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Article

Mapping Environmental Risk of Fluoride Exposure of Drinking Water in a Community of Zacatecas, México

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Abstract: Fluoride (iF) is important inorganic pollutant in groundwater in central México. The aim of this study was estimated environmental risk exposure of iF in residents of Jerez Zacatecas México. Fifty-five tap water samples were collected from households to determine iF concentrations using the ion-selective electrode method. Based on these environmental values, the Hazard Quotient (HQ) was estimated following the methodology of the Pan American Health Organization (PAHO). Subsequently, risk maps were generated from these values to visualize the spatial distribution of high-risk areas within the city. iF in tap water ranger in concentration from 1.3 – 7.3 mg/L (ppm). We estimated HQ in a range of 0.63 to 3.73. iF exposure in tap water is a health problem that must be addressed, carrying out risk communication actions that allow the population to take care of their health through simple actions, avoiding future health costs.

Keywords: fluoride; health; water

1. Introduction

Access to water in adequate quality and quantity for the entire population is an inalienable human right, essential for human development and health care. In Mexico, efforts have been made to ensure that the majority of households to have access to piped water. In the central state of Zacatecas, 94.4% of households are reported to have access to piped water [1] while in the city of Jerez, 93.2% of households are reported to have access to piped water [2]. It is important to note that the quality of the water that reaches homes through the supply system remains constant under routine operating conditions; however, it has been reported that when wells are overused or during dry periods, water is drawn from lower levels of the wells, causing an increase in the concentration of toxic compounds.

Fluorine (iF) is an inorganic compound, abundant in the Earth's crust. It is found as a fluoride ion in groundwater because it dissolves from minerals such as fluorite and apatite present in the rocks with which it comes into contact, the limit in water for human use that most countries have adopted, including Mexico, is based on the WHO guideline of 1.5 mg/L [3], since exposure to this element has been associated with dental fluorosis at doses of 0.06 mg/kg/day [4], endocrine and neurological effects and skeletal fluorosis have also been associated [5–9].

iF has high bioavailability, which is why the consumption of water with this element is a cause of concern for public health, especially considering the vulnerability of the child population and

pregnant women [10–12]. In areas where there are high concentrations of iF in the tap water, there is a high prevalence of dental fluorosis [13,14]. In Mexico, 3.05 million people are exposed to fluoride in water [10]. There is a history of this problem in the state of Zacatecas [10,15], where it's estimated that approximately 22% of the population is at risk of suffering health effects from exposure to iF [16].

The availability of a tool for identifying geographical regions with increased risk, enables decision-makers to optimize resource allocation and improve public health. Thus, the aim of this study is to highlight areas requiring intervention due to elevated fluoride concentrations in water intended for human consumption, employing risk maps as an analytical tool.

2. Materials and Methods

2.1. Study Site

The municipality of Jerez is located in the Central Region of the State of Zacatecas, with an average elevation of 2027 meters above sea level. It borders to the north with the municipalities of Fresnillo and Calera, to the south with Villanueva and Tepetongo, to the east with Zacatecas, and to the west with Valparaiso and Susticacán, approximately 50 kilometers from the state capital, see Figure 1.

Jerez has a population of 43,046 inhabitants, representing slightly over 74% of the total municipality, and it largely concentrates economic activity. Traditionally, it is a city where tourism holds significant economic value, leading to its designation as a “Pueblo Magico”, granting access to specially allocated economic resources. However, its residents persist in primary economic activities such as livestock farming and agriculture, making it a region conducive to the production of apples and peaches [17].

