Original Research
Cariology

Level of natural fluoride in public water supply: geographical and meteorological factors in Brazil's Northeast

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Abstract: This study analyzed the relationships between the concentration of natural fluoride in public water supply and meteorological and hydrographic factors in a northeastern region of Brazil. This was a descriptive, analytical, ecological, longitudinal, and field study conducted by collecting water in 23 municipalities (2019 to 2020) of four macroregions of Paraíba (Brazil): coast (1), borborema (2), agreste (3), and outback (4). Four collection sites were selected per municipality: two near and two distant from the water treatment plant. Fluoride concentration was determined using a combined ion-specific electrode and classified according to the Collaborating Center of the Ministry of Health in Oral Health Surveillance. Meteorological, hydrographic, and population characteristics were also collected. All analyzed samples showed natural fluoride; macroregions 2 and 4 showed the highest mean fluoride concentration, macroregion 4 presented the highest mean temperature, and all macroregions showed a similar pattern of precipitation. The mean fluoride concentration of the four macroregions was below the appropriate value to prevent caries. An increase in precipitation would decrease the fluoride concentration in water. In conclusion, the concentration of natural fluoride varied according to meteorological and hydrographic factors. The concentration in surface waters increased during periods of low precipitation. Therefore, this study provided important information to support implementation of community water fluoridation in this region.

Keywords: Fluorides; Water; Water Supply; Drinking Water.

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Introduction

Environmental fluoride is a natural process involving volcanic emissions and movement of soil particles, which can be transported or removed from the atmosphere through wet deposition.[1] Many factors may interfere with fluoride concentrations in public potable water, such as mineral decomposition of rocks, precipitation, and water and air temperature. For example, groundwater has a high concentration of natural fluoride because water in deep wells is warmer than water in shallow wells in the same location due to geothermal gradients of the earth's crust. In addition, the increase in water temperature increases fluorite solubility and fluoride concentration,[1,2] while a high pH of water and soil favors fluoride concentration due to the anionic exchange of hydroxyl (OH) to fluoride (F) in clay minerals.[3,4]

Fluoride has been detected in all major types of rocks (*e.g.*, igneous, sedimentary, and metamorphic).[5] Moreover, environmental geology controls the dissolution rate of fluoride minerals, which is favored by alkaline conditions.[6] Precipitation also impacts surface waters by increasing water volume and decreasing the concentration of naturally occurring chemical elements (*e.g.*, fluoride).[7] In contrast, acid rain has a high fluoride concentration and increases its penetration in the soil.[8] Air temperature is directly associated with water consumption, and the concentration of chemical elements (including fluoride) in potable water may affect population health.[9-11]

The assessment of fluoride concentration is essential to evaluate the quality of the water for public consumption since it can prevent dental caries and fluorosis.[12] As water with fluoride concentration > 1.5 mg F/L is not suitable for human consumption,[13,14] analyzing and establishing safety intervals of fluoride concentration in public water supplies may prevent diseases and protect human health.[15]

Studies performed in some Brazilian regions reported a high concentration of natural fluoride in public water supplies.[16-19] For example, a study covering 176 municipalities in northeastern Brazil observed a high variation in concentrations of natural fluoride in public water supplies.[20] However, few studies explored fluoride levels in drinking water and meteorological conditions in the region.

In addition, there is a lack of studies monitoring the concentration of natural fluoride in public water supplies on a longitudinal basis.

The analysis of natural fluoride concentration, temperature, precipitation, and access to water allows implementing projects for artificial fluoridation and ensures the effectiveness and safety of this method. The World Health Organization and the International Association for Dental Research recommend adjusting fluoride concentration in public water since it helps prevent dental caries.[21-23] Therefore, this study evaluated the relationships between the concentration of natural fluoride in public water supply and meteorological and hydrographic factors in the state of Paraíba.

Methodology

This was an ecological, descriptive, analytical, longitudinal, and field study developed in the state of Paraíba. Paraíba has 4,018,127 inhabitants distributed in 223 municipalities and divided into four geographical macroregions: a)coast, b)borborema, c) agreste, and d) outback. The average temperature of the state ranges between 26.7 and 32.5ºC, and approximately 81.4% of Paraíba receives potable water; the water system for human consumption supplies 80.23% of the population. Among the Brazilian states, Paraíba has the 13th highest population and the 24th Human Development Index (0.658), with a Gini Coefficient of 0.559.

