

**GEOSCIENCES,  
DISTRIBUTION  
OF FLUORIDE IN  
GROUNDWATER AND  
PREVALENCE OF  
ENDEMIC DENTAL  
FLUOROSIS IN THE  
MUNICIPALITY OF  
SANTANA, BAHIA (BR)**

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***Manuel Vitor Portugal Gonçalves***

PPG Territory, Environment and Society  
(UCSAL), Salvador, Brazil

<https://orcid.org/0000-0002-1463-6224>

***Carlos Alberto Machado Coutinho***

Geochemistry of Interfaces (UFBA),  
Salvador, Brazil

***Manoel Jerônimo Moreira Cruz***

PPG in Geology (UFBA), Salvador, Brazil

<http://orcid.org/0000-0002-8488-4936>

***Rodrigo Alves Santos***

Geochemistry of Interfaces (UFBA),  
Salvador, Brazil

<https://orcid.org/0000-0002-5760-6594>

***Débora Carol Luz Porciúncula***

PPG Territory, Environment and Society  
(UCSAL), Salvador, Brazil

<https://orcid.org/0000-0002-2723-4873>

***Antonio Bomfim da Silva Ramos Júnior***

Professor of the Agronomy Course (UEFS),  
Feira de Santana, Brazil

<http://orcid.org/0000-0002-5980-851X>

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**Abstract:** This research aimed to investigate the relationship between the consumption of groundwater with natural levels of fluoride and the prevalence of dental fluorosis in the municipality of Santana, West Bahia, Brazil. Water samples were collected from 52 tubular wells located in the rocks of the Bambuí Group. The physical-chemical variables were measured in situ, with the aid of a multiparameter probe, and aliquots were taken for the analysis of cations (ICP-OES), anions, and even F<sup>-</sup> (SPADNS). The epidemiological research included a cross-sectional study of the prevalence of dental fluorosis in 159 schoolchildren at 12 years of age (Dean Index). Cluster analysis contributed to understanding the prevalence of dental fluorosis, which was 53% (17% in moderate to severe degrees) and its relationship with groundwater consumption with fluoride levels between 0.05 and 8.88 mg. L<sup>-1</sup>, where 43% of the samples exceeded the local optimum potability limit (0.80 mg. L<sup>-1</sup>). This profile of dental fluorosis in the municipality of Santana was in disagreement with data from the national epidemiological survey of oral health and was considered suggestive of an endemic area.

**Keywords:** Geosciences and Health, Bambuí Aquifer, Fluorite, Multivariate Analysis.

## INTRODUCTION

Churchill (1931) observed in his health research carried out in the United States a direct correlation between stains on the enamel of permanent homologous teeth and the levels of fluoride in the water supply, that is, dental fluorosis. Dean et al. (1941) found a correlation between the prevalence of caries or dental fluorosis and fluoride levels in public water supplies in four US cities. Thus, exposure to fluoride by drinking water and the beneficial or harmful effects on human health was consolidated among the topics

most studied by the scientific community (SAEED; MALIK; KAMAL, 2020).

The application of fluoride (F<sup>-</sup>) in preventive dentistry, mainly in water supply, represents a public health measure that has contributed to the reduction of the prevalence of dental caries in several countries (MARTHALER, 2004). This decline in dental caries worldwide has been accompanied by an increase in the prevalence of dental fluorosis, from very mild to mild degrees (AOBA; FEJERSKOV, 2002). Lima et al. (2019) warned that dental fluorosis can be prevalent where fluoride control is poorly performed in public supply water, or when it is not performed at all, especially in groundwater with natural and toxic levels of fluoride.

Cangussu et al. (2002) and Souza et al. (2013) point out that higher proportions of prevalence in moderate to severe degrees of dental or skeletal fluorosis are expected in endemic areas. This is because the profile of fluorosis depends, among other risk factors and social processes, on the exposure of the population to toxic levels of fluoride through drinking water.

Komati and Figueiredo (2013); Frazão et al. (2018) and Barathi et al. (2019) highlight that the control of fluoride levels in the water supply represents an efficient measure to prevent fluorosis and dental caries. Zuo et al. (2018) and HAN et al. (2021) highlight dental fluorosis, skeletal fluorosis, kidney damage, gastrointestinal effects, loss of intelligence and neoplasms among health problems due to chronic fluoride intoxication.

Fluoride is considered an important trace element for dental and skeletal growth and for systemic health, as long as the intake occurs within an optimal limit (MCDONAGHET et al., 2000; MESSAITFA, 2008). Gopalakrishnan and Viswanathan (2012), Castilho et al. (2015), Terra et al. (2016) and Raju (2017) and Chowdhury et al. (2019) highlight that

drinking water with toxic levels of fluoride and above the optimal limit represents the main risk factor for endemic, dental, skeletal or disabling fluorosis.

Fluoride levels found in water can come from both anthropogenic and geogenic sources (MIKKONEN et al., 2018). Marimon (2005), Colombani et al. (2018), Li et al. (2019) and Stepec et al. (2019) highlighted that human activities, the use of phosphate fertilizers, herbicides, the steel, aluminum, glass and tile industries and the burning of coal are among the main anthropogenic sources of fluoride for water.

Minerals constitute rocks and represent the primary sources of chemical elements to the components of ecosystems. In this sense, fluorite is the most fluorine source mineral found in nature, although fluorine can be found in amphiboles, micas, fluorapatite, topaz, lepidolite and cryolite. Naseem et al. (2010) highlights that chemical weathering of minerals that constitute or are accessory to rocks makes chemical elements available to ecosystems and can change environmental quality and human health.

The present research focuses on the relevance of geogenic sources of fluorine to the waters of the Bambuí Aquifer, in Western Bahia. Furthermore, it is noteworthy that Silva, Viglio and Quintarelli (2020) proposed a megaprovince with fluoride occurrence and risk of endemic fluorosis in the middle course of the São Francisco River, in Bahia and Minas Gerais, based on a low-level geochemistry survey. density. These authors were also based on hydrochemical studies or on the fluoride-health relationship by Velásquez et al. (2006), Ferreira (2007), Costa (2011), Cruz, Coutinho and Gonçalves (2015), Carvalho (2017), Gonçalves et al. (2018), Gonçalves et al. (2019) and Gonçalves et al. (2020).

This way, the importance of groundwater in the municipality of Santana is highlighted,

collected from tubular wells arranged in the rocks of the Bambuí Group, for the public supply of water in peripheral and rural areas. However, the integration of water quality and health information, such as the fluoride-health relationship in the Bambuí Aquifer in Bahia, is incipient. Therefore, this research aimed to investigate the relationship between the consumption of groundwater with natural levels of fluoride and the prevalence and severity of dental fluorosis in the municipality of Santana, in the west of Bahia, Brazil.

## **MATERIAIS AND METHODS**

### **AREA OF STUDY, CLIMATE AND HYDROGEOLOGY**

The municipality of Santana is located in Western Bahia (Figure 1), has an area of 1,820.20 km<sup>2</sup> and in 2010, according to census information from the IBGE (2010), had a population of 24,750 inhabitants and a Gross Domestic Product (GDP) of 130,550 thousand reais. In 2010, it had a value of the Human Development Index - Municipal (HDI-M) of 0.608 (ATLAS BRASIL, 2013). In addition, in 2010, in relation to the 417 municipalities in Bahia, it was ranked 125th in terms of HDI-M, 133rd in terms of income, 134th in terms of GDP and 257th in terms of adequate sanitation. These are the basic conditions available to individuals and public authorities to overcome inequities in health and for human and social development.

The municipality of Santana is part of the regional climate context that varies from sub-humid to semi-arid, with average annual temperature values of 24.3 °C and average rainfall distributed between 800 and 1,010 mm/year (INMET, 2016). Furthermore, rainfall is concentrated between the months of November to April and the drought is distributed between May and September.

