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Probabilistic Risk Assessment of Exposure to Fluoride in Drinking Water in Victoria de Durango, Mexico

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Abstract: In Mexico, it is estimated that millions of people are affected by the presence of fluoride in groundwater. In wells in the Guadiana Valley in Durango, Mexico, the presence of fluoride has been identified, exceeding the maximum allowable limit established by Mexican regulations (1.5 mg/L). The main purpose of this study was to evaluate the non-carcinogenic risk to the health of the adult population due to fluoride contamination of water, using a Monte Carlo simulation. To this end, the wells were monitored, fluoride concentrations were analyzed according to Mexican regulations and possible concentrations in the vicinity of the sampling zones were determined by applying the ordinary Kriging geospatial tool. Crystal Ball software was used for the simulation, also using data collected through surveys. In terms of dental fluorosis, around 30% of the population mentioned through surveys as having some characteristic of this disease. Of the 70 wells and 2 tanks that were sampled, 90% of them were found to exceed the levels allowed by the regulations. In more than 70% of the wells, the adult population had a non-cancer hazard quotient (HQ) greater than 1. Overall, the HQ for ingestion exceeded 1.8 at the 95th percentile, indicating a significant risk of fluoride-related health problems for the population.

Keywords: fluoride; probabilistic risk assessment; hazard quotient

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1. Introduction

Groundwater has a crucial role in the well-being and economic development of Mexico, especially in arid and semi-arid regions. Reliance on groundwater for drinking water supply is a reality in many parts of the country. However, it is important to note that the presence of potentially toxic elements, such as fluorides, in water raises serious public health concerns [1,2]. The water with the highest concentration of fluorides corresponds to water resources located in mountainous areas, and the presence of this element in groundwater is influenced by various factors such as pH, temperature, well depth, hydrogeology, and tectonic characteristics [3–5].

In Mexico, the central and northern region is an endemic area for fluorosis and arsenicosis, and research has shown elevated fluoride concentrations in the central and northern regions of the country (>1.50 mg/L) and arsenic (>10.0 µg/L) in drinking water samples [4]. Previous research has estimated that approximately 20 million people living in Mexico are exposed to fluoride concentrations greater than 1.50 mg/L through water consumption [6]. And although it is estimated that the main source of water consumption of the inhabitants is bottled water, the second source of consumption is tap water or the public water supply, which represents an important source for Mexicans [7].

In addition to exposure to fluoride through drinking water, there are other sources of intake of this compound. For example, some foods may accumulate concentrations of fluoride when cooked or rinsed with water containing fluoride, as has been observed in the case of rice whose fluoride accumulation is evidenced by iodine staining [8]. In addition, studies have revealed that low-cost beverages called Mexican juices and soft drinks (82 products) contain concentrations of fluoride, which represents a risk to human health, especially in vulnerable groups such as children aged 3 to 6 years in areas with endemic hydroflourosis [9].

High fluoride concentrations can trigger a number of significant health problems. These include dental and skeletal fluorosis, as well as possible toxic effects ranging from lower IQ to kidney and liver damage. In addition, it has been suggested that they may be associated with an increased risk of neurotoxic effects [10–16]. Studies conducted in the last two decades have revealed that a significant proportion of the population exposed to high fluoride intakes experience a variety of damages that manifest themselves in different degrees of fluorosis affecting teeth, bones, blood, lungs, kidneys, nervous system, and genetic and hormonal disorders, as well as affecting the reproductive system and having carcinogenic effects [17].

In Mexico, dental fluorosis due to a high concentration of fluorides in groundwater used for human consumption has been reported in cities such as Aguascalientes, Chihuahua, Durango, Hermosillo, Salamanca, San Miguel de Allende, and San Luis Potosí [18–21].

Specifically, in the city of Victoria de Durango, the water that supplies the city is groundwater extracted from the Guadiana Valley aquifer through wells. For many years, the aquifer has been in shortage due to overexploitation, and most of the water that is extracted is used for the demand of the urban area; however, this demand exceeds its renewal, causing negative effects and putting at risk the quality of water and its structure. For approximately 26 years, several articles have shown that water from the Guadiana Valley aquifer has high fluoride concentrations [3,22–24].

Despite the abundance of studies on fluoride concentration levels in Mexico and specifically in Durango [3,4,22,23,25], much of the attention has focused on the potential health threat, especially in children, through deterministic approaches and epidemiological studies focused on specific population groups [16,20,21,26]. However, there is a marked shortage of information regarding health risk analysis from a probabilistic perspective, particularly in the adult population, and substantial knowledge about skeletal fluorosis in adults is absent.

Human health risk assessment is a methodology used to evaluate the potentially harmful effects on human health resulting from exposure to certain chemical agents during a given period [27]. The risk is a function of the hazard of the substance and the magnitude of exposure. Health risk analyses are presented as critical tools in the assessment and management of risks associated with public health issues. In this context, health risk analyses based on probabilistic approaches, such as the Monte Carlo (MC) method, emerge as essential elements to accurately and comprehensively model and understand public-health-related risks. These analyses not only contribute to more informed decision making but also improve resource allocation and facilitate the communication of uncertainty, aspects that together promote more effective health risk management [27].

The MC method is a quantitative technique that makes use of the probability to imitate, through mathematical models, the random behavior of real phenomena. It examines the role of input parameters as a potential factor for health risk models and determines the output as a probability function of the risk quotient of a particular chemical [28–32]. The findings can be further presented and analyzed by geographic information systems (GIS) according to spatial and temporal variability over a wide study area [31–34].

The main purpose of this study was to evaluate the non-cancer risk to the adult population due to fluoride exposure in the city of Durango. To achieve this objective, an analysis of the fluoride concentrations present in the city's wells was carried out and complemented with surveys of the population. Using the data collected, the Monte Carlo simulation technique was applied. This methodology is of great importance as it allows us to understand the probability of health risk derived from fluoride exposure. Although other approaches exist, such as stochastic simulations and other deterministic methods, the accuracy and acceptability of the Monte Carlo method are firmly established in the scientific community.

The significance of this study lies mainly in its ability to shed light on the level of adult population risk, with a special focus on the areas of greatest vulnerability. Through this detailed knowledge, it becomes feasible to implement specific measures aimed at the protection and welfare of the society.

2. Materials and Methods

2.1. Study Area Specification

The study was conducted in the urban area of the city of Victoria de Durango in the State of Durango in northern Mexico, with geographical coordinates of 24°1′13.2″ N 104°39.454′ W (Figure 1). The population is 688,697 inhabitants. The city of Durango is currently supplied by water from the Guadiana Valley aquifer through 93 wells and 2 distribution tanks distributed throughout the city.