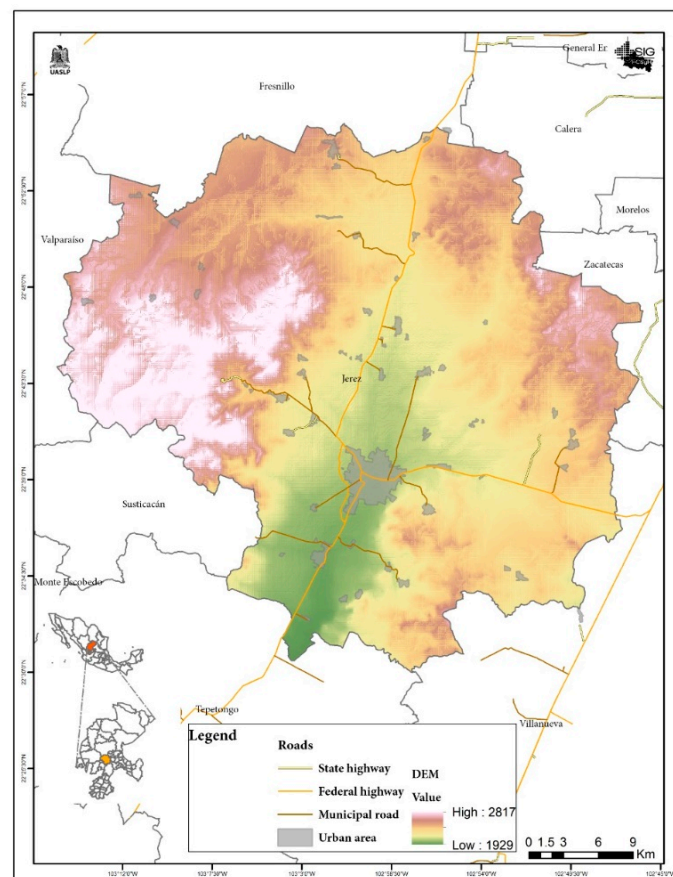


Figure 1. Location of the municipality of Jerez de Garcia Salinas, Zacatecas, México.

2.2. Physiography and Geology

The city is located according to the physiographic regionalization of the National Institute of Statistics and Geography [1] in the Sierra Madre Occidental Province, Sub - province of Zacatecan Sierras and Valleys. These are characterized by its high mountain ranges, with elevations ranging from 2300 to 2700 meters above sea level. According to the Köppen classification, the climate is semi-arid temperate BS1kw, with an annual mean temperature varying between 18 and 22°C. The mean temperature of the coldest month is below 18°C, with cool winters and a summer rainfall regime. Winter precipitation accounts for between 5 and 10.2 percent of total precipitation. The rainy season typically features two periods of occurrence: one in summer from June to September, when peak values are recorded, and another of winter rains from November to February, with less significant precipitation mainly due to cold fronts affecting the region. The dry season spans from March to May.

The most significant superficial water in the area, is the Jerez River, a tributary of the Colotlan River, which converges into the Grande de Santiago River. Jerez is part of a broad valley floor with some nearby hills. Its stratigraphy is represented by lithostratigraphic units of volcanic sedimentary origin with regional low-grade metamorphism (green schist facies), sequences of calcareous-clayey and sandy-clayey rocks, intermediate and acidic, with ages ranging from the upper Triassic to the quaternary. Within the area, there are seven active wells with two pumping systems and eight elevated tanks, Figure 2 show city map whit the ubication of wells.

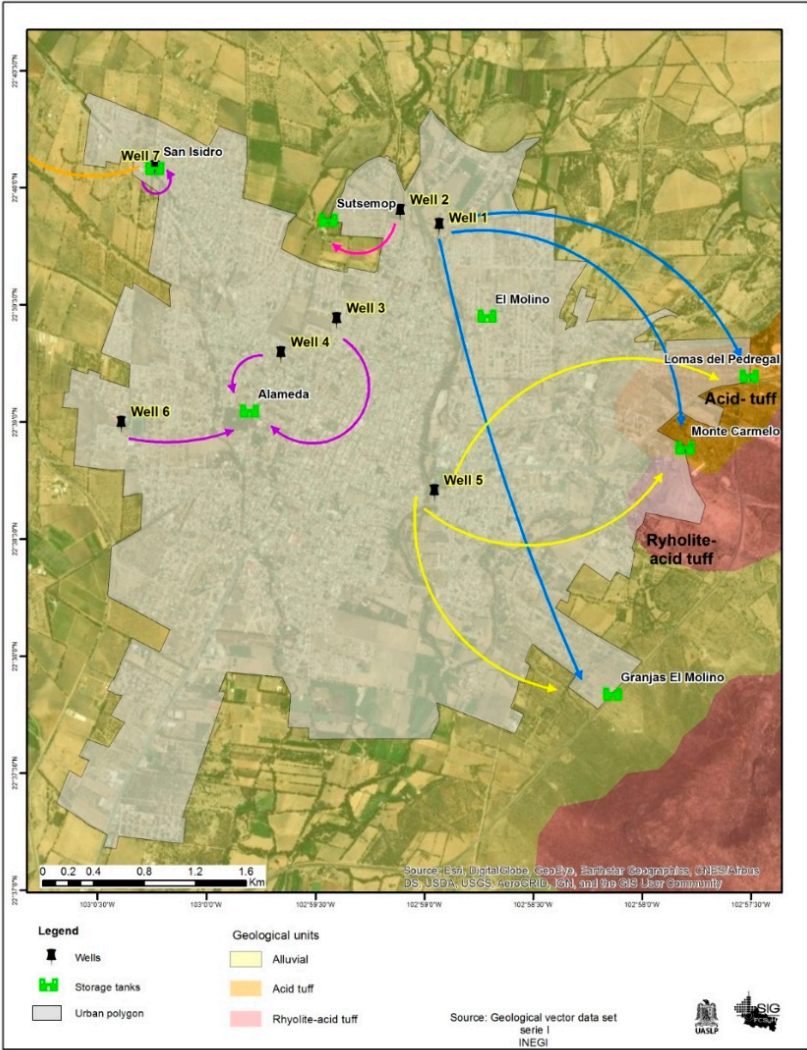


Figure 2. Stratigraphy with extraction points, pumping systems, and elevated water tanks.