In regional geology, Neoproterozoic pelitic and carbonate rocks of the Bambuí

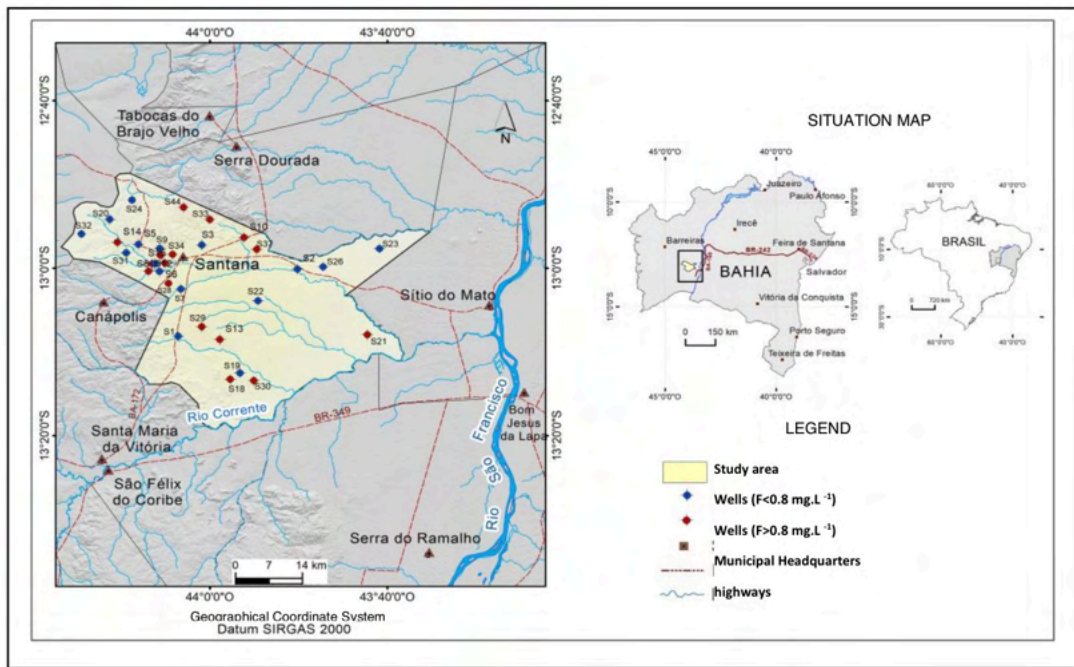


Figure 1 - Map of location, situation and indication of the distribution of fluoride levels in groundwater samples from the municipality of Santana, in the west of Bahia, Brazil.

Source: Prepared by the authors.

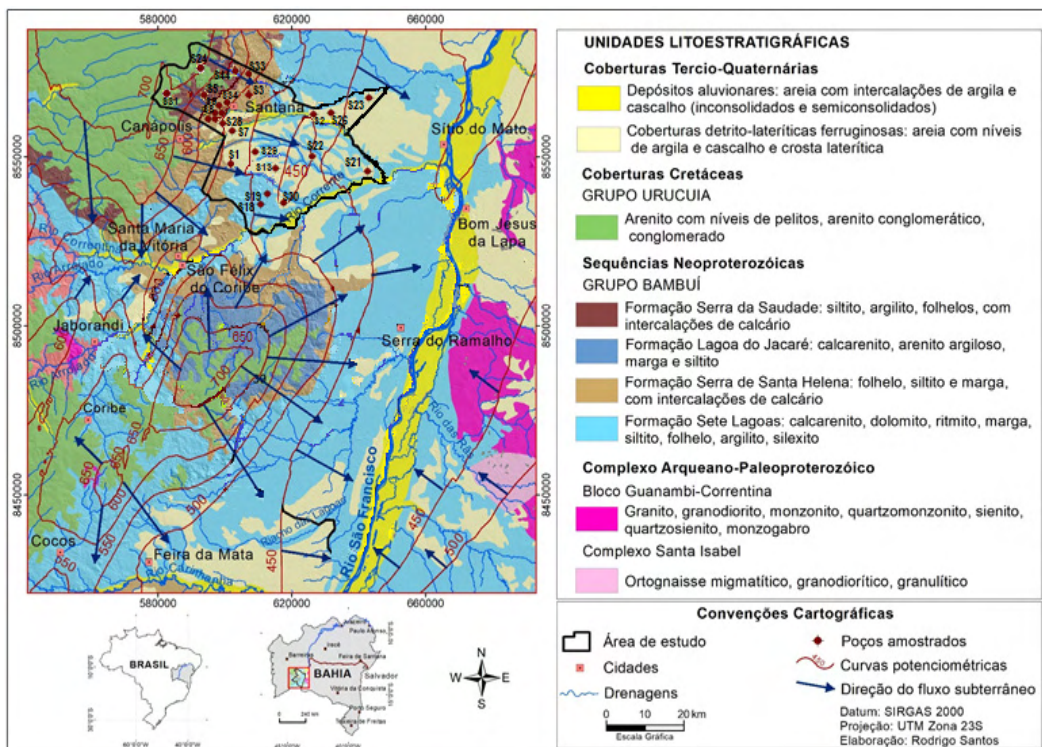


Figure 2 - Simplified geological map and distribution of potentiometric surfaces in the Bambuí Aquifer, Bahia. The spatial indication of the sampling points is also indicated.

Source: Prepared by the authors.

Group emerge, deposited on Archean-Paleoproterozoic rocks of the gneissic-migmatitic basement (MISI et al., 2011) (Figure 2). In this context, there is the fluorite mineral phase disseminated and hosted in the limestones and dolomites of the Sete Lagoas Formation of the Bambuí Group, in western Bahia (MISI et al., 2007). Furthermore, Silva, Viglio and Quintarelli (2020) proposed the megaprovince of occurrence of fluorine in the lower course of the São Francisco River, between Bahia and Minas, where the fluorite mineral phase is associated with pelito-carbonate rocks of the Bambuí Group.

In local geology, the limestones and dolomites of the Sete Lagoas Formation, or the Lagoa do Jacaré Formation, and the pelites (siltites, shales, claystones, slates), with subordinate limestones, from the Serra de Santa Helena Formation or the Serra da Saudade Formation, outcrop. (Figure 2). The presence of detrital and alluvial Tercio-Quaternary covers and the Cretaceous sandstones of the Urucua Group can also be observed on the pelito-carbonate covers of the Bambuí Group. In addition, the tubular wells are located in the pelito-carbonate rocks of the Bambuí Group, which host the Bambuí Aquifer System.

In the regional hydrogeological context, a preferential direction of groundwater flow can be observed, moving from west to east, towards the São Francisco River, considered a regional base level (Figure 2). It is also possible to observe the high topography of Serra do Ramalho, which represents a regional watershed, and a secondary flow direction, which moves from west to southeast, towards the Carinhanha River. Furthermore, in the surroundings of the municipality of Santana, it is observed that the more local flow of groundwater moves to the southeast towards the encounter with the Corrente River, or from west-east-southeast

towards the São Francisco River.

## **HEALTH RESEARCH AND STATISTICAL ANALYSIS: DENTAL FLUOROSIS**

Data on the prevalence and severity of dental fluorosis in the municipality of Santana were compiled from Coutinho's master's research (2014). It included a cross-sectional study of the prevalence and severity of dental fluorosis, which adopted a descriptive design from an epidemiological survey with 159 schoolchildren enrolled in the municipality of Santana, of both sexes and at 12 years of age. The choice of this age group for the study of dental fluorosis is explained because children at 12 years of age have most of their permanent teeth erupted (FEJERSKOV et al., 1994).

Schoolchildren aged 12 years were examined in 2014 by dentist Dr. Carlos Alberto Machado Coutinho, respecting the precepts of ethics in health research. The nature of the research, its objectives, methods, expected benefits, potential risks and inconveniences that it represents for the participants, and all stages of the evaluation of the prevalence and severity of fluorosis were previously informed to those involved in the study with the help of the Free and Informed Consent (ICF) and the Free and Informed Assent Term (TALE), addressed to children, in accordance with Resolution number: 466/2012 of the National Health Council (BRASIL, 2012).

In this context, students were examined in public or private schools, according to the sample selection inclusion criteria: (i) born and residing in the municipality of Santana until the date of the clinical study, according to information provided by the Department of Education ; (ii) the presence of the person in charge at the time of the examination and interview; and (iii) the sample included only children whose parents signed the informed consent.

The sample was obtained from a population of  $\pm 423$  adolescents at 12 years of age, according to information from the IBGE (2010). The sample was representative from the size of 118 individuals, whose estimate was made by simple finite random sampling, without repetitions, with a proportion estimator (prevalence or incidence), with a significance level of 0.05% and prevalence of 0.815, at 12 years old, adopted from the results of the epidemiological research carried out in the north of Minas Gerais by Velásquez et al. (2006).

Oral examinations in schools followed the recommendations of the Dean index, recommended by the World Health Organization (WHO, 1997) (Table 1). They were performed by a calibrated and trained dentist, with the aid of images provided by the National Oral Health Survey of SB Brasil 2010 (BRASIL, 2010), in a school environment with natural light, aided by a spatula and wooden gauze. Furthermore, in the calibration, the agreement of the results was evaluated by

the Kappa statistics (WHO, 1993), until an adequate inter-examiner agreement was found (Kappa = 0.85).