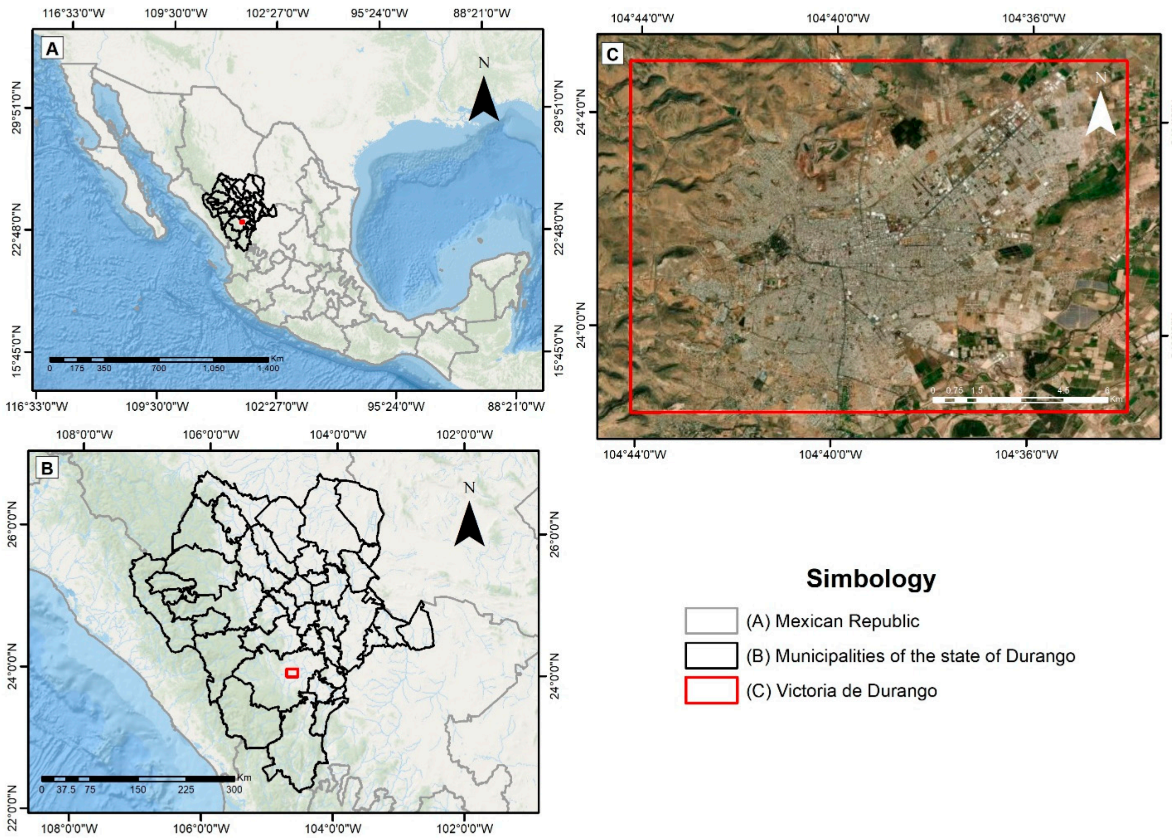

Figure 1. Study area. (**A**) Mexican Republic; (**B**) municipalities of the state of Durango; (**C**) Victoria de Durango.

2.2. Sampling and Analytical Procedures

Sampling was performed according to Mexican Official Standard NMX-AA-077-SCFI-2001 [35]. The fluoride analysis was performed using the potentiometric method with a HANNA instruments HI5522-01 model potentiometer and a HANNA instruments HI4110 model fluoride selective ion electrode. The equipment was calibrated with standard fluoride solutions of 1 ppm, 2 ppm, and 10 ppm (HI4010-11, HI4010-12, and HI4010-10 from HANNA instruments). At extreme pH values, interferences are generated, so it was ensured that the samples were at a pH between 5 and 8. Accuracy was determined using the certified reference standard High Purity Standard of 100 ppm F-, with which three concentrations per standard dilution were prepared: low (1 ppm), medium (5 ppm), and high (10 ppm) within the working range. The pH analysis was carried out according to NMX-AA-008-SCFI-2016 [36].

2.3. Health Risk Assessment

For risk assessment, surveys were applied, in which (a) water consumption patterns and (b) clinical data according to the stages of dental and skeletal fluorosis were considered. The survey considered age, sex, water use, frequency of water consumption, and its management at home. In addition to drinking water, there are other routes of exposure, such as consumption of fluoridated salt, food supplements, and toothpaste, which were not considered in this study, although they could be a factor to be taken into account in future studies. The respondents are of legal age and participated voluntarily in the study, without any sampling or auscultation; their information is confidential, and the persons could decline to participate in the study.

2.4. Determination of the Location of the Sampling Points Using a Geographic Information System (GIS)

GIS has been used in several studies to determine the spatial relationship between environmental pollution factors and their effect on population health, such as the study by Ali et al. [31] in northern India to assess the probability of fluoride exposure risk in drinking water; that of Aghapour et al. [34] in Isfahan, Iran, to assess the health risk of natural fluoride from drinking groundwater resources; and that of Aslani et al. [37] to assess the health risk of fluoride in drinking water supply in rural areas of Maku and Poldasht in Iran. In this study, GIS was used to systematically determine the location of three sampling points within the zone of influence of each well by performing a spatial analysis using ArcGIS 10.6 software.

The estimation of the zones with the highest fluoride concentration in the study area was carried out using a GIS using the geospatial technique of ordinary Kriging interpolation because it showed the best prediction, similar to the results of other studies on the distribution of fluoride concentrations [33,38].

2.5. Exposure and Risk Assessment

The probabilistic risk analysis was carried out following the guidelines established by the Office of Emergency and Remedial Response U.S. Environmental Protection Agency (USEPA) [27].

To assess exposure through both ingestion and dermal contact, USEPA quantitative models were used in a probabilistic manner. These models are described by Equations (1) and (2) [27].

$$CDI = [C \times Ir \times EF \times ED]/[Bw \times AT] \tag{1}$$

$$CDI_{dermal} = [C \times SA \times Kp \times ET \times EF \times ED \times AT1]/[Bw \times AT] \tag{2}$$

where

CDI = Chronic daily intake;
CDI_{dermal} = Chronic daily intake dermal;
CC = Concentration of the hazardous substance in water mg/L;
Ir = Ingestion rate (L/day);
EF = Frequency of exposure (days/years);
ED = Duration of exposure (years);
Bw = Body weight of the exposed person (kg);
AT = Correction factors for averaging time (ED × EF);
SA = Extent of the contact surface between the skin and water (cm^2);
Kp = Dermal permeability coefficient of the substance (cm/h);

ET = Daily duration of the exposure event (h/day);
AT1 = Correction factor for surface area and volume units (10.000 $cm^3/m^2 \times 0.001$ L/cm^3).

