and beta probability distribution

	Normal	Distribution functions weibull	beta	Estimated standard error
Parameters				
Shape1			1.984	**0.354**
shape2			2.402	**0.438**
shape		2.271		**0.243**
scale		0.509		**0.031**
Mean	0.454			**0.0273**
SD	0.205			**0.019**
Performance criteria				
AIC	-14.656	-12.786	-14.254	
BIC	-10.606	-8.735	-10.204	
Goodness-of-fit statistics				
Kolmogorov-Smirnov statistic	0.072	0.066	0.082	
Cramer-von Mises statistic	0.041	0.034	0.067	
Anderson-Darling statistic	0.266	0.293	0.459	

Hierarchical clustering is one of the most widely used clustering methods. In this method, the distance between the two observations is calculated first. After determining the distance between the two observations, due to the proximity of the observations to each other, the observations together form a new cluster. This goes so far that all observations are in one cluster. In Fig. 6, fluoride concentrations at different sampling points were categorized using hierarchical clustering based on Euclidean distance method. The results showed that of the samples in Sargaz Ahmadi and Hassan Langi regions, which had the most similarities in terms of fluoride concentration, were merged, and in the later stages, the classification was completed.

Calculation of fluoride optimal concentration

According to Galagan and Vermillion's formula, the calculated regional optimal concentration of fluoride in drinking water of Hormozgan province is 0.77 mg/l, while the maximum standard fluoride concentration was considered by EPA is 1.5 mg/l. Accordingly, the fluoride concentration in all sampling points was lower than the calculated optimal concentration. In the study of Zozoli et al., the optimal concentration of fluoride in all Iranian provinces was determined in the range of 0.64-1.04 mg/l, but in Alborz, Khuzestan, and Hormozgan provinces, the fluoride concentration was less than acceptable [25]. In another study in one of the cities of Hormozgan, there was a direct relationship between fluoride concentration and DMFT index and with increasing fluoride concentration, DMFT index also increased [41].

Since fluoride enters the body through routes other than water, such as food and beverages, especially tea, and skin absorption, the lack of fluoride concentration in drinking water will probably be compensated. drinking water will probably be compensated. However, failure to provide the fluoride needed by the body can lead to problems such as tooth decay [42, 43].

Human health risk estimation

EDI and HQ related to fluoride in drinking water in different cities of Hormozgan province for three age groups (children, teenagers, and adults) are presented in Table 3.

Table 3. Calculation of EDI and HQ of fluoride for different age groups

NO.	City	EDI			HQ		
		Children	Teenagers	Adults	Children	Teenagers	Adults
1	Jask	0.035	0.019	0.015	0.58	0.32	0.26
2	Bashagard	0.018	0.010	0.008	0.30	0.17	0.13
3	Minab	0.026	0.014	0.011	0.43	0.23	0.19
4	Sirik	0.029	0.016	0.013	0.48	0.26	0.21
5	Rudan	0.021	0.012	0.009	0.35	0.19	0.16
6	Bandar Abbas	0.041	0.023	0.018	0.69	0.38	0.31
7	Hajjiabad	0.033	0.018	0.015	0.55	0.30	0.25
8	Khamir	0.024	0.013	0.011	0.40	0.22	0.18
9	Bastak	0.016	0.009	0.007	0.26	0.14	0.12
10	Parsian	0.035	0.019	0.016	0.58	0.32	0.26
11	Bandar Lengeh	0.040	0.022	0.018	0.67	0.36	0.30

The highest EDI rate belonged to Bandar Abbas in the children group (0.041) and the lowest EDI rate belonged to Bastak in the adult group (0.007). Also, the average HQ for children, teenagers, and adults in different cities of Hormozgan province were 0.48, 0.26, and 0.21, respectively, which indicated that children were the most sensitive group to fluoride exposure through drinking water. The HQ for different cities of Hormozgan province in different age groups is given in Table 3. Because the HQ obtained in all groups was <1 for the cities of Hormozgan province, so there was no non-carcinogenic risk of fluoride in drinking water for the residents of this province. The results also showed that the health risks of fluoride in the children group were 1.8 and 2.2 times higher than in the teenager and adult groups. In the study of Ghaderpoori et al. in the Mashhad drinking water network, the calculate HQ of fluoride for women, men, and children were < 1 [33]. Nevertheless, in the study of Yousefi et al., the HQ in the drinking water of Showt city, Iran for children, teenagers, and adults had health hazards in 54.55%, 31.82%, and 22.73% of samples respectively (HQ> 1). This was due to the presence of high fluoride concentration in drinking water, which has caused endemic dental fluorosis in the inhabitants of this area [44].