Characterization of municipalities

Twenty-three municipalities in Paraíba were selected using a purposive sampling. The municipalities were included according to the following criteria: a) those with medium or large population (> 50,000 inhabitants) and regular system for water treatment and supply; b) those located in one of the four geographic-climatic regions of the state; c) with good accessibility (paved roads); d) with available data. After listing the municipalities, it was expected that at least 40% of the state's population would be represented. In fact, the list corresponded to 46% of the total population of Paraíba. Only one municipality in Paraíba had artificial fluoridation and was excluded from the study.[22] Finally, the sample included 23 municipalities. Table 1 shows the characteristics of the macroregions and municipalities, while Table 2 shows the hydrographic characteristics of the selected municipalities.[24]

Data collection

Water from public water supply of 23 municipalities was collected monthly from October 2019 to October

2020. Water samples were identified and classified according to origin and date and collected in 10-mL polyethylene containers (all information was noted on labels). Data collection teams were trained according to the Collaborating Center of the Ministry of Health for Oral Health Surveillance.[25-27]

Collection points were established according to quantity and location of the water treatment plant of each municipality following the National Water Agency Atlas and CECOL.[25] Four collection sites (two near and two distant from the WTP) were selected from public buildings in each municipality (e.g., schools, health units, or squares); the two closest points were considered an internal control for each other.[22,27] The water was collected at the flow line before it entered the building to ensure it came from the distribution system and WTP.[25] In municipalities with more than one WTP, samples from the four sites were collected for each WTP.

Data regarding location and quantity of WTP were obtained based on the National Water Agency. Meteorological characteristics (*i.e.*, precipitation

Table 1. Macroregions and municipalities included in the study.

Macroregion/Municipality	Population	Health region	HDI	Gini	% Water charging
1					
João Pessoa	800,323	1st	0.76	0.62	95
Santa Rita	135,807	1st	0.62	0.47	67
Bayeux	96,55	1st	0.64	0.48	92
Cabedelo	66,68	1st	0.74	0.70	95
Pedras de Fogo	28,389	12th	0.59	0.53	82
Juripiranga	10,717	12th	0.54	0.54	94
2					
Campina Grande	407,472	2nd	0.72	0.58	92
Alagoa Grande	28,482	3rd	0.58	0.55	51
Sumé	17,007	2nd	0.62	0.50	97
3					
Guarabira	58,492	2nd	0.67	0.53	34
Sapé	52,443	4th	0.56	0.51	92
4					
Araruna	20,215	2nd	0.56	0.53	84
Patos	106,984	6th	0.70	0.56	90
Sousa	69,161	10th	0.62	0.54	87
Cajazeiras	61,776	9th	0.67	0.56	21
Catolé do Rocha	30,346	8th	0.64	0.50	99
Princesa Isabel	23,215	11th	0.60	0.48	95
São João do Rio do Peixe	17, 941	9th	0.60	0.53	58
Tavares	14,103	11th	0.58	0.53	93
Riacho dos Cavalos	8,587	8th	0.56	0.44	65
Marizópolis	6,565	10th	0.60	0.52	79
Vieirópolis	5,323	10th	0.57	0.45	31
São Francisco	3,349	10th	0.58	0.48	79

IBGE, 2018.
HDI: Human Development Index.

and temperature) were used to identify possible relationships with fluoride concentration in public water supply.

Initially, containers were identified by municipality, collection point, month, and year. Right after, the sample was sent to the laboratory for analysis. Data

Table 2. Hydrographic characteristics of included municipalities.