In the context of health risk analysis (HRA), exposure is determined by dividing the average daily doses of the substance in question by the threshold doses. These threshold doses represent the levels below which no toxicological effects are expected in the exposed individual during the period considered. This relationship is known as the *HQ* risk quotient (Equation (3)). If the value of this quotient equals or exceeds unity, it is interpreted as meaning that the level of risk is significant [31].

$$HQ = CDI/RfD \tag{3}$$

In this analysis, the toxicological threshold dose for chronic non-carcinogenic effects was based on the oral and dermal reference dose (*RfD* and *RfDD*, respectively) established by the USEPA. The threshold dose or *RfD* used was 0.06 mg/kg/day for both oral and dermal exposure [27].

2.6. Monte Carlo Simulations and Sensitivity Analyses

The simulation was performed under an urban scenario designed for adult individuals. Both water ingestion by the individual and dermal contact during showering were considered, without considering other factors such as accidental ingestion or inhalation.

With the data from the well concentrations, surveys, and previous studies, a model was created in a spreadsheet representing the input (assumptions) and output (predictors) variables according to Equation (1) or (2) (for *CDI* y CDI_{dermal}, respectively) and Equation (3) (*HQ*). In Crystal Ball software (version 11.1.1.1, Oracle, Inc., Austin, TX, USA), the probability distributions of each of the input variables were defined, and the simulation was carried out with 10,000 iterations to calculate the probability distributions of the *HQ* for the population associated with each of the wells. For the overall analysis, the averages of the assumptions were taken, and the simulation was performed with the same iterations. Table 1 shows the parameters of Equations (1) and (2), their probability distribution, the values taken for the *HQ*, and the source from which they were taken.

Table 1. Exposure parameters and their probabilistic distributions.

	Intake				
	Parameter	Units	Distribution	$\underline{X}$	Data Source
C	Concentration of the hazardous substance	mg/L	Log normal	4.48	Data obtained from fluoride analysis
Ir	Daily intake rate	L/day	Log normal	1.425	Compilation of information from surveys
EF	Frequency of exposure	days/years	Triangle	365 *	Mukherjee et al. [17]
ED	Duration of exposure	Years	Log normal	70 *	Mukherjee et al. [17]
Bw	Body weight of exposed person	kg	Log normal	73	Collection of survey data
AT	Correction factors for averaging time	Days	Triangle	25,550 *	Peluso et al. [39]
	Dermic				
SA	Extent of the contact surface between the skin and water	cm^2	Log normal	18.182 *	Mukherjee et al. [17]
Kp	Extent of the contact surface between the skin and water	cm/h	Triangle	0.001 *	Mukherjee et al. [17]
ET	Daily duration of exposure event	h/day	Log normal	0.175	Collection of survey data
AT1	Correction factor for surface area and volume units	L/m^2	Triangle	0.01 *	Peluso et al. [39]

Note: $\underline{X}$ is the mean value used for the simulation. * values taken from the bibliography.

In addition, sensitivity analysis was performed in the same software to identify the input parameters that have a greater weight on the response or output parameters.

3. Results and Discussion

3.1. Sampling

Of the 93 wells in the city, only 70 wells and 2 macrotanks were analyzed since the others were under repair or out of service during the sampling. The water samples were taken after passing through the chlorination system, except for six wells that did not have a chlorination system. The fluoride concentration and pH data for the two periods June 2022 and March 2023 are shown in Table 2. Figure 2 shows the wells that are outside the maximum permissible limit (MPL) according to NOM-127-SSA1-1994 and the WHO [40,41].

Table 2. Physicochemical parameters (fluorine and pH) for the two sampling periods.

	June (2022)				March (2023)			
Well	Fluoride (mg/L)		pH		Fluoride (mg/L)		pH	
	Mean	SD	Mean	SD	Mean	SD	Mean	SD
1	1.89	0.127	7.321	0.106	1.81	0.161	7.322	0.087
6	4.86	0.08	8.197	0.078	4.82	0.083	8.151	0.084
9	5.67	0.055	7.763	0.037	4.79	0.151	7.763	0.031
12-A	1.36	0.061	7.964	0.067	1.14	0.045	7.965	0.027
13	2.02	0.009	7.878	0.029	2.11	0.054	7.941	0.038
14	3.13	0.089	7.517	0.04	3.45	0.169	7.518	0.033
15	3.94	0.36	7.522	0.155	5.40	0.177	7.522	0.127
16	5.45	0.058	8.204	0.026	5.35	0.208	8.185	0.037
17-B	4.77	0.125	7.497	0.048	3.70	0.146	7.497	0.040
18	3.52	0.313	7.409	0.018	2.23	0.033	7.409	0.015
19	3.47	0.163	8.168	0.052	3.16	0.096	8.152	0.021
20	3.57	0.065	8.088	0.032	3.71	0.193	8.102	0.013
21	7.41	0.287	7.863	0.053	7.74	0.256	7.759	0.078
23	3.2	0.22	7.57	0.018	1.41	0.248	7.570	0.015
24	5.5	0.19	9.042	0.12	4.32	0.247	9.043	0.099
25	3.76	0.19	9.139	0.018	4.13	0.160	9.139	0.015
26	4.51	0.046	8.239	0.037	4.33	0.152	8.239	0.038
27	5.94	0.85	7.914	0.034	4.38	0.065	7.915	0.028
28	3.09	0.252	7.808	0.073	3.21	0.119	7.808	0.060
30	4.43	0.175	8.037	0.016	4.54	0.122	8.049	0.005
31	3.61	0.243	7.937	0.026	3.39	0.201	7.925	0.012
32	4.49	0.038	8.127	0.018	4.22	0.097	8.095	0.020
33	0.227	0.005	7.824	0.124	0.28	0.010	7.768	0.093
34	6.21	0.061	8.039	0.126	6.07	0.051	8.028	0.107
37	3.49	0.447	7.498	0.014	3.61	0.117	7.498	0.012
39	3.58	0.232	7.817	0.061	3.55	0.077	7.849	0.058
40	7.76	0.498	8.794	0.047	5.83	0.529	8.794	0.039
41	1.85	0.041	7.522	0.256	1.91	0.017	7.656	0.066
42	4.5	0.061	7.937	0.013	4.80	0.094	7.957	0.011