## RESEARCH IN GEOSCIENCES: HYDROCHEMISTRY AND (GEO) STATISTICS

The geosciences approach was based on the analysis and interpretation of integrated hydrochemical data from the doctoral research by Gonçalves (2014) and master's degree by Coutinho (2014), with the aid of descriptive and multivariate statistics. Geochemical data from water samples collected in 52 tubular wells arranged in the rocks of the Bambuí Group (Figures 1 and 2), from three campaigns of the rainy season (2011) (n=20) and dry season (2012 and 2012) were compiled. 2014) in the municipality of Santana. In the 2014 campaign, the largest sample was obtained (n=52) and integrated hydrochemical research and epidemiological research on dental fluorosis. In addition, the ORP, pH, EC and STD variables were

Classification	Value	Diagnostic Criteria
Normal	0	Fluorosis absent. Tooth enamel has usual translucency and semi-vitelliform structure, surface is smooth, polished, light cream color.
Questionable	1	Enamel has a slight difference from normal translucency and occasional whitish stains. Applicable if "normal" classification is not justified.
Very light	2	Small whitish, opaque spots occur, which spread unevenly across the tooth (<25% of the surface). Includes clear opacities between 1 and 2 mm at the tip of molar cusps (nevadas).
Light	3	White spots occur and the opacity is more extensive (< 50% of the surface).
Moderate	4	White spots occur on more than 50% of the tooth surface and wear is observed along with small brown spots. All tooth enamel is affected and areas subject to friction appear worn down. There may be brown or yellowish spots, often disfiguring.
Severe	5	Hypoplasia is generalized and the shape of the tooth itself may be affected. The most obvious sign is the presence of depressions in the enamel, which appears to be eroded. Widespread brown spots.

Table 1 – Criteria and values for classifying teeth with enamel lesions associated with fluorosis by the Dean Index (DEAN, 1934), recommended by the WHO (1997).

Source: Modified from the national oral health survey of Projeto SB Brasil (2010).

measured in situ to obtain the samples, with the aid of a multiparameter probe (Horiba U-50), and aliquots were taken to enable the analyzes in the Plasma Laboratory of Federal University of Bahia (UFBA).

Aliquots were stored in polyethylene (0.5L and 1L) and amber (100 mL) containers, according to APHA guidelines (2005). The 0.5 L aliquots passed through a 0.45  $\mu\text{m}$  cellulose acetate filter and addition of nitric acid until reaching  $\text{pH} < 2$  in the cation analysis ( $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ). While the 1L aliquots were used for anionic analysis ( $\text{CO}_3^{2-}$ ,  $\text{HCO}_3^-$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ) and kept in natura at a temperature of  $\pm 4^\circ\text{C}$ .

Cation contents were measured in duplicate, with 20% triplicates to improve analytical quality, using inductively coupled plasma optical emission spectrometry (ICP-OES). Anions were read using the titrimetric ( $\text{HCO}_3^-$ ,  $\text{Cl}^-$ ) and UV-VIS (Varian) ( $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ) spectrophotometric methods. Furthermore, the reading of fluoride ( $\text{F}^-$ ) was performed by means of colorimetry (SPADNS), with the aid of a fluorimeter (LS 4000).

The statistical approach included the normality test (Shapiro-Wilk) and comparison tests for Tukey-Kramer parametric data (ANOVA) or for non-parametric data (Kruskal-Wallis), with a significance level of 0.05% and the cluster analysis. It is used in cluster analysis of the similarity between individuals to classify the samples hierarchically into groups, admitting all the variables determined for each individual (LANDIM, 2011). The Euclidean distance was chosen as a measure of distance or similarity between the sample points, together with the Ward method, for the analysis of the link between the groups. Ordinary kriging was adopted in the geostatistical approach, with the aid of the ArcGIS 9.0 program.

The saturation index (SI) of the samples

was calculated with the aid of the Diagrammes 6.52 program, where the samples could be grouped as undersaturated classes ( $\text{IS} < -0.5$ ), under chemical equilibrium conditions of the mineral phase with the solution ( $\text{IS}: -0.5$  and  $0.5$ ) and supersaturated ( $\text{IS} > -0.5$ ). Merkel and Planer-Friedrich (2012) suggest the admission of IS values greater than the  $0 \pm 0.5$  interval, due to the uncertainties inherent in the IS calculation, the dissolved mineral equilibrium constant and chemical analyses.

## RESULTS AND DISCUSSION

### FLUORIDE IN GROUNDWATER AND DENTAL FLUOROSIS

Clinical examination revealed that the moderate to severe forms of dental fluorosis were the most critical, the clinical aspects of which are represented in Figure 3 d. They stand out, according to Fejerskov et al. (1994), among the diagnostic clinical aspects of dental fluorosis, the appearance of opaque enamel stains on the dental counterparts and the presence of yellowish or brownish-brown areas in severe alterations. In addition, the appearance of depressions on the tooth surface can be verified in the most severe forms.

The results of the descriptive analysis of the profile of the prevalence and severity of dental fluorosis in 159 schoolchildren at 12 years of age are summarized in Table 2. It was found that 87% of those examined lived in rural areas and that 60% were female, being observed in the interview or in the clinical examination that male students were less inclined to be examined, which configured a bias in obtaining the sample.

In this context, there was a prevalence of dental fluorosis of 53% (Dean Index), where the moderate or severe (severe) forms totaled 17% of those examined (Table 2). This profile of dental fluorosis differed from the result of the national oral health survey of the SB Brasil Project (BRASIL, 2010). This oral health



(A - Normal).



(B - Light).



(C - Moderate).



(D - Severe).

Figure 3 - Clinical aspects of dental fluorosis obtained from the clinical examination of 12-year-old schoolchildren living in the municipality of Santana, in the west of the State of Bahia, Brazil.

Source: Obtained from Coutinho's dissertation research (2014).

Fluorosis	Man		Woman		Total	
	N	%	n	%	n	%
Absent (0)	35	22.0	38	24.0	73	46.0
Questionable (1)	4	2.5	5	3.0	9	5.5
Very light (2)	11	7.0	17	11.0	28	18.0
Light (3)	5	3.0	17	11.0	22	14.0
Moderate (4)	5	3.0	10	6.0	15	9.0
Severe (5)	4	2.5	8	5.0	12	7.5
<b>Total</b>	<b>64</b>	<b>40.0</b>	<b>95</b>	<b>60.0</b>	<b>159</b>	<b>100.0</b>

Table 2 - Absolute and relative frequency of the categories indicated by the Dean index in a sample of 12-year-old schoolchildren in the city of Santana, Western Bahia.

Source: Prepared by the authors based on data from Coutinho's dissertation (2014).



survey revealed a national mean prevalence of dental fluorosis of 17% at 12 years of age (Dean index), with 15% very mild or mild and 1.5% moderate or severe, where the lowest prevalence of fluorosis was obtained in the North region (10%) and the largest in the Southeast region (19%).

## HYDROGEOCHEMISTRY OF FLUORIDE AND DENTAL FLUOROSIS

In the hydrogeochemical context, the samples were classified as calcium bicarbonate waters ( $\text{HCO}_3^- - \text{Ca}^{2+}$ ) (40%), calcium mixed waters (mixed -  $\text{Ca}^{2+}$ ) (20%), sodium bicarbonate waters ( $\text{HCO}_3^- - \text{Na}^+$ ) (27%) and chlorinated waters sodium ( $\text{Cl}^- - \text{Na}^+$ ) (13%) (Figure 4). The uncertainty of the ionic balance was at the maximum of 20%, based on practical error (LOGAN, 1965). In the  $\text{HCO}_3^- - \text{Ca}^{2+}$  or mixed -  $\text{Ca}^{2+}$  facies, the ion contents were, in decreasing order:  $r\text{Ca}^{2+} > r\text{Na}^+ > r\text{Mg}^{2+} > r\text{K}^+$  and  $r\text{CO}_3^{2-} > r\text{HCO}_3^- > r\text{Cl}^- > r\text{SO}_4^{2-} > r\text{F}^- > r\text{NO}_3^-$ . While in the hydrochemical facies  $\text{HCO}_3^- - \text{Na}^+$  and  $\text{Cl}^- - \text{Na}^+$ , the contents of the cations found were distributed, in decreasing order:  $r\text{Na}^+ > r\text{Ca}^{2+} > r\text{Mg}^{2+} > r\text{K}^+$ .

Cluster analysis allowed the grouping of samples and the integration of dental fluorosis and hydrogeochemical research ( $n=32$ ), and the assessment of water potability, with the aid of visual observation of the dendrogram (Figure 4). The cut line was marked on the dendrogram at a distance of 10, forming three Integrating Groups (G1, G2, G3a and G3b). Groups G1 and G2 showed the highest average proportions of dental fluorosis prevalence and can be distinguished by proportions of moderate to severe dental fluorosis and fluoride levels. Furthermore, a relationship was found between the prevalence and severity of dental fluorosis and fluoride levels in groundwater samples

from Santana (Table 3).