Uncertainty analysis

In the present study, Monte Carlo simulation were used to determine the quantity and uncertainty in the fluoride risk assessment process according to the input parameters.

The histogram corresponding to HQ in different groups is shown in Fig. 8. The HQ's results showed that the confidence interval in the children group was 89.4%. Therefore, there is a risk of dental fluorosis in them, but in teenagers and adults, no particular problem has been observed due to increased fluoride concentration. In studies conducted in Iran, China and India, children were identified as the most sensitive group to fluoride exposure, which was determined by the higher ratio of water consumption with respect to body weight [45-47]. Sensitivity analysis was used to determine the most effective factor in HQ for different groups. Tornado charts showed the median estimates of the spearman's rank correlation between the input variables and the non-carcinogenic effects for three groups that consumption of drinking water. As shown in Fig. *8*, the concentration of fluoride in drinking water was the most effective factor in the non-carcinogenic effects of drinking water in Hormozgan province. The rate of water consumption has a positive effect on indicating higher level of health risk with increasing water consumption. However, body weigh showed negative effect since the amount of HQ decreased with increasing body weight. The results of this analysis were consistent with studies performed on drinking water fluoride in Bangladesh [48].

Conclusion

This study investigated the distribution of fluoride in drinking water and its health risk assessment in Hormozgan province located in southern Iran. The average fluoride concentration in this province was less than the amount recommended by the EPA and the calculated optimal dose. The results of health risk assessment in this study showed that exposure to fluoride through water consumption in all groups was within acceptable limits. The calculated HQ was < 1, but the health risk in the probabilistic method in children was 6.91% > acceptable range and the order was as HQchildren > HQteens > HQadults. However, because the consumption of foods, tea, and the use of fluoride-containing toothpaste was not considered in this study, the amount of fluoride intake may be higher and may cause problems in the long period. Hence, a regular monitoring program of the water source and proper education about the proper diet for the residents of this province should be done.

Declarations

Funding: This study was supported by Shiraz University of medical sciences.

Ethical approval: Not applicable

Competing interests: The authors declare no competing interests.