Table 2. *Cont.*

	June (2022)				March (2023)			
Well	**Fluoride (mg/L)**		**pH**		**Fluoride (mg/L)**		**pH**	
	Mean	**SD**	**Mean**	**SD**	**Mean**	**SD**	**Mean**	**SD**
43	2.07	0.05	7.733	0.026	1.28	0.210	7.734	0.022
44	6.18	0.282	8.185	0.037	6.28	0.148	8.197	0.008
45	1.83	0.055	7.463	0.005	1.91	0.019	7.463	0.004
46	3.75	0.116	8.063	0.036	3.50	0.086	8.051	0.026
48	4.12	0.158	7.268	0.04	2.15	0.061	7.268	0.033
49	4.48	0.189	7.984	0.018	4.34	0.106	7.932	0.029
50	7.38	0.299	8.76	0.065	7.46	0.090	8.845	0.079
52	3.67	0.196	7.993	0.022	3.88	0.094	7.993	0.023
53	4.57	0.261	7.896	0.2	4.77	0.029	7.872	0.009
55	4.94	0.282	7.724	0.048	4.41	0.202	7.725	0.039
56	4.73	0.047	8.106	0.039	4.52	0.153	8.114	0.031
59	4.03	0.362	8.099	0.043	4.11	0.319	8.099	0.036
60	4.63	0.29	7.427	0.016	4.49	0.166	7.427	0.014
61	6.39	0.149	8.099	0.015	6.14	0.124	8.063	0.031
62	5.78	0.096	7.982	0.036	5.73	0.119	8.019	0.029
64	6.27	0.04	7.988	0.011	6.31	0.041	7.953	0.032
65	3.96	0.1	9.013	0.009	2.40	0.252	9.014	0.008
66	5.59	0.38	8.127	0.027	5.54	0.187	8.148	0.009
67	5.72	0.222	8.185	0.028	5.70	0.144	8.221	0.017
68	5.44	0.11	8.538	0.115	3.17	0.114	8.539	0.095
69-A	5.13	0.218	9.139	0.042	3.88	0.306	9.139	0.034
69-B	3.6	0.025	9.285	0.12	3.28	0.082	9.286	0.098
70	2.82	0.205	8.076	0.049	3.15	0.052	8.077	0.040
72	6.05	0.245	8.992	0.032	4.29	0.262	8.992	0.027
73	4.38	0.114	8.153	0.033	4.19	0.148	8.154	0.051
74	1.84	0.175	7.308	0.003	2.06	0.094	7.309	0.003
76	6.5	0.595	8.905	0.103	7.24	0.125	8.905	0.084
77	4.28	0.124	7.576	0.058	4.56	0.048	7.577	0.048
78	5.89	0.596	7.46	0.033	4.37	0.090	7.461	0.027
79	3.25	0.081	7.328	0.272	4.09	0.052	7.328	0.223
81	4.52	0.217	7.681	0.006	3.54	0.352	7.681	0.005
82	5.54	0.375	7.611	0.054	4.92	0.103	7.611	0.045
85	7.81	0.095	8.032	0.027	7.80	0.090	8.043	0.027
86	7.68	0.102	8.031	0.006	7.58	0.188	8.004	0.021
87	2.93	0.245	7.589	0.021	2.32	0.245	7.589	0.018
88	6.51	0.166	8.014	0.003	6.52	0.126	8.008	0.023
89	6.12	0.711	7.555	0.016	5.39	0.086	7.555	0.014
90	4.75	0.305	7.467	0.103	4.78	0.105	7.468	0.085
91	3.34	0.273	8.187	0.131	3.70	0.103	8.188	0.107

Table 2. *Cont.*

Well	June (2022)				March (2023)			
	Fluoride (mg/L)		**pH**		**Fluoride (mg/L)**		**pH**	
	Mean	**SD**	**Mean**	**SD**	**Mean**	**SD**	**Mean**	**SD**
92	8.54	0.201	8.236	0.055	8.16	0.189	8.267	0.046
93	3.65	0.175	8.024	0.039	5.30	0.149	8.041	0.026
Tank	1.81	0.133	7.648	0.121	1.27	0.111	7.648	0.099
Base II	3.65	0.175	7.914	0.03	3.61	0.137	7.917	0.005

Note: SD = standard deviation.

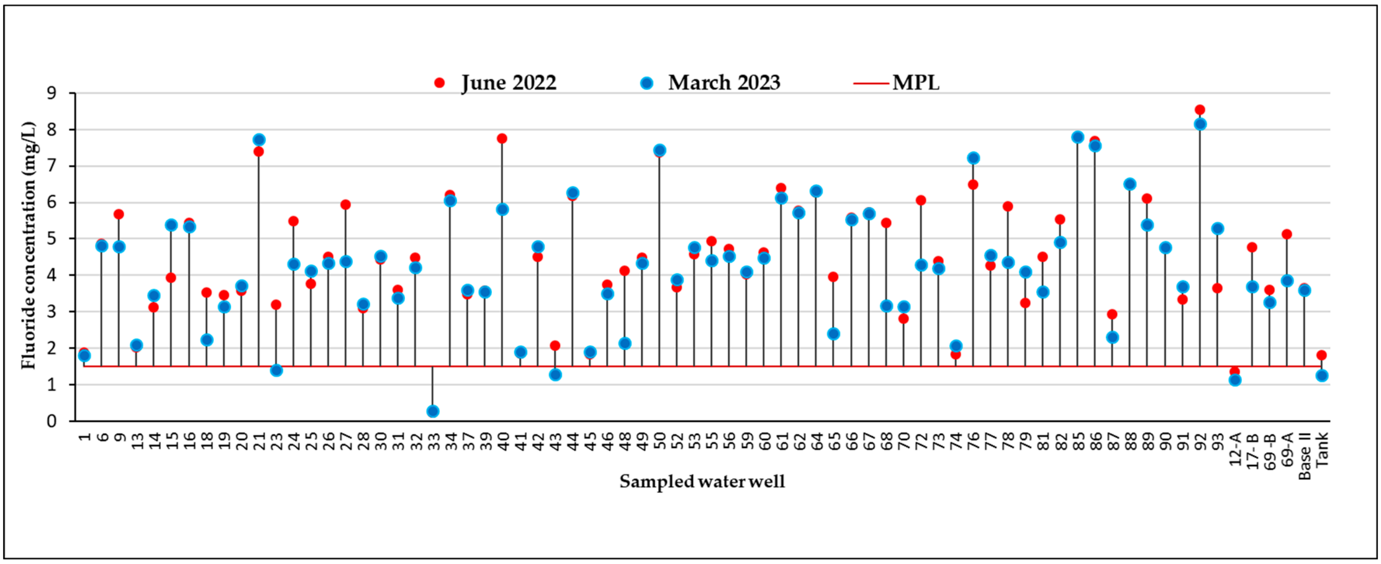

Figure 2. Fluoride concentration in the wells sampled during June 2022 and March 2023.

Of the wells sampled, over 90% did not comply with the Mexican standard. Fourteen percent of the wells exceeded the MPL by more than four times, reaching concentrations above 8 mg/L. The well with the highest concentration was well 92, with a concentration of 8.236 mg/L for the year 2022 and 8.16 for the year 2023. And the lowest concentration corresponded to well 33 with 0.227 and 0.28 mg/L (for 2022 and 2023, respectively).