Median fluoride levels ranged from 0.47 (Integrating Group G3a) to 3.67 mg. L<sup>-1</sup> (Integrating Group G1) (Table 3). It was verified that the levels of fluoride in the groundwater of Santana were from 0.07 to 8.88 mg. L<sup>-1</sup>, with a Median of 0.54 mg. L<sup>-1</sup> (Table 4), where 42% of the samples, those from Groups G1 and G2 exceeded the local optimum potability limit for fluoride (0.78 mg. L<sup>-1</sup>). The local optimum potability limit for fluoride (C) in Santana groundwater was obtained from the Average regional air temperature (T), according to Galagan and Vermillion (1957) (Equations 1 and 2).

$$\epsilon(T) = 10,3 + 0,725T \quad (\text{Equation 1})$$

$$C = 22,2/\epsilon \quad (\text{Equation 2})$$

Table 4 presents a statistical summary compared between the fluoride levels obtained in the samples from Santana (current research) or in the groundwater of its neighboring municipalities, in Western Bahia, according to data from selected wells from SIAGAS (CPRM). Fluoride levels were higher than the local optimum potability limit (0.8 mg. L<sup>-1</sup>) in samples from Western Bahia, especially in the municipalities of Santana (43%), São Félix do Coribe (53%) and Serra Dourada (45%), where prolonged ingestion of these waters can threaten the fundamental right to health. Median fluoride levels in groundwater differed between samples from Santana and its neighbors, according to the Kruskal-Wallis test ( $p=0.0003$ ) (Figure 6).

In this context, the presence of natural and toxic levels of fluoride in the municipalities of Western Bahia was made explicit, on a more regional scale, whose consumption of these waters represents a risk factor for oral and systemic health, and the emergence of dental fluorosis in the city of Santana. Velasquez et al. (2006), Costa et al. (2013), Coutinho (2014) and Cruz et al. (2015) studied dental fluorosis in school-age children in Northern Minas

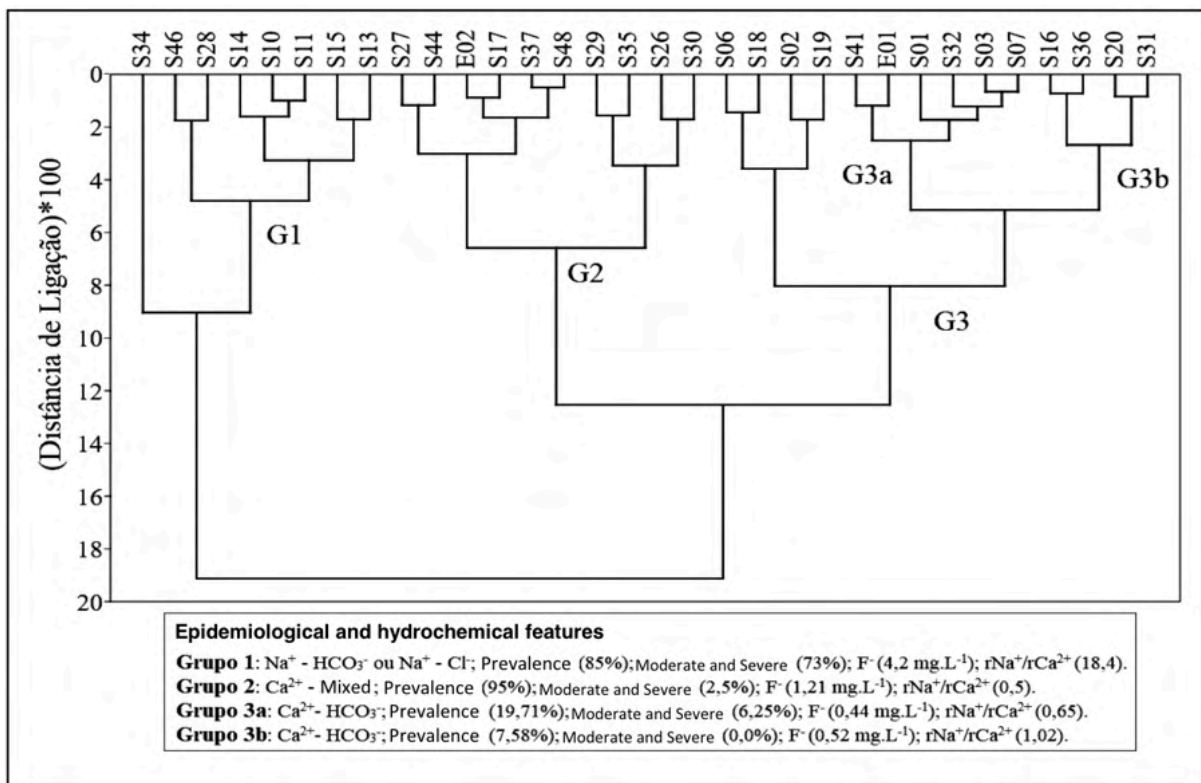


Figure 4 - Dendrogram of integration of the epidemiological classification of fluorosis and the hydrochemistry of Santana.

Source: Prepared by the authors based on data from Coutinho (2014) and Gonçalves (2014).

Point	Degrees			pH	mg. L <sup>-1</sup>					rNa/ rCa	rHCO <sub>3</sub> / rCa	HCO <sub>3</sub> / DT	
	Prevalence	2 e 3 <sup>A</sup>	4 e 5 <sup>B</sup>		STD	Ca <sup>2+</sup>	Na <sup>+</sup>	Cl <sup>-</sup>	F <sup>-</sup>				DT
Grupo 1 (G1)													
S34	50.00	50.00	50.00	8.78	1683.50	4.00	353.60	478.76	8.88	7.21	76.92	30.16	51.04
S28	82.00	35.00	47.00	7.60	650.00	96.00	167.20	68.00	1.88	138.00	1.52	1.35	2.86
S46	50.00	25.00	25.00	8.40	720.75	32.00	107.52	87.50	2.40	133.76	2.92	2.71	1.98
S14	100.00	0.00	100.00	8.27	477.00	22.00	97.40	61.78	3.31	71.46	3.85	3.74	3.51
S10	100.00	0.00	100.00	8.34	590.83	21.47	161.80	121.15	4.03	97.30	6.56	3.74	2.52
S11	100.00	0.00	100.00	7.97	712.83	12.93	136.79	90.03	2.76	51.50	9.21	6.80	5.21
S15	100.00	0.00	100.00	8.57	799.90	5.67	191.68	152.57	4.88	25.87	29.42	14.40	9.63
S13	100.00	40.00	60.00	8.65	670.23	14.01	263.48	71.10	5.22	35.34	16.36	9.37	11.33
<b>Average</b>	<b>85.25</b>	<b>18.75</b>	<b>72.75</b>	<b>8.32</b>	<b>788.13</b>	<b>26.01</b>	<b>184.93</b>	<b>141.36</b>	<b>4.17</b>	<b>70.06</b>	<b>18.35</b>	<b>9.03</b>	<b>11.01</b>
Median	100.00	12.50	80.00	8.37	691.53	17.74	164.50	88.77	3.67	61.48	7.88	5.27	4.36
Grupo 2 (G2)													
S27	100.00	100.00	0.00	8.30	460.00	119.00	62.33	98.34	1.25	211.10	0.46	0.63	1.08
S44	100.00	100.00	0.00	8.30	611.00	183.00	45.53	55.00	0.60	289.00	0.22	0.16	0.31
S17	100.00	100.00	0.00	7.72	553.33	99.89	73.33	88.57	1.29	228.23	0.64	1.06	1.41
E02	100.00	100.00	0.00	7.82	619.58	63.21	35.22	91.00	0.54	264.00	0.48	1.48	1.08
S37	100.00	100.00	0.00	7.75	262.00	63.42	75.69	82.30	1.60	265.10	1.04	1.35	0.99
S48	100.00	100.00	0.00	7.75	361.00	60.00	62.33	93.00	1.25	250.80	0.90	1.56	1.14