Consent to participate: Not applicable

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Figures

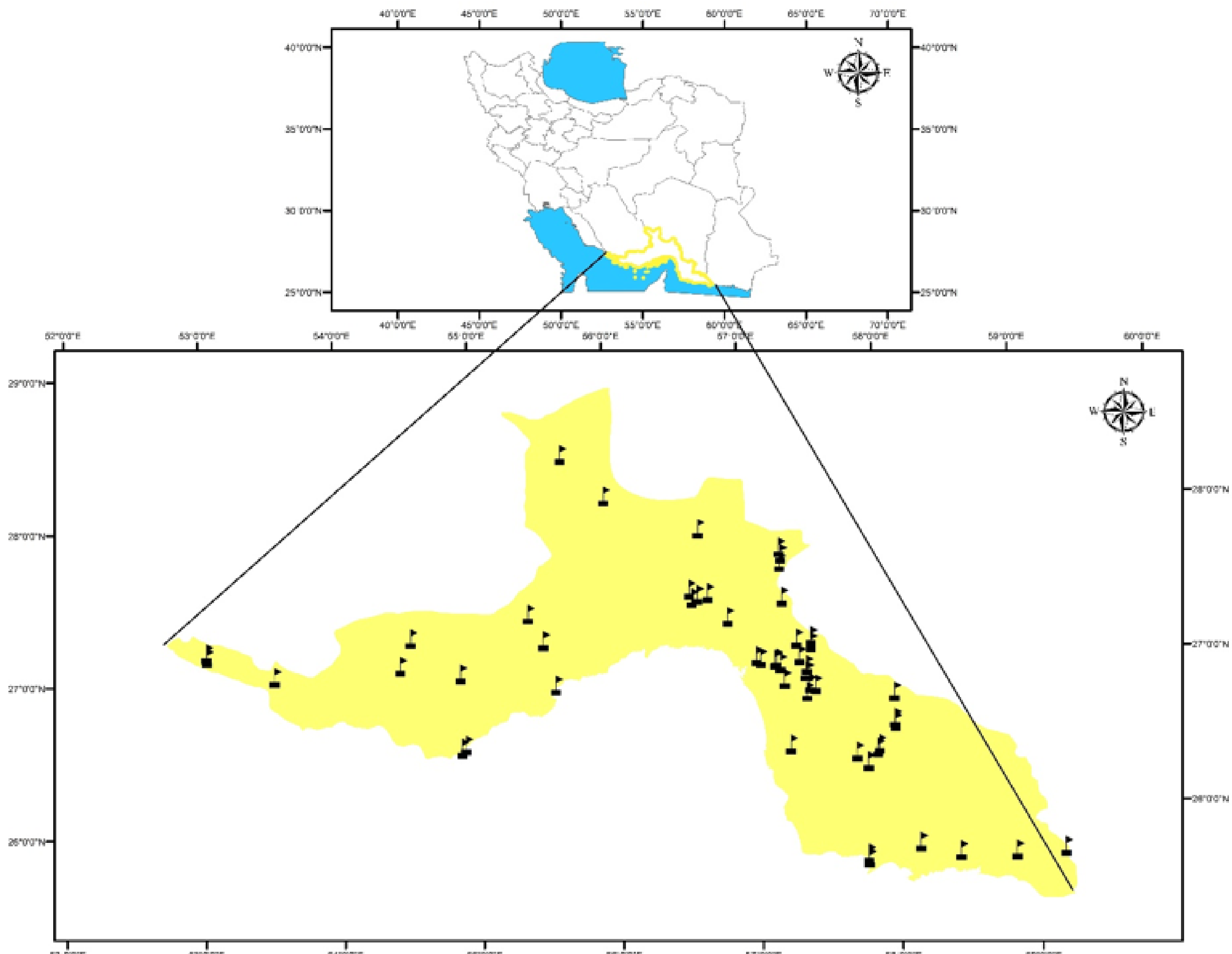

Figure 1