Municipality	Hydrographic region	Reservoir type	System type	Subbasin 1	Subbasin 2
João Pessoa	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	Low Paraíba/Mamanguape/ Gramame
Santa Rita	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	Low Paraíba/Mamanguape/ Gramame
Bayeux	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	Low Paraíba/Mamanguape/ Gramame
Cabedelo	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	High Piranhas/Açu
Pedras de Fogo	Eastern Northeast Atlantic	Surface	Isolated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	Low Paraíba/Mamanguape/ Gramame
Juripiranga	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	Low Paraíba/Mamanguape/ Gramame
Campina Grande	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	High and Middle Paraíba/ Taperoá
Alagoa Grande	Eastern Northeast Atlantic	Surface	Isolated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	Low Paraíba/Mamanguape/ Gramame
Sumé	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	High and Middle Paraíba/ Taperoá
Guarabira	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	Low Paraíba/Mamanguape/ Gramame
Sapé	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	Low Paraíba/Mamanguape/ Gramame
Araruna	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	High and Middle Paraíba/ Taperoá
Patos	Eastern Northeast Atlantic	Surface	Integrated	Piranhas	Seridó/Piancó/Espinhares
Sousa	Eastern Northeast Atlantic	Surface	Integrated	Piranhas	High Piranhas/Açu
Cajazeiras	Eastern Northeast Atlantic	Surface	Integrated	Piranhas	High Piranhas/Açu
Catolé do Rocha	Eastern Northeast Atlantic	Surface	Integrated	Piranhas	High Piranhas/Açu
Princesa Isabel	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	Seridó/Piancó/Espinhares
São João do Rio do Peixe	Eastern Northeast Atlantic	Surface	Isolated	Piranhas	High Piranhas/Açu
Tavares	Eastern Northeast Atlantic	Surface	Isolated	Piranhas	Seridó/Piancó/Espinhares
Riacho dos Cavalos	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	High Piranhas/Açu
Marizópolis	Eastern Northeast Atlantic	Surface	Integrated	Coastal Paraíba/Pernambuco/ Rio Grande do Norte	High Piranhas/Açu
Vieirópolis	Eastern Northeast Atlantic	Underground	Isolated	Piranhas	High Piranhas/Açu
São Francisco	Eastern Northeast Atlantic	Surface	Isolated	Piranhas	High Piranhas/Açu

National Water Agency, 2019.

from the Executive Agency of Water Management regarding temperature and level of precipitation of each municipality were monitored monthly.

Analysis of fluoride concentration

Initially, a combined ion-specific electrode for fluoride (ORION 9409BN) and the reference electrode (ORION 900200) were calibrated and connected to an ion analyzer (ORION 710A). Standard solutions (TISAB II) ranged from 0.02 to 6.4 mg F/L, and all solutions and samples were agitated before the analysis. Readings (in mV) were conducted in triplicates for each standard solution and converted into fluoride concentration (mg F/L) using the Excel® software. Millivolt potentials (mV) were converted to mg/L using a standard curve with a coefficient of determination ≥ 0.99.

Parameters used for fluoride concentration analysis

Fluoride concentration was obtained using the mean of three readings from each collection point and classified based on CECOL.[25] The CECOL establishes maximum and minimum values according to the mean high temperature in the region to evaluate prevention of dental caries and risk for dental fluorosis.

The temperature of the included municipalities ranged from 21.7 to 30.9°C from October 2019 to October 2020. A fluoride concentration between 0.65 and 0.96 mg F/L was considered for municipalities with mean temperature < 26.3°C, whereas concentrations between 0.55 and 0.84 mg F/L were used for municipalities with 26.3 and 32.5°C.

Statistical analysis

Several models (*e.g.*, ordinary least squares, weighted least squares, MM-estimation, mixed-effects models, and generalized linear models) were tested to explore the relationships between fluoride concentration and temperature, precipitation, macroregion, and time. After testing different distributions and link functions, the generalized linear model based on the Gaussian distribution with inverse link was considered the best fit. The variable temperature was removed from the model due to its collinearity with precipitation. This model passed the tests for global fit (pseudo-R^2 = 48.23%), normality, linearity, and independence of errors but failed at the test for homoscedasticity of errors. Thus, we used heteroscedasticity and autocorrelation consistent estimator for the variance-covariance matrix of the coefficient estimates since the violation of homoscedasticity may bias the coefficient estimates. All analyses were performed using the R programming language (version 4.1.1), and statistical significance was set at $p < 0.05$.

We used a non-linear link function (inverse or reciprocal function) because variations in fluoride concentration caused by changes in independent variables were also non-linear. A β_0 value of 5.6848 defined the mean fluoride concentration in macroregion 1 for a given precipitation level at time 1 (October 2019). For instance, the estimated fluoride concentration was $1/((\beta_0 + \beta_1 \times 10 + \beta_5)) = 1/((5.6848 + 0.0069 \times 10 – 0.2259)) = 0.1809$ for a 10-mm precipitation. Regarding precipitation levels (x_1), $\beta_1 = 0.0069$ represented the variation in fluoride concentration caused by precipitation changes.

Results

Descriptive data for temperature (mean or median), precipitation, and fluoride concentrations are presented in Figures 1, 2 and 3 respectively. Macroregion 2 presented the lowest mean temperature throughout the analyzed period. Macroregion 4 had the highest mean temperature from October 2019 to January 2020 and July 2020 to October 2020, whereas macroregion 1 presented the highest mean temperature from February 2020 to June 2020 (Figure 1).