S35	100.00	100.00	0.00	7.91	559.00	186.00	148.00	58.70	1.82	289.00	0.69	0.73	1.44
S29	100.00	75.00	25.00	7.20	528.45	197.00	63.47	40.52	1.28	261.29	0.28	0.39	0.89
S26	50.00	50.00	0.00	7.10	812.50	290.00	27.59	40.32	0.74	378.64	0.08	0.35	0.82
S30	100.00	100.00	0.00	7.25	812.50	303.00	80.27	48.19	1.72	380.00	0.23	0.42	1.02
<b>Average</b>	<b>95.00</b>	<b>92.50</b>	<b>2.50</b>	<b>7.71</b>	<b>557.94</b>	<b>156.45</b>	<b>67.38</b>	<b>69.59</b>	<b>1.17</b>	<b>281.72</b>	<b>0.50</b>	<b>0.81</b>	<b>1.02</b>
Median	100.00	100.00	0.00	7.75	556.17	151.00	62.90	70.50	1.27	264.55	0.47	0.68	1.05
Grupo 3a (G03a)													
S02	0.00	0.00	0.00	7.26	476.50	217.12	22.58	61.66	0.26	514.44	0.09	0.43	0.56
S19	0.00	0.00	0.00	8.08	527.00	249.00	33.95	47.86	0.53	324.73	0.12	0.33	0.77
S06	50.00	50.00	0.00	7.62	741.37	158.05	25.47	125.35	0.46	416.65	0.14	0.55	0.63
S18	100.00	50.00	50.00	7.54	620.50	184.80	48.97	140.53	0.86	414.09	0.23	0.45	0.61
S20	0.00	0.00	0.00	8.19	424.23	92.42	16.14	32.42	0.07	284.67	0.15	0.93	0.92
S31	7.70	7.70	0.00	8.22	500.00	130.00	37.51	46.65	0.57	175.00	0.25	0.74	1.68
E01	0.00	0.00	0.00	7.60	582.17	62.31	33.33	86.30	0.29	260.45	0.47	1.38	1.01
S16	0.00	0.00	0.00	7.77	568.33	7.66	32.93	32.10	0.47	31.57	3.74	14.32	10.60
<b>Average</b>	<b>19.71</b>	<b>13.46</b>	<b>6.25</b>	<b>7.79</b>	<b>555.01</b>	<b>137.67</b>	<b>31.36</b>	<b>71.61</b>	<b>0.44</b>	<b>302.70</b>	<b>0.65</b>	<b>2.39</b>	<b>2.10</b>
Median	0.00	0.00	0.00	7.70	547.67	144.03	33.13	54.76	0.47	304.70	0.19	0.64	0.85
Grupo 3b (G03b)													
S36	12.50	12.50	0.00	7.40	600.00	133.00	45.15	33.82	0.80	393.00	4.79	10.46	0.54
S41	0.00	0.00	0.00	7.10	487.50	125.36	36.37	29.58	0.57	250.72	0.25	0.51	0.77
S01	33.00	33.00	0.00	7.62	557.50	134.53	30.06	95.85	0.51	360.03	0.19	0.59	0.67
S32	0.00	0.00	0.00	7.12	566.00	136.00	70.90	129.00	0.37	220.00	0.45	0.36	0.68
S03	0.00	0.00	0.00	7.42	562.75	125.69	33.73	79.60	0.35	329.35	0.23	0.66	0.76
S07	0.00	0.00	0.00	7.33	728.43	130.51	33.43	119.92	0.51	344.81	0.22	0.55	0.64
<b>Average</b>	<b>7.58</b>	<b>7.58</b>	<b>0.00</b>	<b>7.33</b>	<b>583.70</b>	<b>130.85</b>	<b>41.61</b>	<b>81.30</b>	<b>0.52</b>	<b>316.32</b>	<b>1.02</b>	<b>2.19</b>	<b>0.68</b>
Median	0.00	0.00	0.00	7.37	564.38	131.76	35.05	87.73	0.51	337.08	0.24	0.57	0.67

<sup>A</sup> severity: mild to very mild; <sup>B</sup> Severity: moderate to severe (most critical situation).

Table 3 - Group obtained in the cluster analysis that integrate the results of surveys on the prevalence and severity of dental and hydrogeochemical fluorosis in the city of Santana.

Source: Prepared by the authors based on data from Coutinho (2014) and Gonçalves (2014).

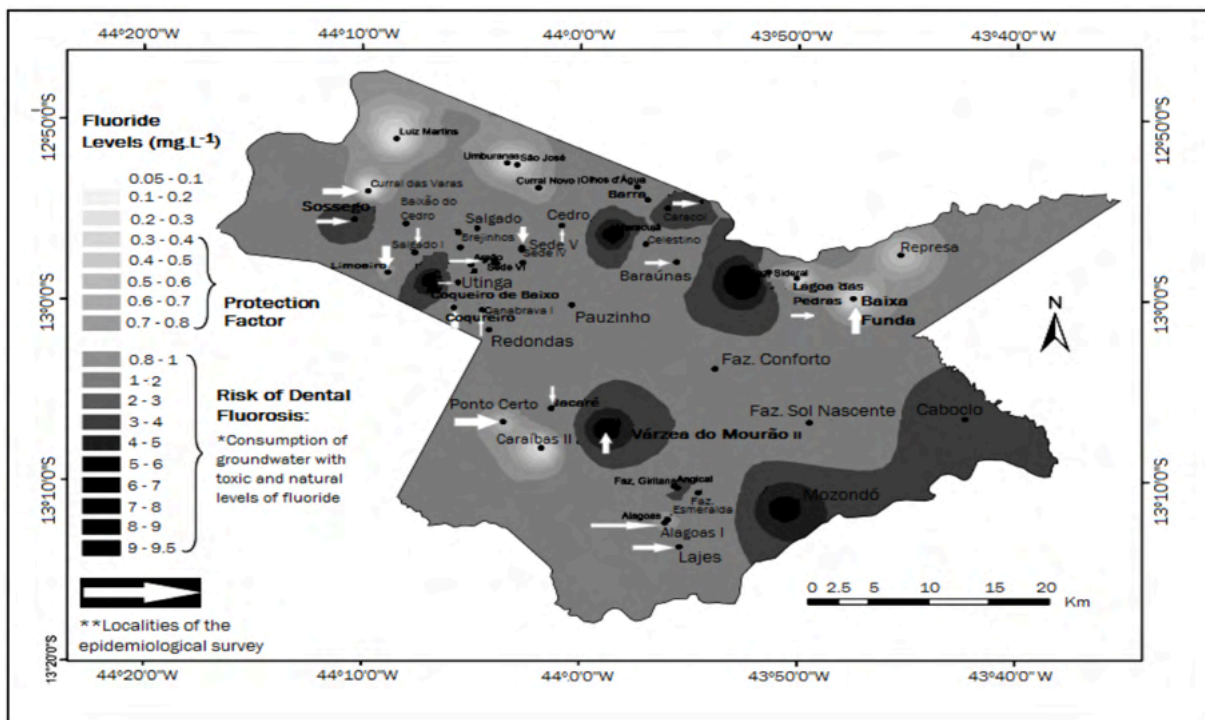


Figure 5 - Distribution map of fluoride levels in groundwater and indication of protection factor categories (0.4-0.8 mg. L-1) and risk of dental fluorosis (> 0.8 mg. L- 1).

Source: Prepared by the authors based on data from Coutinho (2014) and Gonçalves (2014).

City	Size (n)	Minimum	Maximum	Average	Median	Standard deviation	Standard Error	CV (%)	SW (p value)	F-> 0,8 mg. L <sup>-1</sup>
Santana (Atual)	52	0,05	8,88	1,36	0,54	1,78	0,25	130,58	$p < 0,0001^B$	42 (%)
S. F. do Coribe <sup>C</sup>	15	0,38	8,13	1,68	0,93	2,04	0,53	121,92	$p = 0,009^B$	53 (%)
Serra Dourada <sup>C</sup>	40	0,06	5,20	1,19	0,78	1,20	0,19	100,43	$p = 0,0004^B$	45 (%)
Baianópolis <sup>C</sup>	26	0,03	4,80	0,58	0,27	1,02	0,20	175,64	$p < 0,0001^B$	12 (%)
Sítio do Mato <sup>C</sup>	12	0,16	13,89	2,45	0,74	3,97	1,15	161,95	$p = 0,0002^B$	33 (%)
Sta. Ma. da Vitória <sup>C</sup>	53	0,02	4,74	0,84	0,52	0,87	0,12	103,21	$p < 0,0001^B$	30 (%)
Serra do Ramalho <sup>C</sup>	10	0,10	1,95	0,56	0,33	0,60	0,19	106,86	$p = 0,013^B$	30 (%)
Canápolis <sup>C</sup>	38	0,14	7,00	0,85	0,65	1,10	0,18	129,20	$p < 0,0001^B$	29 (%)
Serra do Ramalho <sup>C</sup>	10	0,10	1,95	0,56	0,33	0,60	0,19	106,86	$p = 0,013^B$	30 (%)

CV: Coefficient of variation; SW: Shapiro-Wilk; <sup>A</sup> Gaussian distribution; <sup>B</sup> non-Gaussian distribution; <sup>C</sup> synthesis of data from the register of selected wells from SIAGAS/CPRM.

Table 4 - Comparative statistical summary of fluoride levels in groundwater in Santana (current) and in the groundwater of the municipal neighbors (SIAGAS), Bahia.

Source: Prepared by the authors based on data from Coutinho (2014), Gonçalves (2014) and the register of wells in the Groundwater Information System (SIAGAS/CPRM).