Geographical position of the study area

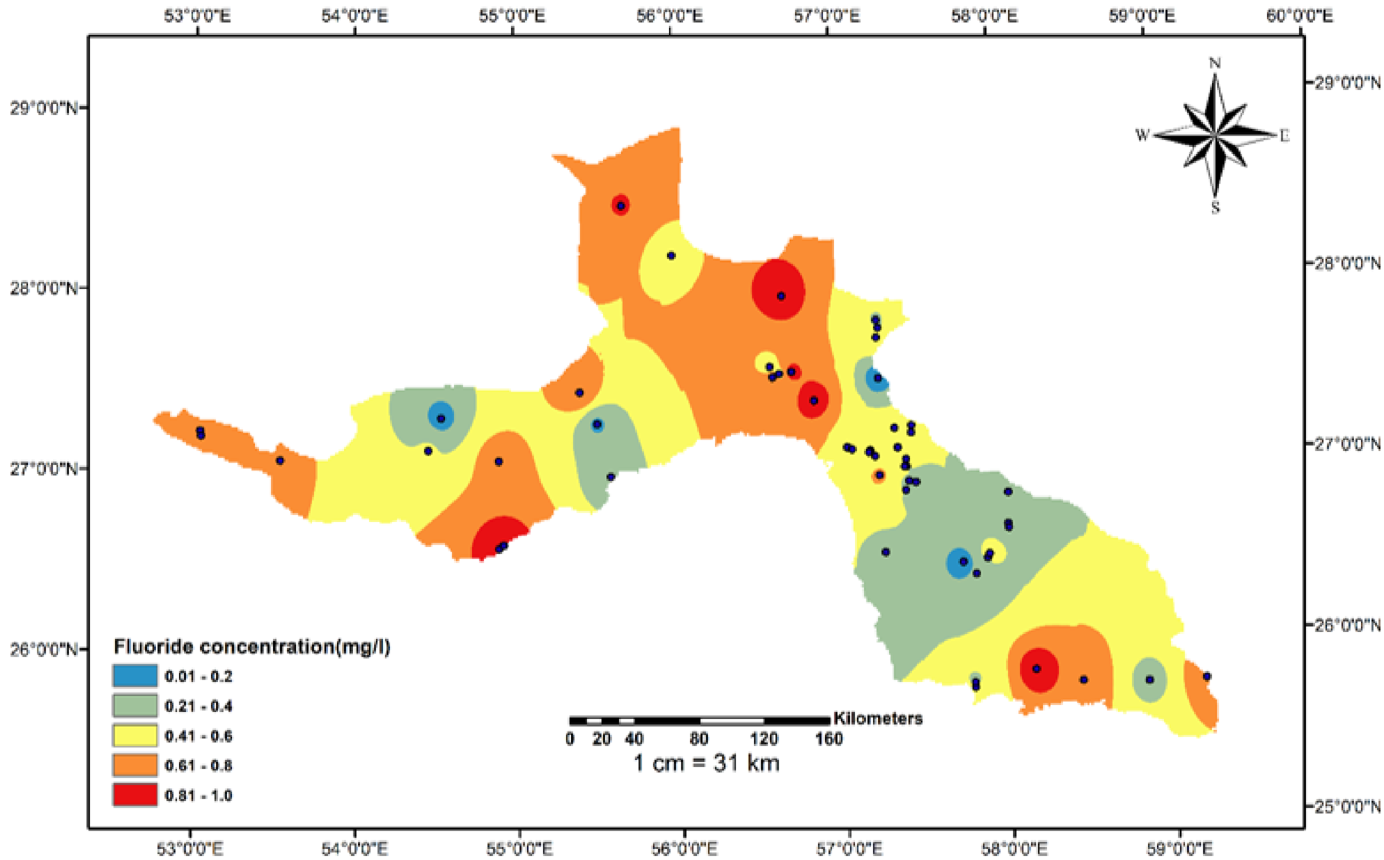

Figure 2

Spatial distribution of nitrate concentration in drinking water of Hormozgan province.