According to Nuñez et al. [18] in their study of the hydrogeochemical characteristics and quality of the Jerez aquifer, specifically focusing on wells in the municipal seat city (see Table 1), it can be stated that the groundwater’s origin corresponds to water families circulating through fractured volcanoclastic rocks at depths exceeding 300 m. This suggests that the behavior of ions such as Na⁺, Li⁺, As⁺ and F⁻ results from an interaction between groundwater and the vitreous mesostasis of volcanoclastic rocks, leading to their devitrification. This process involves an exchange of hydrogen ions from the solution primarily for Na⁺ and Li⁺, with concentration of Li observed to correlate with those of F⁻. These ions are considered to originate from a regional flow system.

Table 1. Wells of supply system of the city of Jerez.

Well	Name	UTM ¹ X	UTM ¹ Y	Altitude (m)	Profundity (m)
1	El molino	707393	2507681	2005	280
2	IMMSA	707086	2507794	2008	300
3	CTM	706589	2506938	2005	280
4	Centro de salud	706147	2506670	2006	200
5	Estadio de beisbol	707353	2505582	1993	300
6	Unidad deportiva	704889	2506121	2006	250
7	San isidro	705156	2508140	2022	274

¹UTM (Universal Transverse Mercator).

2.3. Sampling

A cluster sampling method was employed by overlaying a grid on the city map, resulting in a total of 36 squares each 1 km². Only 14 squares, where the highest population density was concentrated, were considered. In each of the selected quadrants, 5 water samples were taken from different households, recording the geolocation of each one using a GarminTM eTrex 20 GPS locator, as well as the elevation above sea level.

Tap water samplers were provided directly by the residents of the household, who were instructed to follow the protocol and were given appropriate containers according to the Mexican standard NOM-230-SSA1-2002 [19]. The standard requires a one-liter water sample to be collected from the tap after letting the water run for at least 30 seconds.

The sample was stored in an amber plastic bottle with a capacity of one liter. The samples were kept at a temperature below 5 degrees Celsius during transportation and were refrigerated until analysis. The pH of each sample was measured at the time of collection using an OrionTM portable pH detector. The time between sampling and fluoride analysis did not exceed 24 hours.

2.4. Determination of Fluorides in Tap Water

Fluoride levels were determined using the ion-selective electrode method described in the Mexican standard NMX-AA-077-SCFE-2001 [20]. We use Thermo Scientific Orion ISE meter with the 9609BNWP fluoride electrode combine to determine fluoride in water. Electrode response follow the Nernst equation. Direct Calibration was used, a series of solutions ranging from 0.5 to 8 ppm were prepared from a 100 ppm standard solution to construct a calibration curve. These solutions were mixed with TISABTM ionic strength adjuster solution in a 1:1 ratio. This solution adjusts the pH and facilitates the formation of metal complexes with ions that could interfere with the measurement. [21].

Three aliquots (50 ml each) of each water sample were taken, and 50 ml of TISABTM was added to each aliquot. Measurements of both standards and samples were conducted by immersing the ion-selective electrode in the solutions with continuous stirring for 3 minutes, the electrode was rinsed with deionized water and carefully dried between each measurement and the final reading (mV) was recorded. Electrode potentials of standard solutions were plotted on the linear axis against their concentrations on the log axis.

As a quality control, it was monitored that the slope of the calibration curve was between -54 to -60 mV as an indicator of the proper functioning of the electrode. The precision of the standard measurements was evaluated, and the accuracy was determined using the CLINICHECK II™ fluoride standard. All standards were at the same temperature as the samples, between 20-25 °C.

2.5. Environmental Health Risk Estimation

To estimate the environmental health risk due to exposure to fluoride in tap water, the PAHO method [22] was used, allowing projection through mathematical formulas based on the fluoride concentration in water.

The steps to estimate the environmental risk are as follows: conduct a dose-response analysis by searching for reference values for health effects due to oral exposure to fluoride in the literature. Then, estimate the population exposure and the risk quotient as detailed below. These estimations are performed for the pediatric population due to their significant vulnerability window and allow for rapid and cost-effective results.