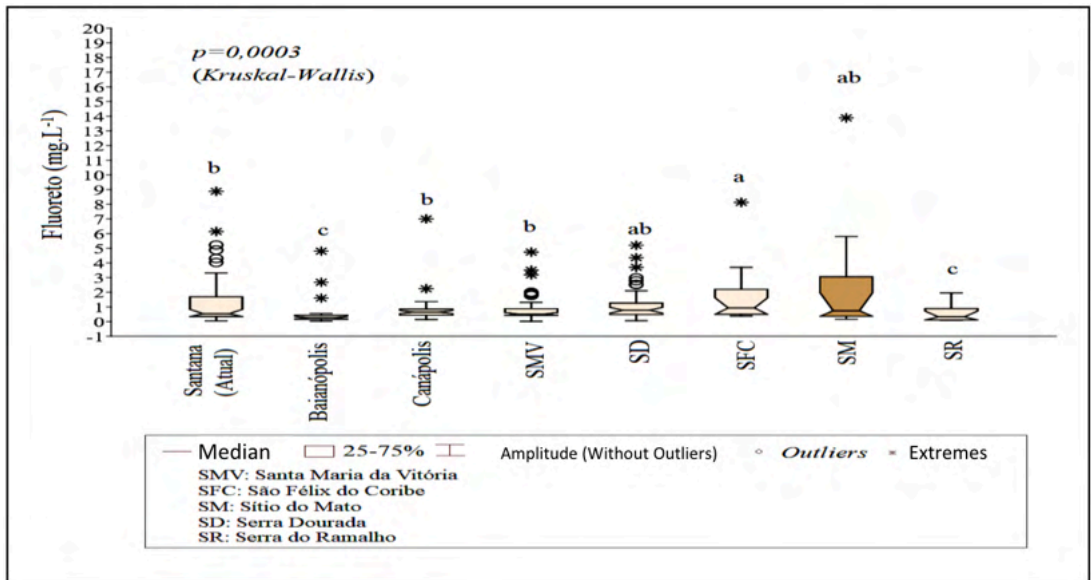


Figure 6 - Box-plot diagram applied to analyze the dispersion of fluoride values in groundwater samples from Santana and neighboring municipal districts, located in the hydrogeological and hydrogeochemical context of the Bambuí Aquifer, in Western Bahia, Brazil.

Source: Prepared by the authors based on data from Coutinho (2014), Gonçalves (2014) and the register of wells in the Groundwater Information System (SIAGAS/CPRM).

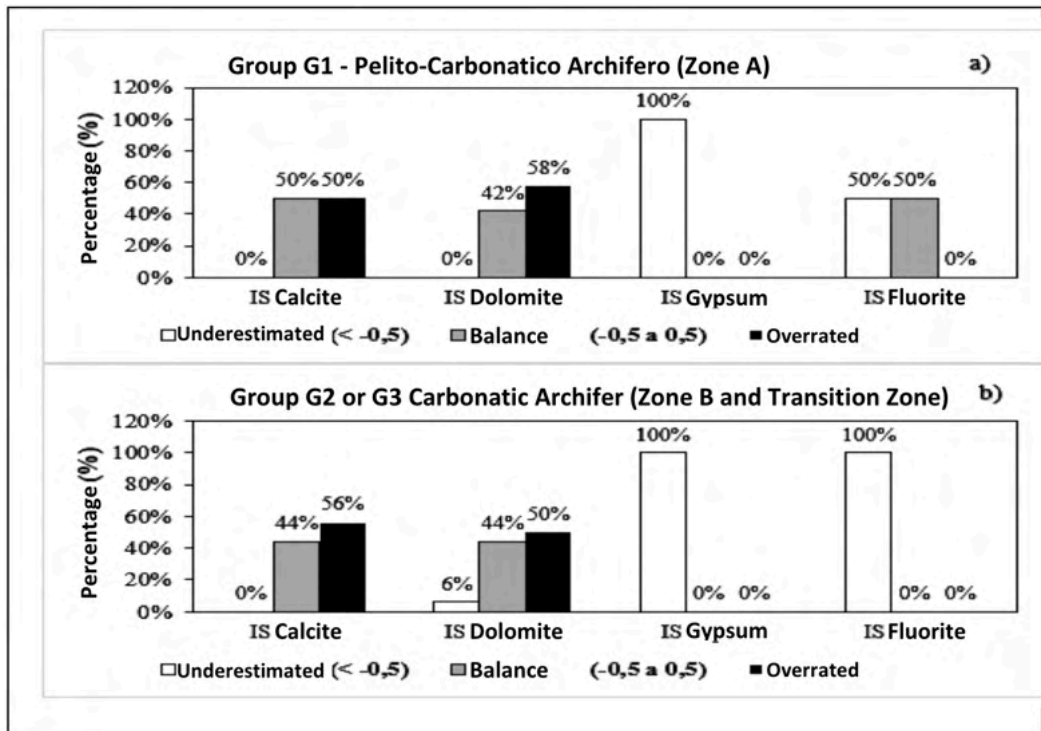


Figure 7 - Distribution diagram of the percentages of saturation indexes of the mineral phases calcite, dolomite, gypsum and fluorite in groundwater samples. A: Hydrochemical Zone A; B: Refers to the Hydrochemical Zone B or the Hydrochemical Transition Zone.

Source: Prepared by the authors based on data from Gonçalves (2014).

Gerai and Western Bahia. The proportions of the prevalence of dental fluorosis, and the severity, in Bahia and Minas Gerais, resulted, to a large extent, from the chronic exposure of children aged 0 to 6 years by the ingestion of water from the Bambuí Aquifer with natural and toxic levels of fluoride.

Cangussu et al. (2002), Narvai (2002), Souza et al. (2013) point out that higher proportions of dental fluorosis, in moderate or severe forms, are expected in endemic areas. This is because in fluorosis-endemic areas, children at the age of formation of the tooth germ may be exposed by drinking water with natural and toxic levels of fluoride, and cultural and socio-spatial, climatic and ecological-environmental ones.

It is noteworthy that the Integrating Group G1 presented the greatest relevance for understanding dental fluorosis and its relationship with fluoride levels in groundwater (Table 3), which represent a risk factor for dental fluorosis for 42% of the samples and bone fluorosis for 21% of samples. Nirgude et al. (2010) point out that chronic exposure through the ingestion of natural waters with fluoride levels that exceed 3.0 mg. L-1 represents a risk factor for the prevalence of dental fluorosis and/or bone fluorosis.

The G1 Integrator Group exhibited the most notable proportions of dental fluorosis and a moderate to severe degree of severity (80%), the most mineralized waters (STD Median: 691.93 mg. L-1) and the most pronounced F- values. (Median of 4.17 mg. L-1) and the geochemical ratios  $rNa+/rCa^{2+}$  and  $rHCO_3-/rCa^{2+}$  (Table 3). It is characterized, in summary, by the presence of sodium waters, which are more alkaline and enriched in Na+ and F-, by the low levels of Ca<sup>2+</sup> and DT and by the greater epidemiological relevance in the investigation of fluorosis.

The G2 Integrator Group revealed a high prevalence of dental fluorosis (100%),

distributed between very mild to mild degrees of severity (100%), fluoride levels that generally exceed the optimal local potability limit, but much lower than levels observed in Group G1 (Table 3). While Group G3 had the least expressive prevalence of dental fluorosis, between very mild to mild degrees of severity, low fluoride levels did not exceed the local optimal limit of fluoride potability. It can be noted that the samples from the Integrating Groups G2 and G3 (G3a and G3b) were characterized by calcic waters, rich in calcium and with the highest levels of total hardness (DT), especially in terms of carbonate hardness.

Based on the interpretation of the integration of fluorosis and geosciences research, from the paradigmatic perspective of Human Ecology, which includes Medical Geology, Medical Ecology and Medical Geography, a theoretical-conceptual model that pointed out two hydrogeochemical zones and related them to the distribution of fluoride in natural waters and to dental fluorosis. It follows, therefore, that the Hydrochemical Zone A would be the one characterized by sodic waters, more mineralized, enriched in F- and Na+ and depleted in Ca<sup>2+</sup>, which would refer to the G1 Group, whose aquifer units are hosted in pelito-carbonate rocks of the Bambuí Group (Pelitic-Carstic Aquifer).

Hydrochemical zone B comprised the G3 Group, characterized by calcium bicarbonate waters, rich in Ca<sup>2+</sup>, depleted in F- and Na+, whose aquifer units are hosted in the carbonate rocks of the Bambuí Group (Karstic Aquifer) (Table 3). The samples of Group G2, defined by the mixed-calcic waters, constitute the Transition Zone. This Hydrochemical Zone is distinguished from the zone B in that it exhibits F- levels that exceed the local optimum potability limit and higher relative Na+ ionic activity. Furthermore, the levels of calcium and magnesium, and the hardness

of carbonates, can be uncomfortable for the consumption of water in zones B and Transition, which can be an important protective factor against dental fluorosis.

In the context of the theoretical-conceptual model of hydrogeochemical zones, based on the doctoral research by Gonçalves (2014), it is noteworthy that the values of the  $r\text{HCO}_3^-/r\text{Ca}^{2+}$  ratio were lower than 1.5 in the Integrating Groups G2 and G3, reporting the relevance of the water-carbonate rock interaction (Table 3). Roisenberg et al. (2003) highlighted that the values of the  $r\text{HCO}_3^-/r\text{Ca}^{2+}$  ratio  $< 1.5$  indicate the role of the dissolution of calcite and dolomite and the subsaturation of the solution in terms of these minerals in the hydrochemistry. It informs about the influence of water transit and water-rock interaction in the Bambuí Aquifer. The hydrogeochemical evolution of karst aquifers depends on the dissolution of calcite and dolomite minerals, climate, transit time and water circulation in the aquifer (FAIRCHILD et al., 2000; SANTOS, 2017).