The study by Vázquez-Bojórquez et al. [42], who conducted a systematic review of original studies, reveals a worrying situation in the northern and western regions of Mexico regarding the levels of fluorides present in tap and bottled water. The authors found that these levels are consistently high and, for the most part, exceed national and international recommendations. In the particular case of Durango, they found that fluoride concentrations ranged from 2.05 to 8.16 mg/L, with a mean of 4.71 mg/L, data similar to those of the present study. For their part, the study by Martinez-Cruz et al. [22], who investigated 97 wells and 7 tanks in Durango during the period from 2012 to 2016, found that fluoride concentrations ranged from 2.3 mg/L to 9.3 mg/L in these water sources. This information reinforces the urgent need to address and mitigate high fluoride levels in these areas to safeguard public health.

In terms of pH, according to the Mexican standard, the permissible limit ranges between 6.5 and 8.5. Table 2 shows that nine of the wells sampled had pH values that exceeded the established limits. In general, the pH value does not usually have a direct impact on human health, although a low pH can increase the acidity of water, which could have an effect on its ability to react [5]. It is important to note that the concentration of fluorides in groundwater is closely related to the presence of fluoride-containing minerals and especially to their decomposition and dissolution through interactions between rock and groundwater [1]. An alkaline environment (7.6–8.6) with a high bicarbonate concentration is shown to be conducive to fluoride dissolution in groundwater, as indicated by the study of Ayoob and Gupta [43]. It is also relevant to note that a high fluoride concentration may be associated with a low Ca/Na molar ratio and with environments with low annual precipitation [4,44]. Figure 3 shows the spatial distribution of concentration for the two periods sampled.

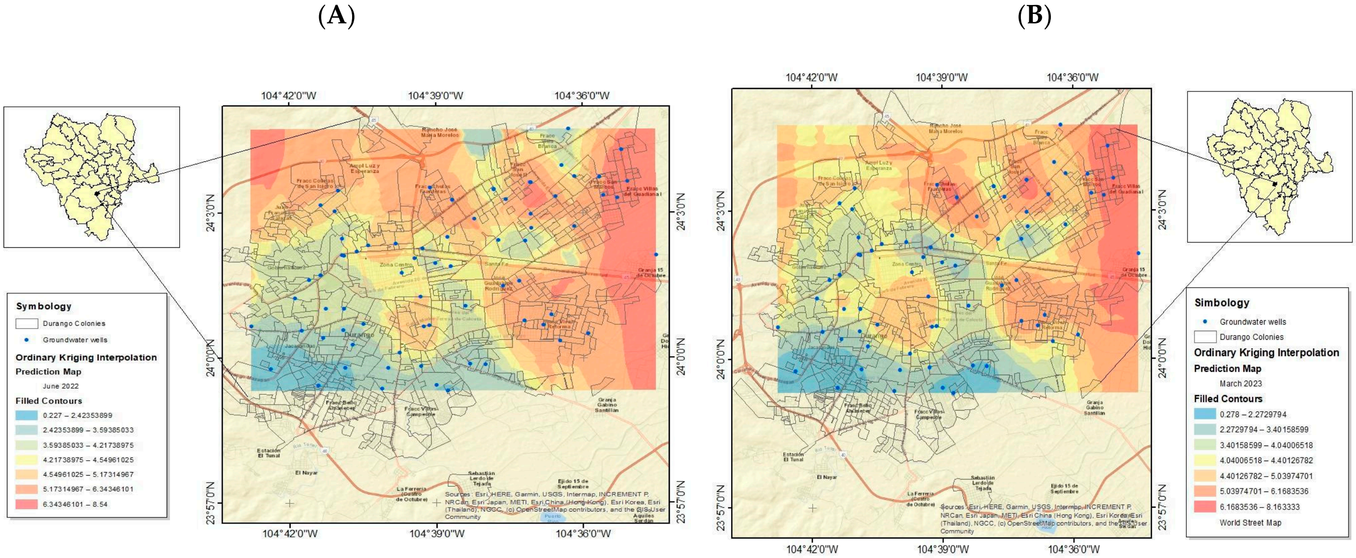

Figure 3. Spatial distribution maps of fluoride concentration in the studied groundwater samples: (**A**) June 2022; (**B**) March 2023.

According to Figure 3, the area with the lowest concentration is the southwest area, and the highest concentration is the northeast area. Similar values were found by Martinez-Cruz [22].

It can be observed that the difference between the two sampling periods was minimal and slightly higher in June 2022; the relationship between the season of the year and fluoride concentration in groundwater can be complex and depends on several interrelated factors. To better understand how the season of the year affects fluoride concentration in a specific region, it is important to consider geological and hydrological characteristics, which were not considered in this study [45,46].

Some studies report significant differences between different times of the year, for example, in southern India, mean fluoride concentrations of 1.26 mg/L and 2.21 mg/L were found before and after the monsoon, respectively [47].

As for the risk ratio that exists between different seasons, this can be influenced not only by the fluoride concentration but also by the amount of water ingested due to the environmental temperature.

3.2. Water Consumption Data

The surveys were applied to an adult population between 18 and 70 years of age, whose average weight ranged between 45 and 124 kg, and an average height of 1.67 m, varying between 1.4 and 1.93 m. The data collected showed that 27% of the population uses water directly from the mains without any type of treatment, while 57% consumes bottled water. From the data collected, it was found that 27% of the population uses water directly from the mains without any treatment, while 57% consumes bottled water. The latter reduces the risk of ingesting high concentrations of fluoride.

According to the results of the surveys, 62% of the population uses water from the municipal network for cooking, consuming quantities that vary between 0.5 and 3 L per day. In addition, it was found that 92% of the population consumes between 0.7 and 2 L of water per day. It is relevant to mention that the intake of 2 L of water daily with concentrations between 5 and 10 mg/L over 10 years can lead to skeletal fluorosis. However, the degree of risk also depends on factors such as age, weight, and nutritional status of the individual [6].

It was also found that the population takes from 2 to 10 min to shower; these data were used to determine the risk due to dermal exposure. The type of toothpaste used by 89% of the population contains a fluoride concentration of 1463 mg/L [48]. If a person with an average weight of 70 kg is exposed to toothpaste with a concentration of 1450 mg/L, 241 g of toothpaste is needed for acute intoxication to occur, and 1545 to 3090 g for a lethal dose [49].

3.3. Clinical Data

According to the clinical data, 34% of the surveyed population presented sporadic pain, 25% presented joint stiffening, 18% presented joint movement limitations, 17% presented chronic joint pain, and 14% presented arthritic symptoms; lower percentages presented spinal cord compression, disabling spinal deformities, and bone fractures for no apparent reason.

Regarding dental fluorosis, about 30% of the population mentioned presenting some characteristic of dental fluorosis (slight lines or striations on the surface of the teeth, appearance of white spots on the enamel, roughness in the enamel, yellowish spots or changes in the shape of the teeth, and dental wear or fractures).