2.6. Dose-Response Analysis

Several reference values are available for fluorides that are useful for estimating environmental risks. However, due to insufficient evidence supporting their carcinogenic effects, only non-carcinogenic risk can be estimated. The No Observed Adverse Effect Level (NOAEL) is 0.06 mg/kg/day; a LOAEL of 0.12 mg/kg/day and the reference dose is 0.06 mg/kg/day, calculated for the critical effect of dental fluorosis, the Minimal Risk Level (MRL) is 0.05 mg/kg-day for the same critical effect [4,23,24].

2.7. Exposure Estimation

To estimate the Average Daily Dose (ADD) (1) for the pediatric population, the equation (1) is used:

$$ADD \left(\frac{\text{mg}}{\text{kg}} \right) = \left\{ \frac{(\text{Environmental concentration (mg/L)} \times \text{ingestion rate (L/day)})}{\text{Body Weight (kg)}} \right\} \times \text{exposure fa} \quad (1)$$

The average body weight for the school-age pediatric population in the municipality of Jerez is 33.912 kg [16], and the water ingestion rate for this population is 0.960 L/day, as estimated for ENSANUT [25]. The environmental concentration corresponds to each water sample, so the ADD was calculated for each of the sampling points.

2.8. Risk Characterization

For the characterization of non-carcinogenic risk, the hazard quotient (HQ) is calculated using the equation (2):

$$HQ = ADD((\text{mg/kg})/\text{day}) / RfD((\text{mg/kg})/\text{day}) \times EF \quad (2)$$

This allows us to estimate the risk faced by the pediatric population for the critical effect, in this case, dental fluorosis [4] for which the reference dose was 0.06 mg/kg/day, was calculated [23]. We calculated HQ for each sampling points. HQ allows us to estimate how many times the exposure dose exceeds the reference dose; a value greater than one represents that the population is at risk, the greater the estimated value, the greater the risk. A value equal to or less than one, is interpreted as that the population is not at risk due to exposure to iF in water in that area.

2.9. Risk Mapping

The mapping of risk areas was based on the fluoride concentration levels detected in tap water samples. This concentration data was used to create a spatial distribution map, highlighting areas with higher fluoride levels. The aim was to identify regions with potential health risk, particularly

for the pediatric population. The cartography of fluorine concentrations and water wells distribution were elaborated in the ArcMap 10.8.2 Geographic Information System (GIS). In particular, the fluorine concentrations map was obtained using the Inverse Distance Weighted (IDW) method, an interpolation process widely used and highly adaptable. The interpolation method uses the average values to estimate the importance of the cells. Thus the values for each cell never exceed the minimum or maximum values of a point. This method assumes that each value in the space has an influence that decreases with distance. The IDW uses the measured values surrounding the prediction location to predict a value for any unmeasured location [26,27].

The fluorine concentrations were captured in a database, exported, and displayed in the GIS. Through the IDW method, the punctual values were interpolated to obtain a layer with continuous concentration values. The map of water wells distribution and its relationship with storage water tanks was obtained using the geographic location of each element and the relate tool of ArcMap™.

3. Results

3.1. Fluoride Concentration

The concentrations of the samples were calculated from the equation of the straight line obtained by plotting the log concentration ($\log C_{iF}$ [ppm]) of the standards on the x-axis against the instrument response (potential, mV) on the y-axis. It was verified that the slope was between -54 and -60 mV when the standards were at a temperature between 20-25°C, as a control of the system's proper functioning [21]. An accuracy percentage of 98% was achieved using Clinicheck™ level II as the reference material. Figure 3 shows an example of a calibration curve; these curves were prepared for each work session.