Another situation was observed in the G1 Integrating Group, whose  $r\text{HCO}_3^-/r\text{Ca}^{2+}$  ratio values  $> 1.5$ , being, therefore, attributed to the water-rock interaction process, where the chemical weathering of the minerals present in the pelitic rocks, judging by the sodium plagioclases, would add bicarbonate and lithogenic sodium ions to the water and contribute to the hydrochemistry. In turn, the samples from the G1 Integrator Group are characterized by the presence of the most expressive values of the  $r\text{HCO}_3^-/r\text{Ca}^{2+}$  and  $r\text{Na}^+/r\text{Ca}^{2+}$  ratios.

The geochemical modeling also included the calculation of the Saturation Index (SI) of the fluorite, calcite, dolomite and gypsum mineral phases (Figure 7). With this, it was verified that the values of the fluorite IS (fluorite IS) differed between the samples of the Integrator Group 1 (G1), associated

with the Hydrochemical Zone A, and those of the Integrating Groups G2 or G3, related respectively to the Hydrochemical Zone B and the Hydrochemical Transition Zone.

It was observed that the value of the Median of the  $\text{IS}_{\text{fluorite}}$  of the Integrator Group G1 approached the equilibrium condition between the mineral phase and the solution, while the values of the Medians of the  $\text{IS}_{\text{fluorite}}$  of Integrating Groups G2 or G3 suggested a fluorite undersaturation condition (Table 5). It is also noted that the samples proved to be undersaturated in terms of the anhydrite, gypsum or halite mineral phases, although they varied from the equilibrium condition with the solution to supersaturation (reports to a tendency for the mineral to precipitate) to the mineral phases calcite and dolomite.

The hematite values reflect a variation of conditions from subsaturation to supersaturation, where samples from the zone B (G2) or from the Transition Zone (G3) revealed a tendency of mineral precipitation, ie, supersaturation (Table 5). Furthermore, it was found that the IS values  $\text{hematite}$  of the Hydrochemical Zone A vary between subsaturation and supersaturation of the samples. As a result, the IS value  $\text{hematite}$  may contribute to the differentiation of samples between Zone A and Zone B or Transition Zone. The origin of iron oxides and hydroxides, such as the hematite, was attributed to the impurities present in the carbonate rocks of the Bambuí Group or to the iron oxides and hydroxides present in the sandstones of the Urucuia Group (see Figure 2).

The evolution of the hydrogeochemical composition involves the dissolution of calcite and dolomite from the limestones and dolomites of the Bambuí Group, which supplies  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  ions to the solution and influences the solubility of other minerals, such as fluorite (Equations 3 to 5). Apambire et al. (1997) emphasize that the saturation

a) Samples from the G1 Group - Pelito-Carbonitic Aquifer (Zone A).

Indexes	Minimum	Maximum	Average	Median	Desvio Padrão	Standard Error	Coefficient of variation (%)	SW (value of p)
IS <sub>Aragonita</sub>	-0,07	0,88	0,38	0,32	0,34	0,09	89,19	$p = 0,21^A$
IS <sub>Calcita</sub>	0,07	1,02	0,55	0,46	0,34	0,10	62,02	$p = 0,25^A$
IS <sub>Dolomita</sub>	-0,16	2,12	0,94	1,02	0,75	0,22	79,83	$p = 0,59^A$
IS <sub>Fluorita</sub>	-0,91	-0,06	-0,53	-0,51	0,30	0,09	56,99	$p = 0,36^A$
IS <sub>Gipsita</sub>	-4,76	-2,48	-3,17	-3,06	0,60	0,17	19,04	$p = 0,24^A$
IS <sub>Anidrita</sub>	-3,79	-2,74	-3,19	-3,19	0,32	0,09	10,16	$p = 0,51^A$
IS <sub>Halita</sub>	-8,39	-5,73	-6,43	-6,30	0,65	0,17	10,06	$p = 0,002^B$
IS <sub>Hematita</sub>	-8,37	16,45	2,67	-0,96	9,03	2,41	338,56	$p = 0,025^B$

<sup>A</sup> Gaussian distribution; <sup>B</sup> Non-Gaussian distribution; SW: Shapiro-Wilk test.

b) Samples from Groups G2 or G3 - Carbonatic Aquifer (Zone B or Transition Zone).

Indexes	Minimum	Maximum	Average	Median	Standard deviation	Standard Error	Coefficient of variation (%)	SW (value of p)
IS <sub>Aragonita</sub>	0,05	2,68	0,52	0,42	0,56	0,13	109,32	$p < 0,0001^B$
IS <sub>Calcita</sub>	0,19	1,01	0,54	0,56	0,21	0,05	39,85	$p = 0,60^A$
IS <sub>Dolomita</sub>	-0,06	1,47	0,54	0,48	0,44	0,10	81,83	$p = 0,19^A$
IS <sub>Fluorita</sub>	-2,23	-0,91	-1,69	-1,81	0,36	0,09	21,50	$p = 0,38^A$
IS <sub>Gipsita</sub>	-2,02	-1,44	-1,71	-1,70	0,17	0,04	9,81	$p = 0,82^A$
IS <sub>Anidrita</sub>	-2,28	-1,71	-1,98	-1,97	0,04	0,03	-8,70	$p < 0,60^A$
IS <sub>Halita</sub>	-7,89	-4,46	-7,09	-7,10	0,71	0,16	10,05	$p < 0,0001^B$
IS <sub>Hematita</sub>	4,02	20,82	14,15	15,35	4,18	0,96	29,56	$p = 0,002^B$

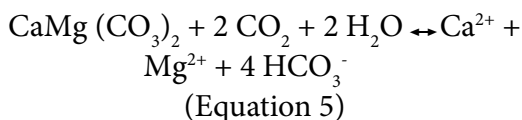
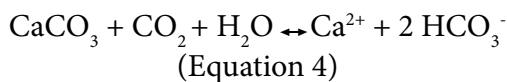
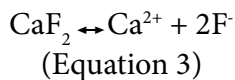
<sup>A</sup> Gaussian distribution; <sup>B</sup> Non-Gaussian distribution; SW: Shapiro-Wilk test.

Table 5 - Statistical summary of the calculated values of the Saturation Indexes of the mineral phases calcite, dolomite, anhydrite, gypsum, halite and fluorite in the groundwater samples, differentiated according to the conceptual model in the category of Hydrochemical Zone A (Group 1) and in the category that includes the Hydrochemical Zone B or Transition Zone (Groups 2 and 3).

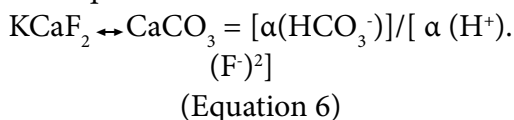
Source: Prepared by the authors based on data from Gonçalves (2014).



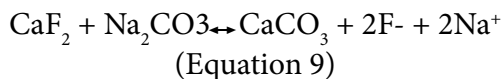
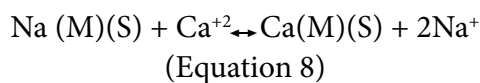
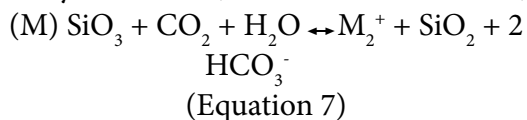
of the solution, water, in the calcite mineral phase controls the solubility of fluorite, due to the common ion effect and the abundance of calcite in the water. Furthermore, Rao (2011) emphasizes that the interdependence of the activities of Ca<sup>2+</sup>, F<sup>-</sup> and HCO<sub>3</sub><sup>-</sup> ions in groundwater must be considered. This author considered that the dissolution of fluorite could occur under conditions of subsaturation of the solution in calcite and dolomite, in more alkaline waters, under high air temperatures and that there is coprecipitation of Ca<sup>2+</sup> and HCO<sub>3</sub><sup>-</sup> ions.



An inverse correlation was observed between the activities of fluoride and calcium in groundwater, which corroborated the proposition that saturation of the solution in calcite would control the dissolution of fluorite. Handa (1975) proposed a geochemical conceptual model for the enrichment of water in fluorine, which elucidated the correlation between F<sup>-</sup>, Ca<sup>2+</sup> and HCO<sub>3</sub><sup>-</sup> ions, under relatively constant pH conditions (Equation 6), where K and α are called respectively the solubility constant and the ionic activity. Gonçalves et al. (2018) proposed that Handa's (1975) conceptual model, after being adapted, would contribute to the geochemical study of fluorine in the Bambuí Aquifer.



It was inferred, from the hydrochemical analyses, of the rNa<sup>+</sup>/rCa<sup>2+</sup> ratio and the IS values of calcite and dolomite, that the activity of the Na<sup>+</sup> ion in Santana's groundwater contributes to the increase of alkalinity and to the dissolution of fluorite, or suggest presence of fluorine geochemical anomaly associated with clay minerals, and pelitic rocks. It is noteworthy that in the interaction of water and clay minerals (M), originated from the chemical weathering of impure carbonates and pelites, Ca<sup>2+</sup> is removed, DT is reduced, and Na<sup>+</sup> is added to the solution from base exchange reactions (Equations 7-9). The activity of sodium increases the alkalinity of the water and interferes in the conditions of saturation, precipitation of calcite and the solubility of fluorite (APAMBIRE et al., 1997).