Macroregions 1, 2, 3, and 4 presented a similar pattern of precipitation throughout the analyzed period, with peaks in May 2020, May 2020, Abril 2020, and March 2020, respectively; macroregion 1 had the highest value (Figure 2).

An increase in fluoride concentration was observed from July 2020 in all macroregions. Macroregion 2 showed the highest fluoride concentration, followed by macroregions 4, 1, and 3 (Figure 3).

Table 3 shows the fluoride concentration, temperature, and precipitation in the four macroregions of Paraíba (*i.e.*, 52 observations). The most interesting

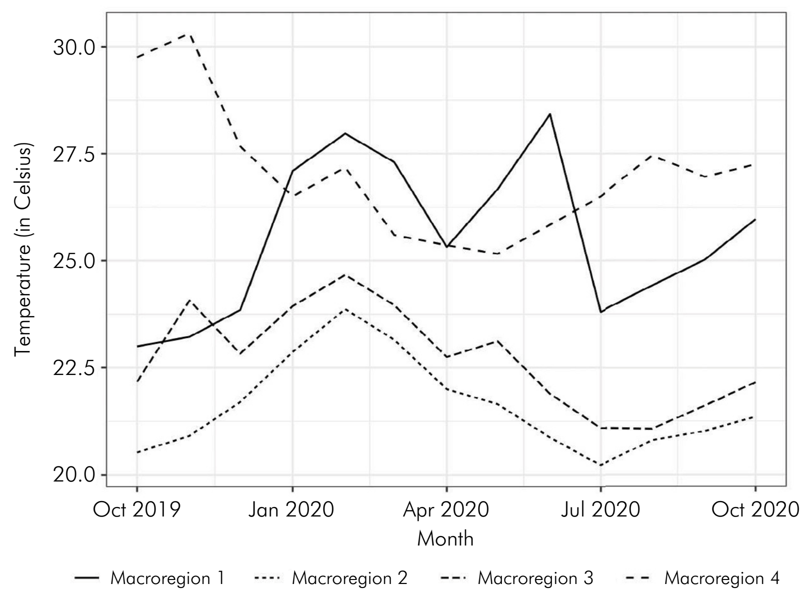

Figure 1. Monthly mean temperature by macroregion.

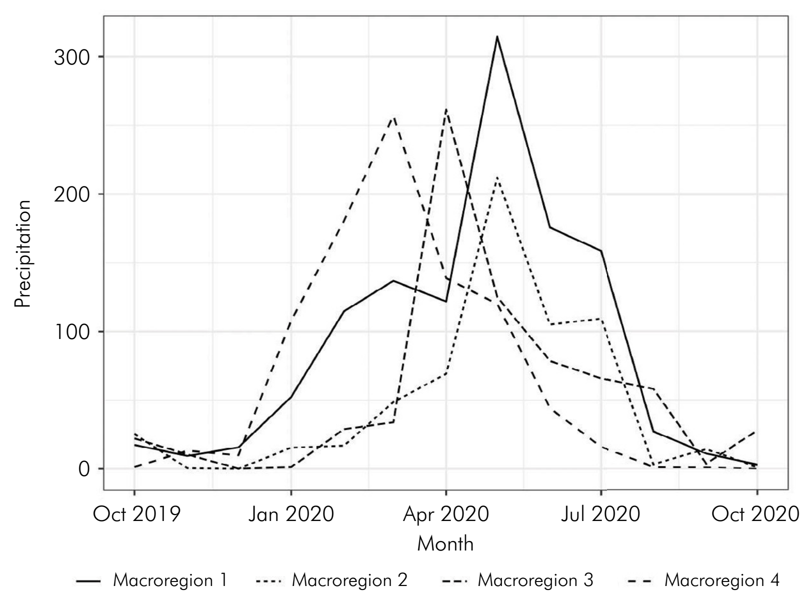

Figure 2. Monthly mean rain precipitation by macroregion.

information is related to Macroregion 4 that showed the highest median fluoride concentration, the highest temperature, and the lowest precipitation compared to the other macroregions.