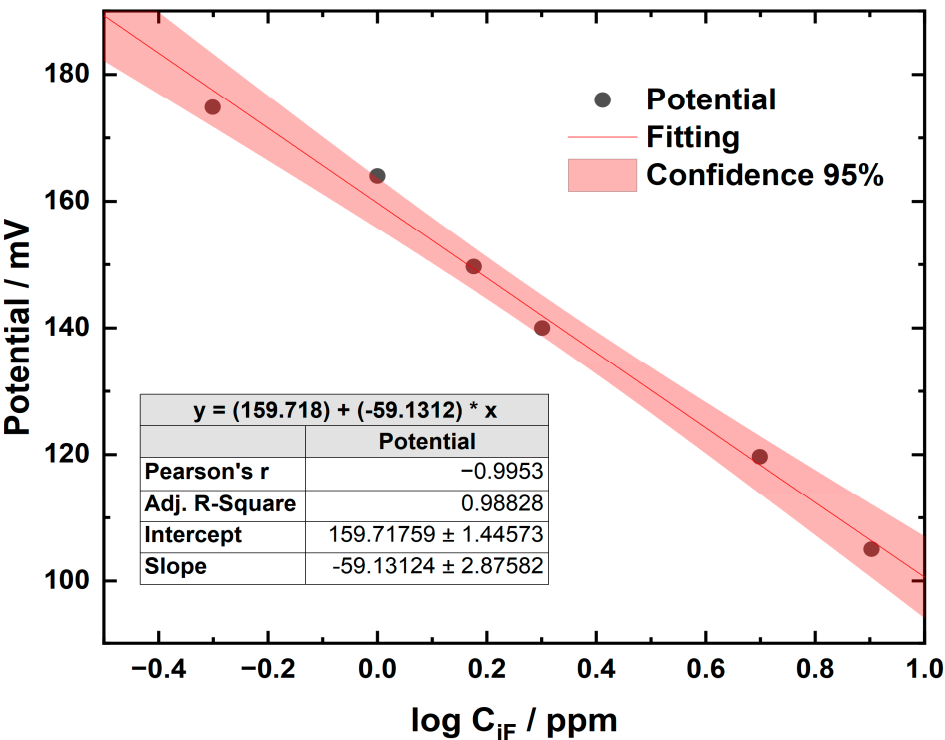


Figure 3. Calibration curve using the ion-selective electrode method for determining the concentraton of fluoride.

The concentration of each sample was obtained by interpolating the response of the electrode against the x axis of the calibration curve. This value corresponds to the logarithm of the concentration, so the antilogarithmic transformation was carried out to obtain the iF concentration

expressed in mg/L. which is the same as in ppm units. As each sample was prepared in triplicate, the mean and standard deviation were calculated as a control for the precision of the measurements.

Most of the samples taken (69 out of 70) showed fluoride concentrations exceeding the limit allowed by NOM-127-SSA1-2021 of 1.5 mg/L [28]. The measured values ranged from 1.34 to 7.30 mg/L.

In Table 2, the results of the fluoride concentration in each quadrant sampled are summarized. The pH of water samples means of 7.8 at the interval of 7.3-8.4.

Only the northwest part of the city had a sample below the permitted limit according to the Mexican Official Standard, whit a concentration of 1.34 mg/L. This area also showed the lowest fluoride values compared to the other quadrants.

Conversely, the area with the highest fluoride levels was the east to northeast part of the city, with concentrations significantly above the permitted limit, the highest being 7.30 mg/L. This information is shown in Figure 4.