Betancourt-Lineares et al. [21] found that the prevalence of dental fluorosis in 28 federative entities in Mexico was 27.9%; the lowest was detected in Morelos (3.2%) and the highest in Durango (88.8%). The highest community fluorosis indexes (ICF) ($1 \leq$ ICF) were observed in Durango, Zacatecas, Aguascalientes, and San Luis Potosí, indicating that in these states, dental fluorosis is a public health problem.

In accordance with Akuno et al. [50], in individuals over 18 years of age, both dental fluorosis and skeletal fluorosis are observed, along with other conditions unrelated to the skeletal system. In this age group, skeletal fluorosis tends to be more common and tends to worsen over time, while dental fluorosis usually remains stable.

According to Onipe et al. [12] fluorosis is divided into three categories according to severity: mild, moderate, and severe. The main symptoms associated with skeletal fluorosis include chronic joint pain, joint stiffness, sporadic pain, ligament calcification, and osteosclerosis. In addition, skeletal deformities, muscle atrophy, neurological deficits, and restriction of joint motion, as well as severe ligament calcification, may be observed. Chronic conditions lead to calcification and ossification of various ligaments of the spine. Joint immobilization leads to the development of flexion deformities of the hip, knee, and other joints, especially in people between 30 and 50 years of age.

Another study by Mohammadi et al. [51] evaluated the association between exposure to fluoride from drinking water and skeletal fluorosis in five villages of Poldasht County, Iran. The results revealed that people exposed to high concentrations of fluoride were 18.1% more likely to develop skeletal fluorosis compared to those exposed to low concentrations. In addition, 54.5% of skeletal fluorosis was found to be in the age group 71 years and older, and it was more common in women than in men.

Rahman et al. [52], according to the World Health Organization [41] and Dissanayake [53], classified the effects on human health into five classes in terms of fluoride concentration: class I with values < 0.5 mg/L: lead to dental caries, class II from 0. 5 to 1.5 mg/L: promote the development of bones and teeth, class III from 1.5 to 4 mg/L: development of dental fluorosis (mottling of teeth), class IV from 4 to 10 mg/L: dental and skeletal fluorosis (back and neck pain), and class V > 10 mg/L: crippling fluorosis. Based on this classification, Figure 4 shows what was found in this study. Fifty-seven percent of the wells contain concentrations between 4 and 10 mg/L, so the population associated with the polygons of these wells, according to the aforementioned classification, is at risk of dental and skeletal fluorosis.

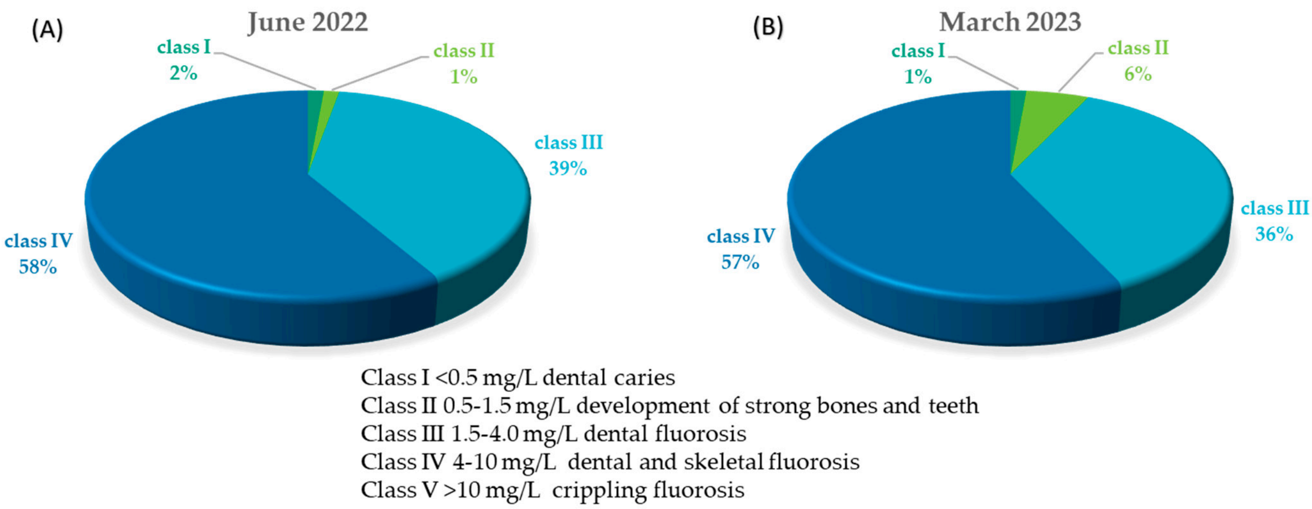

Figure 4. Potential human health effects associated with fluoride ingestion: (**A**) June 2022; (**B**) March 2023.

Skeletal fluorosis represents a significant health problem characterized by the accumulation of fluoride in bones; however, no clear early symptoms are shown, and some may be confused with other conditions. This disorder primarily affects the joints of the body, leading to the appearance of stiffness in individuals of all ages. The chronic symptoms that develop are unfortunately irreversible and permanent, leading to deterioration of health to the point of affecting the ability to work and thus negatively influencing the development of a country. The effects of fluorosis can be reversed by eliminating the source of fluoride, and including a diet rich in calcium and antioxidants can be beneficial [12].

In Mexico, Alarcón-Herrera et al. [54] found a variation between 1.5 and 16.0 mg/L in wells in Durango and showed a linear correlation between the frequency of bone fractures (in children and adults) and the severity of dental fluorosis. According to this study, the prevalence of bone fractures in people (13 to 60 years of age) who consumed water with a fluoride concentration in the range of 1.5 to 8.5 mg/L for nine years was 30%.

According to recent research, such as that carried out by Solanki et al. [10], exposure to fluoride has caused damage to the parathyroid gland, leading to the development of hyperparathyroidism. Ran et al. [55] suggests a possible relationship between chronic fluoride exposure and neurotoxicity, which could affect cognitive function and neurodevelopment in adults.

The limitation of studies on adult fluorosis can be explained by several reasons including a lower prevalence compared to the pediatric population and lack of awareness of skeletal fluorosis in adults, and the intrinsic difficulty in diagnosis as symptoms can be vague and non-specific. In addition, fluoride accumulation in bone over time may be a gradual process and not as evident as dental problems [56].

Despite these limitations, research on adult fluorosis is important because it can have significant consequences for people's health and quality of life. In areas where fluoride exposure is a problem, it is essential that studies be conducted to better understand its prevalence, risk factors, and best prevention practices.

3.4. Risk Assessment of Fluoride

The calculated CDI value was 0.083 mg/kg/day on average for the two years, a value higher than the reference dose. The CDI is calculated based on the average daily consumption of drinking water, fluoride concentration in drinking water, and body weight. It is expressed in units of milligrams per kilogram of body weight per day.