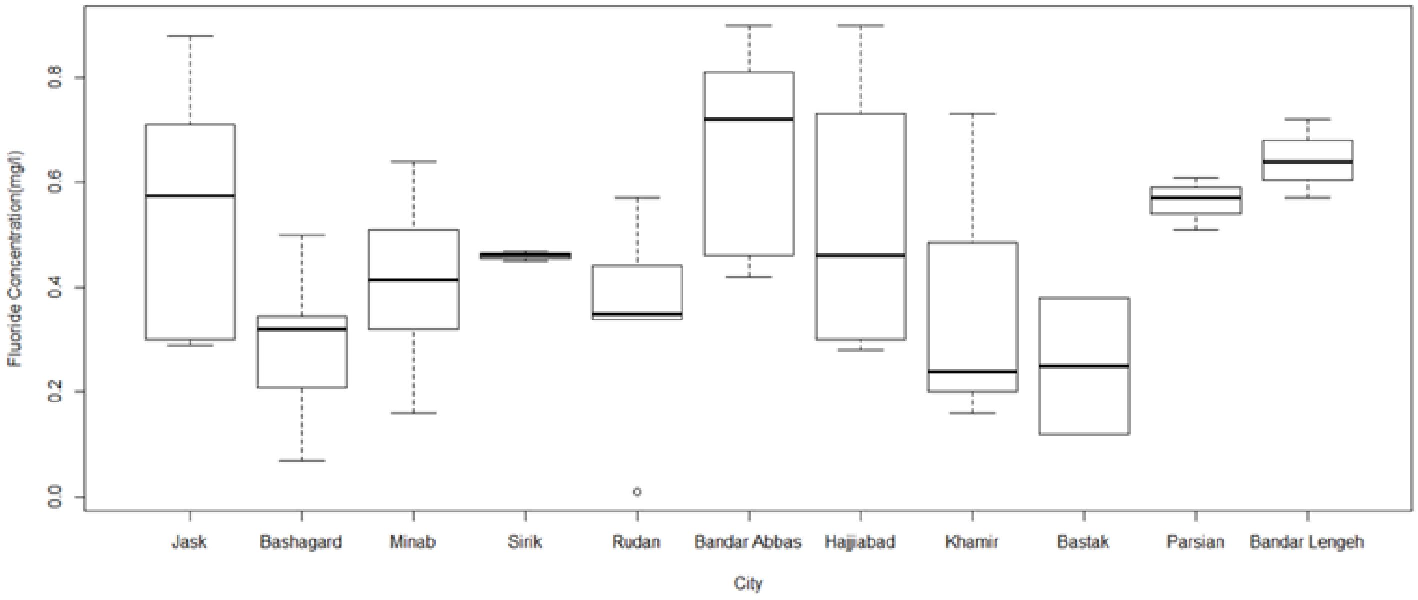

Figure 3

Box Plot of Kruskal-Wallis test in different cities of Hormozgan province.

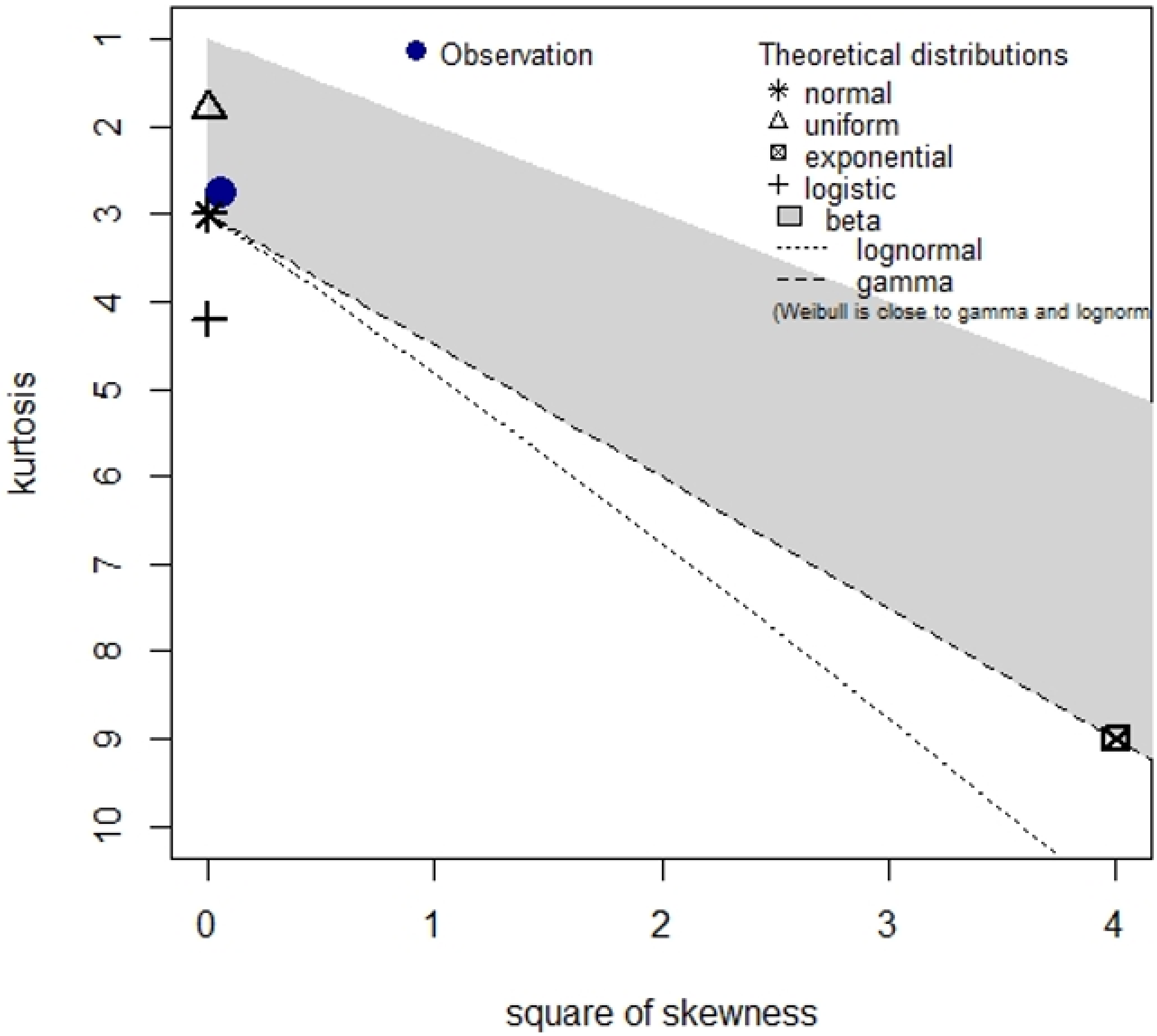

Figure 4

Comparison of different statistical distributions for drinking water fluoride concentrations