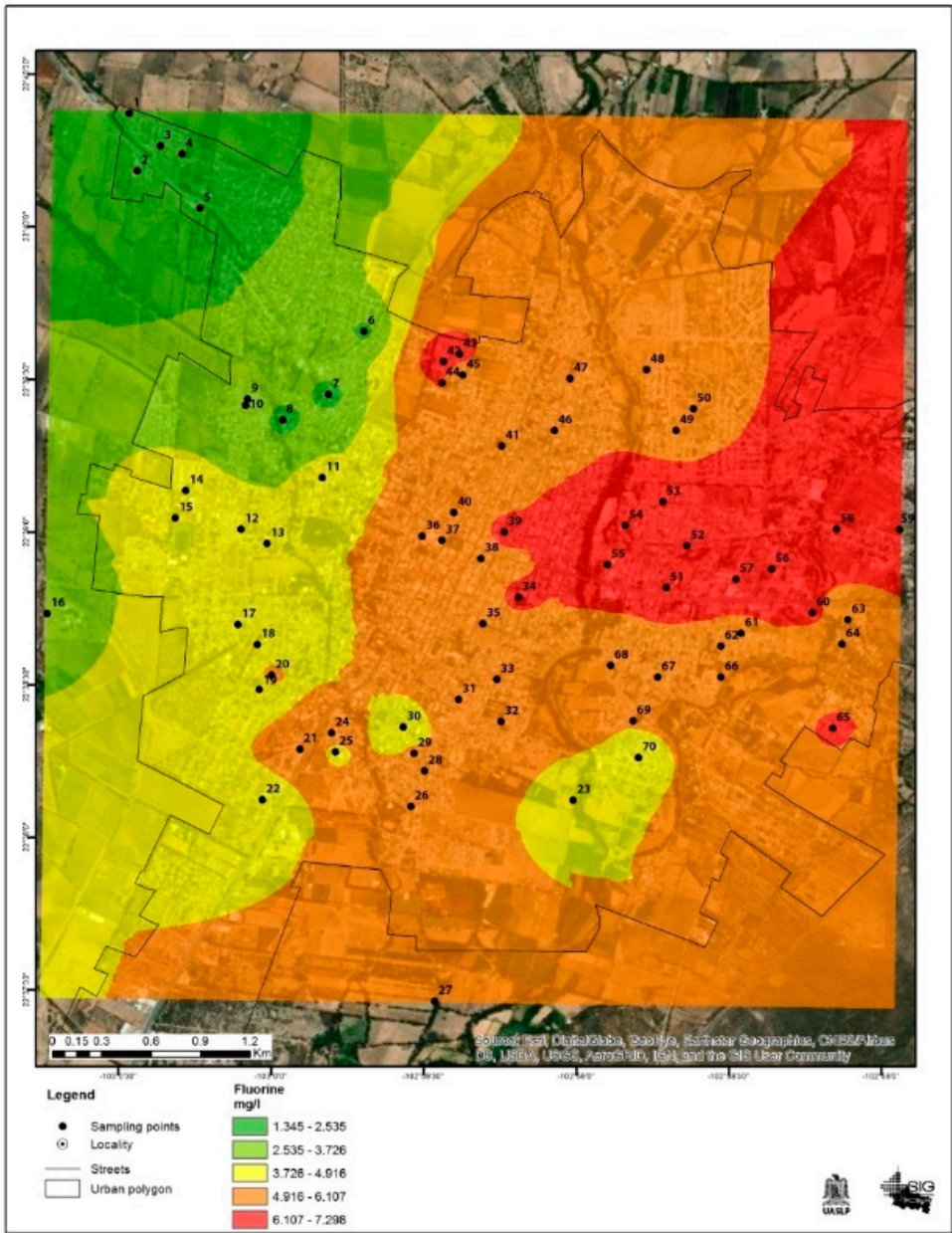


Figure 4. Fluoride concentrations in drinking water in the city of Jerez, Zacatecas México. The black dots represent the sampling sites, and the colors indicate the fluoride concentration.

Table 2. Wells of supply system of the city of Jerez.

Quadrant	n	[F- mg/l]	Average Daily Dose (ADD) ^{1 2}	HQ
		mean (min-max)	mean (min-max)	mean (min-max)
1	5	1.75 (1.34-2.19)	0.05 (0.04-0.06)	0.8 (0.6-1.0)
2	5	2.59 (2.19-3.19)	0.07 (0.06-0.09)	1.2 (1.0-1.5)
3	5	4.05 (3.74-4.36)	0.11 (0.106-0.123)	1.9 (1.8-2.1)
4	5	4.15 (2.55-5.08)	0.12 (0.07-0.14)	2.0 (1.2-2.4)
5	5	4.93 (4.47-5.54)	0.14 (0.13-0.16)	2.3 (2.1-2.6)
6	5	5.02 (4.28-5.46)	0.14 (0.12-0.15)	2.4 (2.0-2.6)
7	5	5.73 (5.40-6.22)	0.16 (0.15-0.18)	2.7 (2.5-2.9)
8	5	5.79 (5.47-6.26)	0.16 (0.15-0.18)	2.7 (2.6-3.0)
9	5	6.14 (4.89-7.19)	0.17 (0.14-0.20)	2.9 (2.3-3.4)
10	5	5.53 (5.32-5.83)	0.16 (0.15-0.16)	2.6 (2.5-2.7)
11	5	6.95 (6.48-7.30)	0.21 (0.20-0.22)	3.6 (3.3-3.7)
12	5	6.97 (6.27 -7.19)	0.21 (0.19-0.22)	3.6 (3.2-3.7)
13	5	5.67 (5.09-6.20)	0.17 (0.16-0.19)	2.9 (2.6-3.2)
14	5	5.01 (4.56-5.45)	0.15 (0.14-0.17)	2.6 (2.3-2.8)

¹ The average weight value for school-age children in the municipality of Jerez is 33.912 kg. This data was obtained from the National Weight and Height Registry 2017-2018. ² The value used for water intake in school-age children at the national level is 0.960 liters. This data was obtained from ENSANUT 2012.

3.2. Exposure Estimation

To estimate fluoride exposure from tap water in children, the exposure dose was calculated, as previously described, based on the fluoride concentrations determined in water. The results can be seen in Table 2. ADD estimated was in ranged of 0.04-0.22 mg/kg/day

3.3. Hazard Quotient

The Hazard Quotient was calculated using data from school-age population of the city, as they are considered the most vulnerable group to adverse effects from elevated fluoride consumption in the water.

In only 4 out of the 70 analyzed points, the risk quotient was less than 1.0, corresponding to the northwest part of the city. The remaining 66 points were distributed in the city and exceed the value of 1.0, in general, HQ ranged in 0.6-3.7. Here, Table 2 show depicts detailed HQ for each quadrant. This indicates that the majority of the school-age population is at risk of experiencing some type of adverse effect due to tap water consumption. These results can be seen in Figure 5.