Fernández Macías et al. [32] in their study conducted in the metropolitan area of San Luis Potosí, Mexico, found an CDI of 0.0285 ± 0.014 mg/kg/day for adults, a lower value than in the present study, however, with concentrations in the range of 0.20 to 3.50 mg/L.

The overall risk probability distributions and sensitivity analysis are shown in Figure 5 for the June 2022 season and Figure 6 for March 2023.

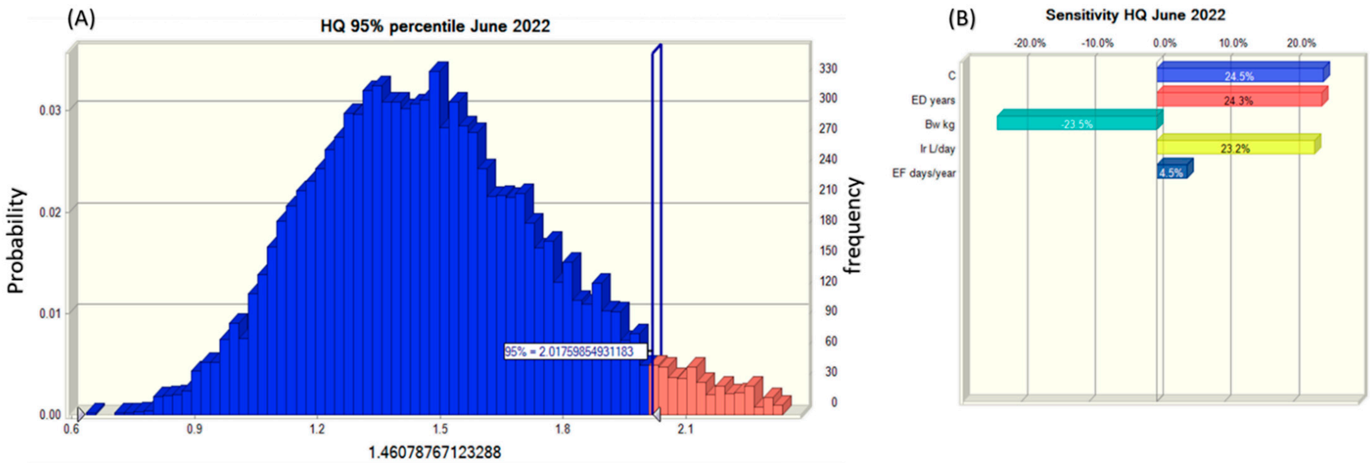

Figure 5. Overall risk probability distribution and sensitivity analysis for the population in June 2022: (**A**) non-cancer risk quotient by intake; (**B**) sensitivity analysis by intake.

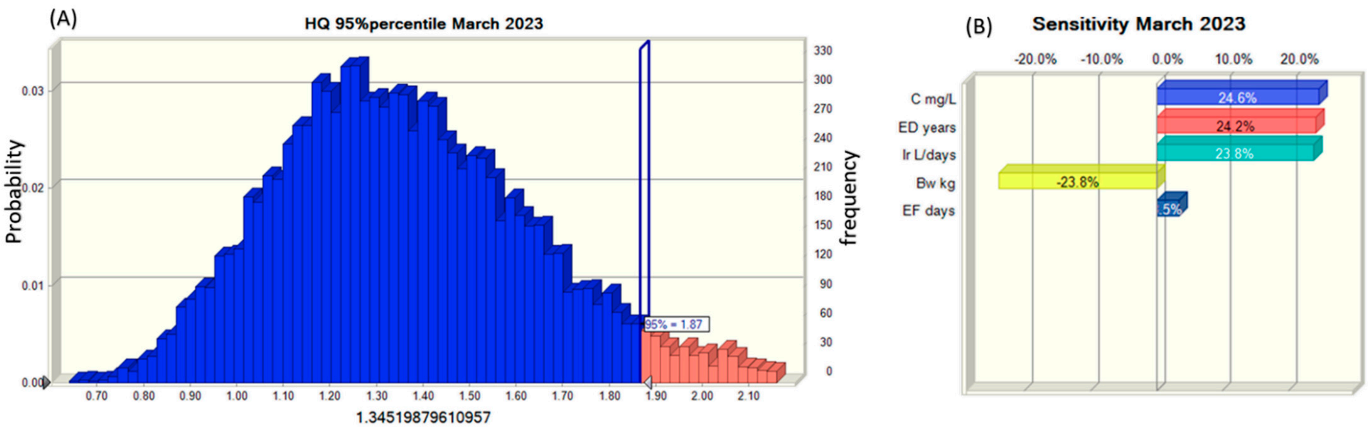

Figure 6. Risk probability distribution and general sensitivity analysis for the population in March 2023: (**A**) non-cancer risk quotient by intake; (**B**) sensitivity analysis by intake.

The HQ for the adult population associated with drinking water consumption from water wells in the city of Durango at the 95% percentile was greater than 1 in both seasons, 2.1 for June 2022 and 1.87 for March 2023. Over 70% of the wells in the two periods had a non-cancer risk quotient greater than 1, which means that there is a high risk of the population having a disease caused by the consumption of fluoride. It is important to emphasize that the consumption of foods, pastes, or mouthwashes with fluoride can significantly modify the calculation of HQ, and this value is significantly higher in children, who are a vulnerable group due to exposure to high concentrations of fluoride according to several studies [32,57].

Similar HQ values were calculated by Kumar Yadav et al. [58], where 71% of the adult population had characteristics similar to those of this study, but with fluoride concentrations ranging from 0.90 mg/L to 4.12 mg/L, having a non-cancer risk greater than 1. Ali et al. [31] in northern India obtained that more than 90% of the adult population has a non-cancer risk quotient greater than 1, with fluoride concentrations ranging from 1.32 to 4.64 mg/L lower than the present study; however, for the overall risk, the average concentration value of this study was found to be 4.48 mg/L.

In Bangladesh, the hazard quotient (HQ) of high-fluoride-water consumption for infants and children was found to have mostly exceeded the threshold value in two seasons:

dry and rainy. And the risk of infants, children, adolescents, and adults at the 95th percentile exceeded 1 in the dry season [52].

Sensitivity analysis studies the difference in an output model that may be related to variations in its input elements, indicating the most relevant factors that influence the output model. Figures 5B and 6B show that intake, exposure duration, weight, and concentration are relevant factors that significantly influence the output model, with weight being inversely proportional to risk [28]. Figure 7 shows the risks calculated by the Monte Carlo method associated with the wells distributed to the population. It can be seen that only 12 wells were at HQ less than unity. The well with the lowest risk was 33, and the one with the highest risk was 85. The risk is associated with other variables such as weight and water intake, so it can be assumed that the weight of the people surveyed in the area corresponding to well 85 was lower than that of well 92, which had a higher concentration in the two periods analyzed.