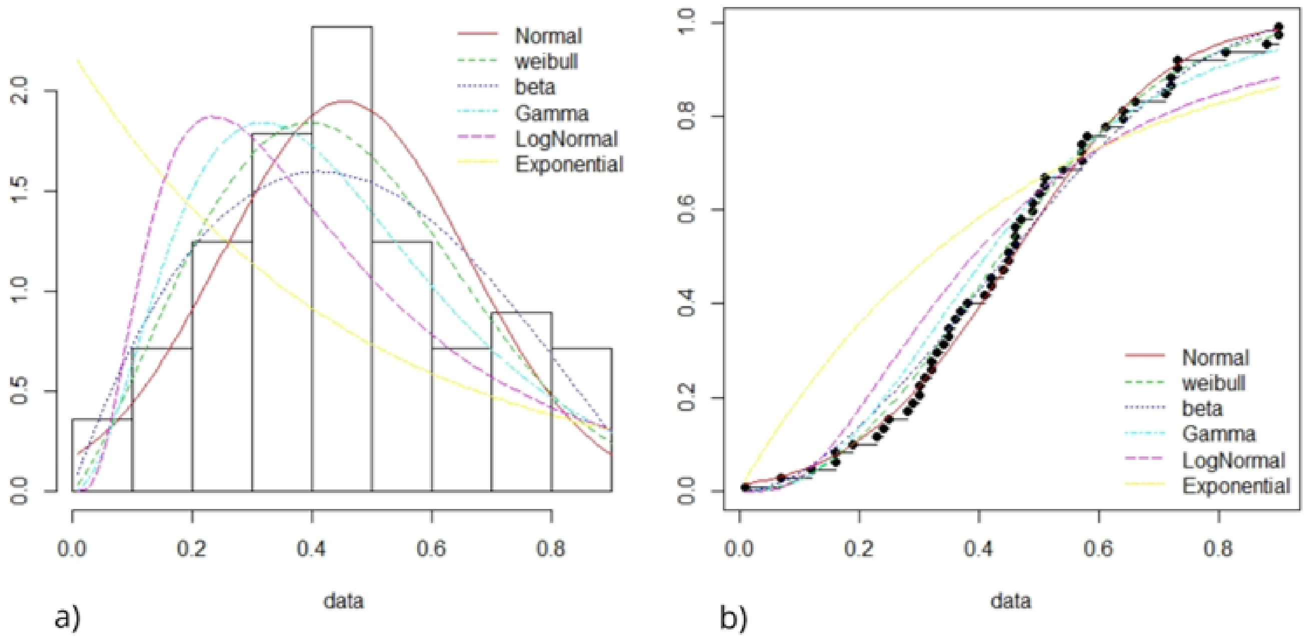

Figure 5

Various theoretical and CFD distributions on fluoride data

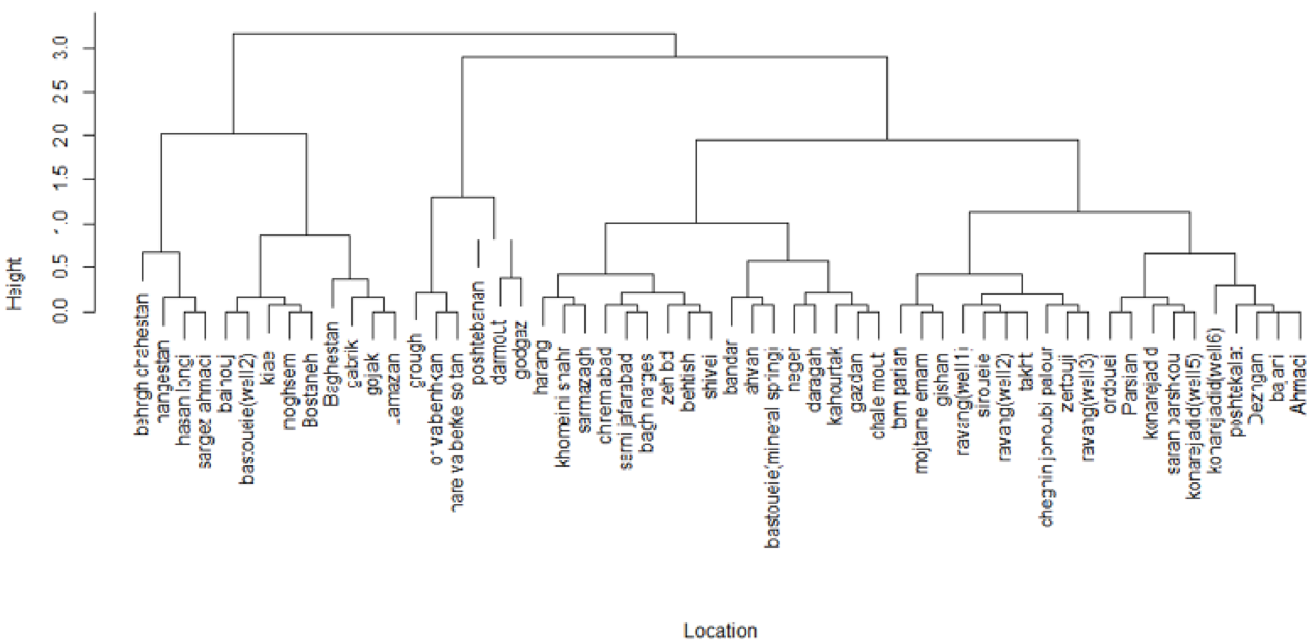

Figure 6

Fluoride concentration dendrogram