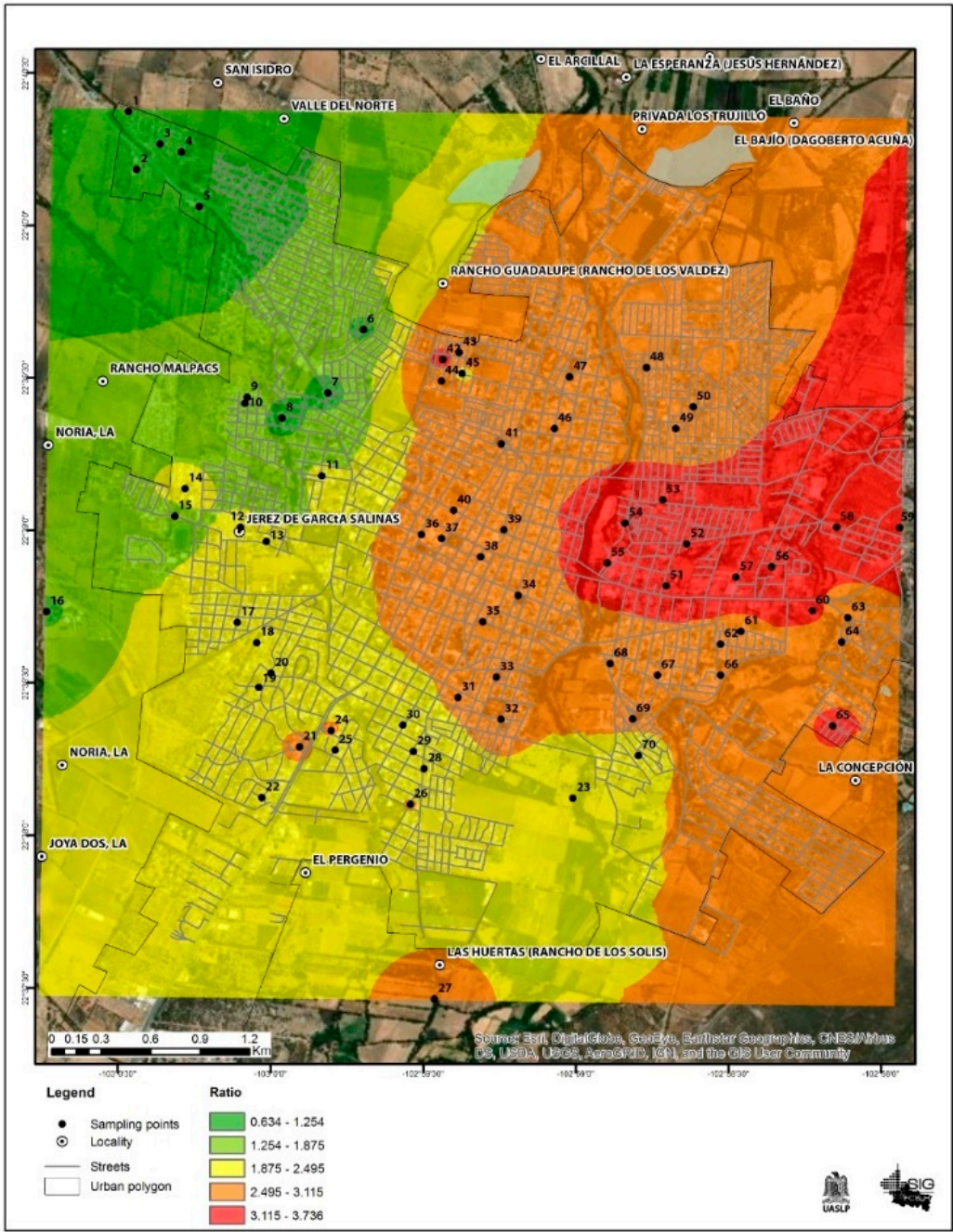


Figure 5. Environmental estimation risk map to Jerez, Zacatecas city.

4. Discussion

It was found that the fluoride levels in the tap water of the city of Jerez, Zacatecas, exceed the limit allowed by the Mexican Official Standard NOM-127-2021 [28] and the WHO of 1.5 mg/L in almost all samples. Only 1 out of the 70 analyzed points complied with this value. Therefore, it can be stated that 98.57% of the analyzed samples are above the permissible limit.