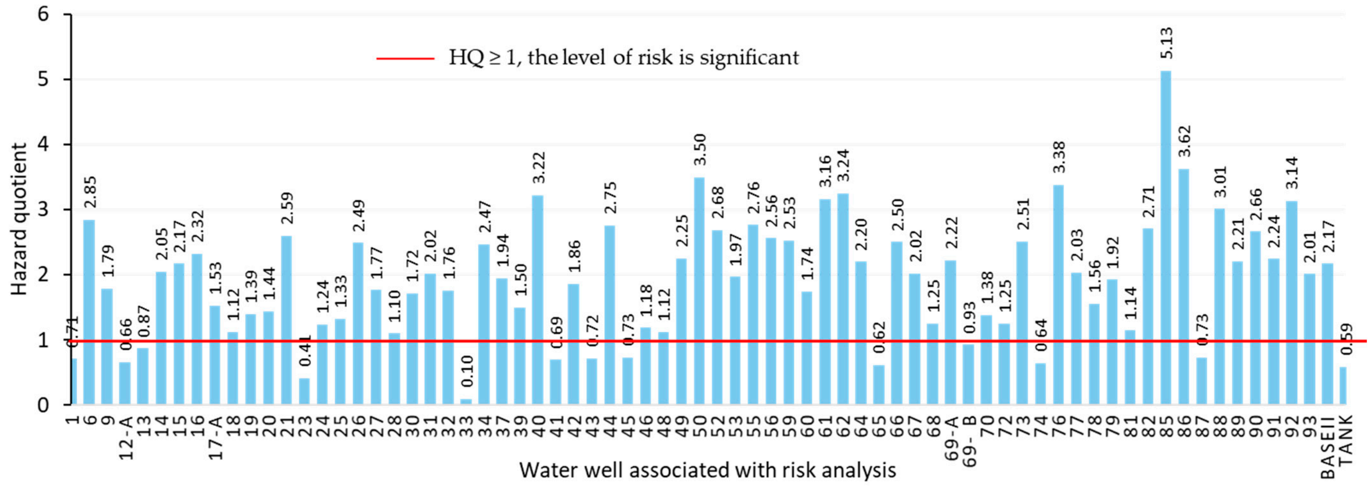

Figure 7. Risk quotient associated with each of the city's water distribution wells. Values above the red line have an HQ greater than 1 and therefore present a higher risk.

The average daily dose from dermal contact was 0.001952 mg/kg/day, and the non-cancer risk associated with fluoride through dermal absorption was 0.05 at the 95% percentile HQ < 1 and well below the HQ from ingestion found in this study. Thus, the main route of exposure is ingestion. The values of the analysis are shown in Figure 8. The sensitivity analysis shows that the contact surface between skin and water apart from the concentration was the most influential factor. Similar values of HQ by dermal contact were found by Mukherjee [17].

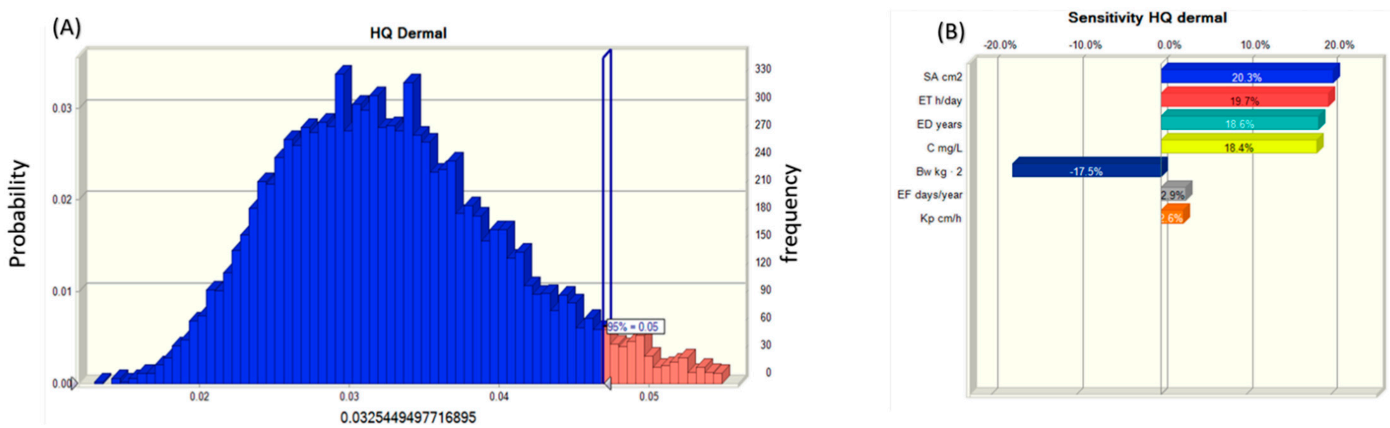

Figure 8. Risk probability distribution and general sensitivity analysis: (**A**) non-cancer risk quotient for dermal contact; (**B**) sensitivity analysis for dermal contact.

4. Conclusions

Ninety-seven percent of the samples analyzed exceeded the maximum permissible limit of 1.5 mg/L of fluoride established by the World Health Organization and the Mexican standard. These concentrations varied from 0.2 to 8 mg/L for both analysis periods and remained consistent in both measurements. It is important to note that the northeastern part of the study area had the highest concentrations. These findings provide essential information on the current landscape and risks associated with fluoride exposure in adults.

The HQ risk quotient for the two periods assessed exceeds unity, indicating that the general population is potentially exposed and faces an elevated risk of fluoride-intake-related diseases. It is highlighted that dermal HQ is notably lower compared to oral intake. In addition, it is identified that the highest HQ is found in the population associated with well 85, highlighting the need to implement mitigation measures especially in that area.

The sensitivity analysis indicates that concentration, intake, and exposure time are the most sensitive factors for the model. For a more comprehensive impact assessment, it is essential to focus on a detailed analysis of the health of the exposed inhabitants. Therefore, it is crucial to focus on the areas with the highest risk. It is also important to highlight the relationship between poverty conditions, unemployment, and social inequalities with human health. These factors make the population even more vulnerable.

Limitations and Future Prospects

In the context of our research, it is important to note that there are certain limitations that must be acknowledged. In particular, an exhaustive evaluation of other possible sources of fluoride exposure, such as food, air, and soil, among other possibilities, was not carried out. It is relevant to note that these alternative sources of exposure may have a substantial impact on the health of the population. In future research, it is essential to address not only this aspect but also to consider socioeconomic factors that may influence fluoride exposure. In addition, it is critical to examine the risk associated with other contaminants that may be present in the aquifer, as these compounds may interact and have combined effects on health. To gain a more complete understanding of potential risks, we also recommend conducting epidemiological studies related to diseases linked to fluoride exposure.

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