diagram for the measured points

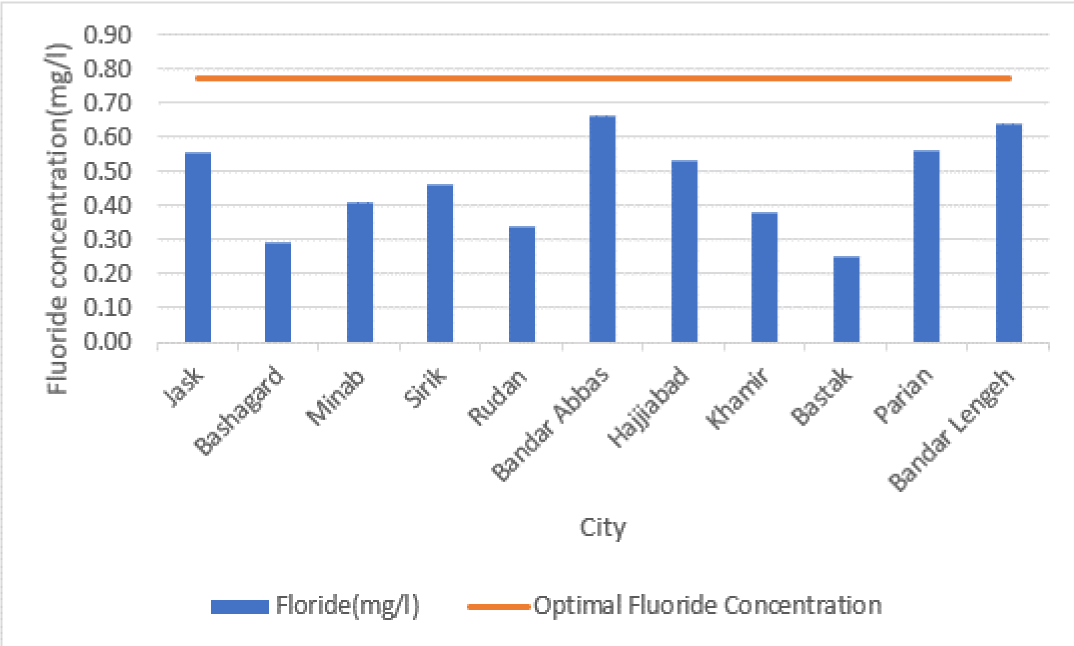

Figure 7

Comparison of fluoride concentration in drinking water in Hormozgan province with calculated optimal dose.