In a previous study conducted in the state of Zacatecas to measure fluoride and arsenic levels, 10 aliquots of tap water were taken, resulting in an average fluoride level of 1.8 mg/L [16]. Although this is above the levels permitted by the NOM and the WHO, it is below the results obtained in this study. In the city of San Luis Potosi, SLP, 190 km from the study site, Perez et al [29] found areas where the mean iF concentration was 2.55 mg/L, estimating a HQ of 1.4 ± 0.980 . These data are consistent with the exposure values, as the pediatric population living in this area exhibited the

highest iF levels. Another study conducted by Farias [30] in estimated a HQ for 1.5 for children exposed to fluoride in tap water in the state of Guanajuato, México. Tap water was identified as the primary source of exposure compared to other drinking water sources. The average concentration of fluoride in tap water was 4.2 mg/L in their study.

In another hand, Jarquin et al. [6] report in a rural population of the state of San Luis Potosí, México, that children exposed to fluoride with mean urinary levels of 3.14 ± 1.09 mg/L all presented dental fluorosis, whit 95% of the cases being severe. All these studies have been conducted in the north-central region of the country, where Alarcón [10] reported a high presence of this element in alluvial aquifers in the arid zones of this region, where volcanics rocks are abundant. The area where the city of Jerez is located is mostly volcanoclastic, so it is possible that, due to the depth of the wells from which the water is pumped, the fluoride comes from the types of rocks and sediments that make up the soil.

The calculation of the HQ was based on the critical effect of dental fluorosis, for which the IRIS reference dose is estimated. In this regard, Gamarra [31] mentions that in areas with higher concentrations of fluoride in drinking water, there is a higher prevalence of dental fluorosis, which is consistent with the estimated HQ values. However, health effects have been reported in the literature that still lack reference doses, such as reduction in IQ scores in children, thyroid dysfunction, kidney dysfunction, and disruption effect [32] indicating that the HQ might be underestimated. Moore [33] suggests considering not only dental fluorosis but also the reduction in children's IQ scores as critical health effects for calculating a safety level for fluoride in drinking water.

Furthermore, the effects of oral fluoride exposure on the human microbiome have not been fully studied [34]. India [35] reported an association between early dietary fluoride exposure and adverse cardiometabolic outcomes in school-aged Mexican children. Qiao [36] reported that oral exposure to sodium fluoride produce immunotoxicity, disrupted riboflavin metabolism, transport and mitochondrial function in mouse model. In humans, Avila, et al [37] refer to chronic fluoride exposure has harmful effects on human health. Besides, preclinical studies associate fluoride toxicity with oxidative stress, inflammation, and apoptosis. Therefore, these health effects should also be considered a risk for the resident population, although the reference doses for these effects are not yet available.

There is evidence of the co-occurrence of iF and arsenic (As) in ground water sources [10,11], which could indicate that the population is at risk of suffering the combined or enhanced toxic effects of these two contaminants, which are still under study. However, the greatest risk is to the pediatric population therefore, it is necessary to take remediation actions to provide them with safe water sources [38]. Locksley F.Castañeda and co-workers reviewed an electrochemical technology to solve this problem, where electrocoagulation is a promising technology for the removal of iF from natural groundwater samples.[39]. However, many remediation technologies are still in the development stage or are very expensive to implement [38].

In a community in central Mexico, an association with biomarkers of early kidney damage was found in a pediatric population exposed to water contaminated with inorganic elements, including As and iF [40], Limon [41] estimated 6 million of children in scholar age would be exposed to fluoride over the Mexican official standards which the authors suggest should be adjusted and enforced to preserve health.

5. Conclusions

This study demonstrates that the population of Jerez, especially children, is at risk of health problems due to exposure to and ingestion of tap water. The risk quotient for this age group ranges from 0.63 to 2.73. It is necessary to implement risk mitigation strategies in the areas with the highest HQ identified on the map in order to provide safe water sources for the population. It is suggested to carry out a health risk communication program focused on educating the population to adopt habits such as not consuming tap water directly, neither for drinking nor for food preparation, as well as reducing the use of products with added fluoride. Additionally, new measures should be

promoted for water treatment and ways to reduce the fluoride levels in it to comply with the NOM standards and protect public environmental health.

In addition, it is necessary to consider routine epidemiological surveillance of diseases associated with exposure to fluoride to prevent and intervene in a timely manner in the exposed population, in order to reduce the environmental burden of these diseases, using as a basis the risk maps generated in this work.

Supplementary Materials: The following supporting information can be downloaded at the website of this paper posted on Preprints.org, Figure S1-S4: Table S1, and Full sampling data.

Author Contributions: Conceptualization, Mónica Martínez-Acuña; Data curation, Luis Estudillo-Wong; Investigation, Rosa Martínez-Esquivel; Project administration, Mónica Martínez-Acuña; Visualization, Humberto Reyes-Hernández; Writing – original draft, Mónica Martínez-Acuña; Writing – review & editing, Sergio Hernández-Covarrubias.

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Conflicts of Interest: The authors declare no conflicts of interest.

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