Table 4 presents the coefficient estimates (β) and respective 95% confidence intervals, standard errors, Z values, and p-values. The intercept and all independent variables were significant ($p < 0.05$). Equations $\beta 2 = -0.9308$, $\beta 3 = 1.2881$, and $\beta 4 = -0.4800$ represented the difference in fluoride concentration from macroregion 1 to macroregions 2, 3, and 4 (respectively) for a given precipitation and a fixed time. For instance, for a 10-mm precipitation at time 5 (February 2020), fluoride concentration

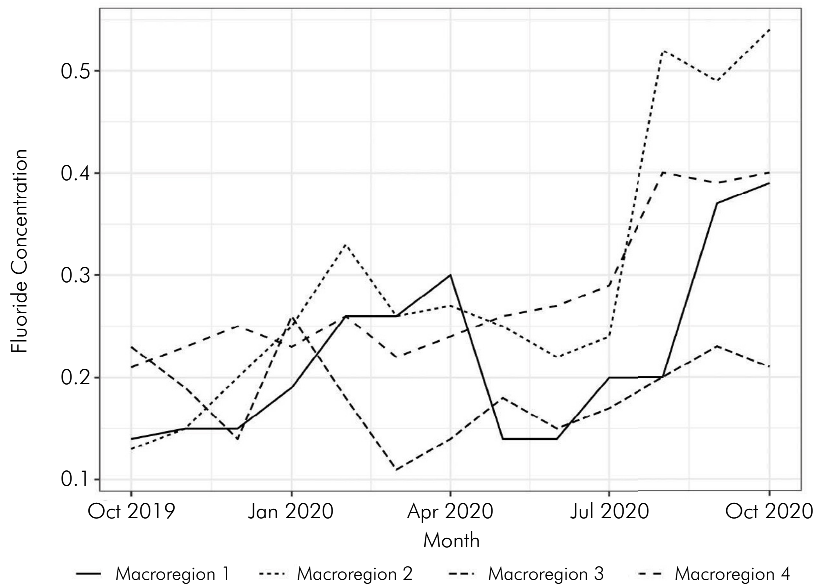

Figure 3. Monthly mean fluoride concentration by macroregion.

Table 3. Characteristics of fluoride concentration. 3.1. Subsection

Characteristic	Macroregion 1	Macroregion 2	Macroregion 3	Macroregion 4	Overall
	n = 13	n = 13	n = 13	n = 13	n = 52
Fluoride concentration (mg F/L)					
Minimum	0.14	0.13	0.11	0.21	0.11
Maximum	0.39	0.54	0.26	0.40	0.54
Median	0.20	0.25	0.18	0.26	0.23
(25%-75%)	(0.15–0.26)	(0.22–0.33)	(0.15–0.21)	(0.23–0.29)	(0.18–0.26)
Mean	0.22	0.30	0.18	0.28	0.25
(SD)	(0.09)	(0.14)	(0.04)	(0.07)	(0.10)
Temperature (°C)					
Minimum	23.00	20.22	21.07	25.16	20.22
Maximum	28.42	23.87	24.67	30.31	30.31
Median	25.32	21.35	22.75	26.96	23.91
(25%-75%)	(23.85–27.10)	(20.87–22.00)	(21.89–23.94)	(25.84–27.46)	(21.97–26.50)
Mean (SD)	25.54 (1.84)	21.61 (1.10)	22.72 (1.18)	27.04 (1.56)	24.23 (2.60)
Rain precipitation					
Minimum	3	0	0	0	0
Maximum	314	212	262	257	314
Median	52	17	29	16	28
(25%-75%)	(16–137)	(3–69)	(10–66)	(1–120)	(10–111)
Mean (SD)	89 (93)	48 (62)	55 (72)	69 (84)	65 (78)

SD: Standard deviation.

Table 4. Summary data of coefficient estimates for fluoride concentration.

Variables	Estimates	Lower	Upper	Standard error	Z value	p-value
Intercept	56.848	46.918	66.779	0.5067	112.203	0.0000
Rain precipitation	0.0069	0.0014	0.0124	0.0028	24.511	0.0142
Macroregion 2	-0.9308	-13.426	-0.5191	0.2101	-44.306	0.0000
Macroregion 3	12.881	0.5313	20.449	0.3861	33.358	0.0009
Macroregion 4	-0.4800	-0.9151	-0.0449	0.2220	-21.622	0.0306
Time	-0.2259	-0.3111	-0.1406	0.0435	-51.942	0.0000

was approximately 25% higher in macroregion 2 than in macroregion 1, approximately 22% lower in macroregion 3 than in macroregion 1, and approximately 11.5% higher in macroregion 4 than in macroregion 1.

The equation $\beta 5 = -0.2259$ represented the variation in fluoride concentration according to time (Table 4), which changed according to macroregion and precipitation since they are inversely related.