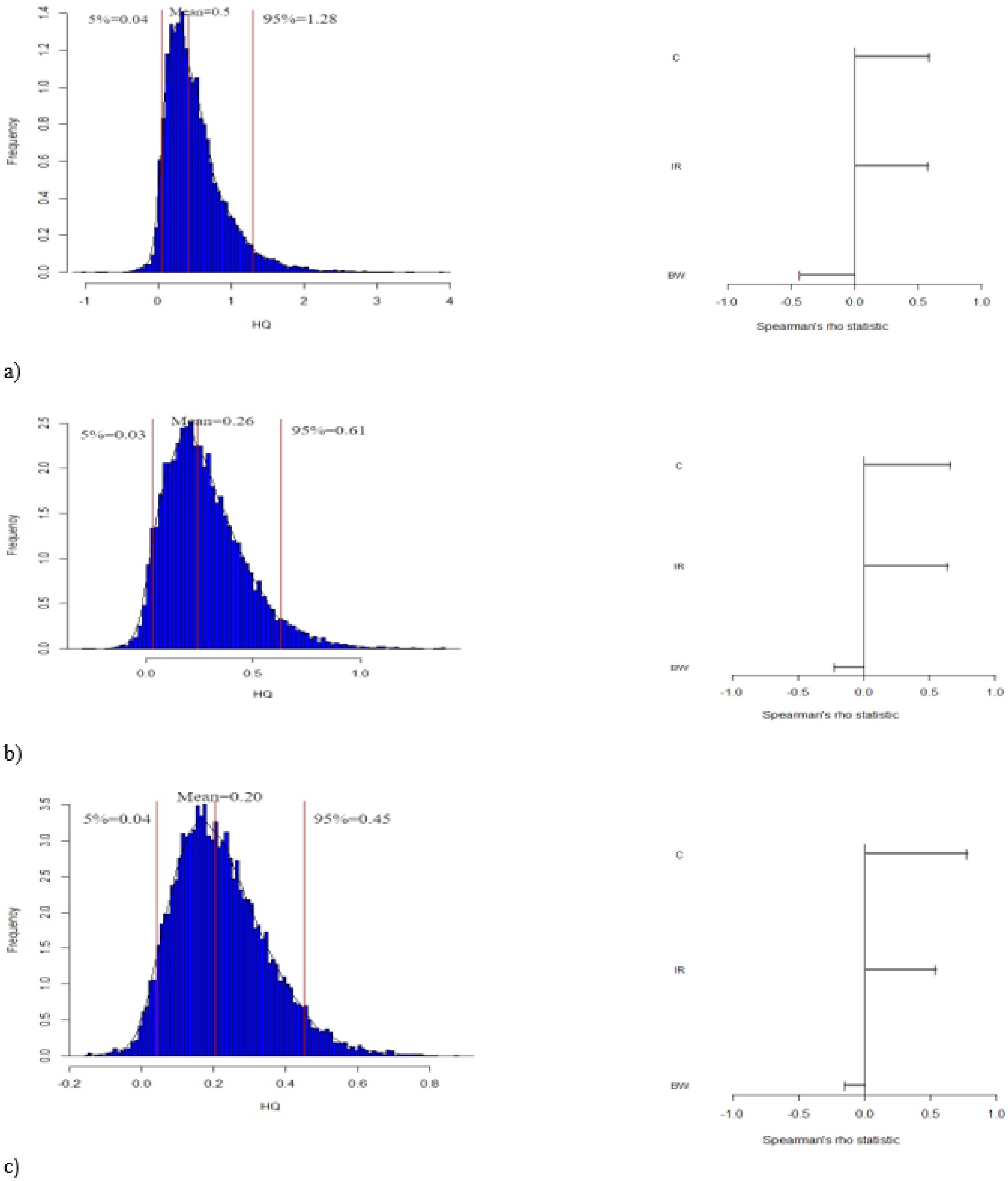

Figure 8

Probabilistic distribution and sensitivity analysis of parameters of HQ in a) children b) teens and c) adults