Discussion

This was the first longitudinal study (13 months) that monitored and mapped the concentration of natural fluoride in public water supply of 23 municipalities from four macroregions of Paraíba (Brazil). All analyzed samples presented natural fluoride, corroborating studies from other Brazilian regions[16,18,19,28] and countries.[29-31] The main finding was the variation of fluoride concentration values according to meteorological and hydrographic factors.

The determination of fluoride concentration in water is included in international guidelines and legal frameworks of the Ministry of Health. It was observed in this study that the mean concentration of natural fluoride in the analyzed macroregions was below the optimal level for the prevention of dental caries, considering the criteria of CECOL,[25] and its magnitude allowed the implementation of projects for adjusting fluoride concentration. Macroregions 2 and 4 presented the highest fluoride concentration (0.30 and 0.28 mg F/L, respectively) and had similar hydrographic characteristics, such as the hydrographic region (Eastern Northeast Atlantic) and subbasin 1 (Paraíba/Pernambuco/Rio Grande do Norte Coast).

Waters with high pH have high fluoride concentration since the surface charge of several minerals is generally negative at high pH, inhibiting fluoride adsorption on mineral surfaces.[1] A high fluoride concentration is also related to groundwater with low Ca/Na ratio since the processes that reduce the dissolved Ca concentration generally promote subsaturation of fluorite and increase the dissolved fluoride concentration.[1] Fluoride in the mineral composition of rocks is released into the water through rock decomposition and may affect its concentration in water for public consumption.[2] Therefore, these conditions may justify the variation in concentration of natural fluoride found in this study.

Although macroregion 4 presented the second highest mean concentration of natural fluoride and the highest mean temperature throughout the analyzed period, values were below 0.84 mg F/L and did not represent a risk for dental fluorosis. Studies showed that increased water consumption with fluoride concentration < 1.5 mg F/L in high-temperature zones could increase the prevalence of dental fluorosis.[9,13] Dental fluorosis was observed in high temperature regions with fluoride levels below ideal concentrations in drinking water.[13]

Fluoride in drinking water, and thus daily fluoride exposure, is inversely related to caries and positively correlated to dental fluorosis.[18,25] A systematic review[31] study highlighted that some factors may be either determinant or confounding factors for dental fluorosis, such as average annual temperature and maximum daily temperature, rainfall, altitude, and well depth.

Studies conducted in municipalities from macroregion 4 showed a high concentration of natural fluoride in reservoir water, which was not suitable for consumption.[18,20,19,32] Furthermore, the results of this study reinforced the importance of studying other water sources in the region since the diverse fluoride concentration associated with home and public water consumption source, temperature, and fluoridated materials may be a risk for dental fluorosis.

The four macroregions showed similar patterns of precipitation, with the highest peaks in March, April, and May 2020. In this sense, the increase in rain precipitation may have decreased the fluoride concentration in water. For example, an increase in precipitation from 0 to 1 mm (without changes in other variables) would decrease fluoride concentration by approximately 0.13%, whereas an increase from 100 to 101 mm would decrease fluoride concentration by 0.11%. Also, an increase in rain precipitation from 0 to 100 mm (without changes in other variables) would decrease by fluoride concentration by 11.2%, whereas an increase from 100 to 200 mm would decrease fluoride concentration by approximately 10.1%. These results corroborated a study that observed low fluoride concentration in water from areas with increased rain precipitation and vice versa, mainly due to changes in fluoride concentration caused by evaporation.[33]

Our results may be essential for the development of methods to spread and improve the surveillance system of drinking water quality implementation, particularly for projects to adjust fluoride concentration and prevent dental caries that benefit populations with difficult access to fluoride sources and reduce social inequalities.[34-36]

This study also reinforced the need to monitor fluoride levels in public water supplies to ensure that potability standards and the quality of contents are met to maximize benefits in preventing dental caries with the minimal risk of dental fluorosis.[25,37,38]

This study had some limitations, such as the number of analyzed municipalities and the lack of alternative water sources used by the population. However, this longitudinal research provided a realistic representation of the natural fluoride concentration in 23 municipalities of the four macroregions of Paraíba (i.e., more than 50% of the state population). Further studies should investigate the oral health conditions of the population in these municipalities, the implementation of community water fluoridation, and a surveillance program to ensure quality according to rules and regulations.

Conclusions

All water samples contained natural fluoride, and most were below the recommended concentration for caries prevention. The concentration of natural fluoride varied according to meteorological and hydrographic factors. The concentration in surface waters increased during the rainy season. Therefore, this study provided important information to support implementation of community water fluoridation in this region.

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