Costa (2011) proposed that chemical weathering of plagioclases could make lithogenic sodium ions available to the solution, thereby promoting the enrichment of the waters of the Bambuí Aquifer, in Minas Gerais. In this study, fluoride contents were obtained in the pelite between 120.0 and 620.0 mg. L<sup>-1</sup> and in the carbonate rocks from 320.0 to 508.0 mg. L<sup>-1</sup>. Thus, the presence of fluorine levels in the rocks of the Bambuí Group, in Minas Gerais, was verified, which exceeded 300.0 mg. L<sup>-1</sup>, considering the average level of fluorine obtained in carbonates proposed by Krauskopf and Bird (1995) and Mendes and Oliveira (2004).

In view of the proposition of a geogenic

origin of fluorine in the Waters of the Bambuí Aquifer, which is relevant for the promotion of oral health, it is noted that mineral deposits and discontinuous extraction of purple fluorite associated with the rocks of the Bambuí Group, in Western Bahia, have been known since 1960, highlighting the work of Miranda (1976), Misi et al. (2000), Conceição Filho et al. (2003), Martins (2001) and Gomes (2005). Silva, Viglio and Quintarelli (2020) suggested the existence of a megaprovince of fluoride occurrence and prevalence of endemic fluorosis in the middle course of the São Francisco River, which extends from the north of the State of Minas Gerais to the west of Bahia, Brazil.

Furthermore, the fluoride anomalies associated with the mudstones of the Bambuí Group demand an understanding of the regional geological history and investigations into the contribution of weathering of igneous rocks in the origin of sediments and the high levels of fluoride in this geological material. Enalou et al. (2018), Dehbandi et al. (2018) and Singh et al. (2018) highlight that high levels of fluoride in groundwater are generally related to chemical weathering of minerals, igneous rocks and clay minerals.

In this sense, the application of cluster analysis contributed to the explanation of the existence of a geochemical control of the distribution of fluoride in the groundwater of Santana, associated with the Bambuí Aquifer, which have epidemiological relevance to the understanding and prevention of endemic fluorosis and the promotion of oral and systemic health of the population of Western Bahia.

## CONCLUSIONS

A prevalence of dental fluorosis of 53% of schoolchildren examined at 12 years of age was obtained, with a proportion of 17% distributed from moderate to severe degrees

in the municipality of Santana, in western Bahia, Brazil. These proportions of prevalence and severity of dental fluorosis were related, to a large extent, to the consumption of groundwater with natural and toxic levels of fluoride. supported by approaches to medical geology and health, and in the training of professionals who consider health in its relations with the environment and society.

The presence of fluoride potability restrictions was verified in 42% of the groundwater samples from Santana, regarding the local optimal limit, whose consumption of these waters represents a risk factor for dental fluorosis. Fluoride levels were observed to represent skeletal health risks for 21% of the samples, as well as being related to the risk of knee and hip deformities or disabling fluorosis. Risk profiles of endemic fluorosis were also observed in the municipalities of Canápolis, Santa Maria da Vitória, São Félix do Coribe, Sítio do Mato and Serra Dourada.

The application of multivariate analysis contributed to the integration of health and geoscience research and to the proposition of the conceptual model that grouped the samples of the Integrator Group G1 in the Hydrochemical Zone A, characterized by the presence of sodic waters, enriched in fluoride and sodium, and less hard, than when comparing the samples of the Integrating Groups G2 and G3, which contemplated, respectively, the categories called Hydrochemical Zone B and Hydrochemical Zone of Transition.

It is concluded that the application of cluster analysis contributed to the understanding of the existence of a geochemical control of the distribution of fluoride levels in groundwater in the municipality of Santana, within the Bambuí Aquifer, as well as pointing out that the profile of dental fluorosis in this municipality would be suggestive of an

endemic area. This research revealed relevance to the understanding of endemic fluorosis in the municipality of Santana and its municipal neighbors, in the west of Bahia. Furthermore, it explained the importance of adopting interdisciplinary approaches, such as Human Ecology and Medical Geology, to the study of the multiple relationships between environment and health.

## THANKS

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## REFERENCES

- AOBA, T.; FEJERSKOV, O. Dental fluorosis: chemistry and biology. **Critical Reviews in Oral Biology & Medicine**, v.13, n.2, p.155-170, 2002. DOI: 10.1177/154411130201300206
- APAMBIRE, W. B.; BOYLE, D.R.; MICHEL, F. A. Geochemistry, genesis, and health implications of fluoriferous groundwaters in the upper Regions of Ghana. **Environmental Geology**, v.33, n.1, p.13-24, 1997. DOI: 10.1007/s002540050221
- APHA - American Public Health Association. 1995. **Standard Methods for Examination of Water and Wastewater**. 19 ed. Washington: American Water Works Association.
- ATLAS BRASIL. **Atlas do Desenvolvimento Humano no Brasil 2013**. Disponível em: <www.atlasbrasil.org.br/2013>. Acesso em: 5/3/2015.
- BARATHI, M.; KUMAR, A. S. K.; RAJESH, N. Impact of fluoride in potable water—An outlook on the existing defluoridation strategies and the road ahead. **Coordination Chemistry Reviews**, v. 387, p. 121-128, 2019. DOI:10.1016/j.ccr.2019.02.006
- BRASIL. Ministério da Saúde. Conselho Nacional de Saúde (CNS). Resolução n. 196, de 10 de outubro de 1996. Aprova as diretrizes e normas regulamentadoras de pesquisas envolvendo seres humanos. Brasília, **Diário Oficial da União**, 16 out. 1996.
- BRASIL. Ministério da Saúde. Secretaria de Atenção à Saúde. Departamento de Atenção Básica. **Projeto SB Brasil 2010 – Condições de saúde bucal da população brasileira 2010: resultados principais**. Brasília: Ministério da Saúde (MS) - Coordenação Nacional de Saúde Bucal (CNSB), 2012.
- CANGUSSU, M. C. T.; NARVAI, P. C.; FERNANDEZ, R. C.; DJEHIZIAN, V. A fluorose no Brasil: uma revisão crítica. **Cad. Saúde Pública**, v.1: p.7-15, 2002. DOI: 10.1590/S0102-311X2002000100002.
- CASTILHO, L. S.; FERREIRA, E.; VELÁSQUEZ, L. N. M.; FANTINEL, L. M. PERINI, E. Fluorose endêmica na América Latina., **Ambiente: Gestão e Desenvolvimento**, v.1, p.15-31, 2015.
- CHOWDHURY, A.; ADAK, M. K.; MUKHERJEE, A.; DHAK, P.; KHATUN, J.; DHAK, D. A critical review on geochemical and geological aspects of fluoride belts, fluorosis and natural materials and other sources for alternatives to fluoride exposure. **Journal of Hydrology**, v.574, p.333-359, 2019. DOI: 10.1016/j.jhydrol.2019.04.033
- CHURCHILL, H. V. Occurrence of fluorides in some waters of the United States. **Journal Industrial & Engineering Chemistry**, v.23, n.9, p. 996-998, 1931. DOI: 0.1021/ie50261a007
- COLOMBO, F.; LIRA, R.; DORAIS, M.J. Mineralogy and crystal chemistry of micas from the A-type El Portezuelo Granite and related pegmatites, Catamarca (NW Argentina). **J. Geosci.**, v.55, n.1, p.43-56, 2010. DOI: 10.3190/jgeosci.058.
- CONCEIÇÃO FILHO, V. M.; MONTEIRO, D. M.; RANGEL, P. D. A.; GARRIDO, I. D. A. Bacia do São Francisco entre Santa Maria da Vitória e Iuiú, Bahia: geologia e potencialidade econômica. **Salvador, CBPM**, 2003. 76p.

- COSTA, D.A. **Controle lito-estrutural e estratigráfico na hidrogeoquímica e nas concentrações de fluoreto no Sistema Aquífero Cárstico - Fissural do Grupo Bambuí, norte de Minas Gerais**. 2011. 131 f. Dissertação (Mestrado), Universidade Federal de Minas Gerais, Belo Horizonte, 2011.
- COSTA, S. M.; ABREU, M. H. N. G.; VARGAS, A. M. D.; VASCONCELOS, M.; FERREIRA, E.; CASTILHO, L. S. Cárie dentária e fluorose endêmica em distritos rurais de Minas Gerais, Brasil. **Revista Brasileira de Epidemiologia**, v.16, p.1021- 1028, 2013. DOI: 10.1590/S1415-790X201300040002
- CARVALHO, J. V. **Hidrogeoquímica e Isótopos estáveis das águas subterrâneas do Aquífero Bambuí (Bahia, Brasil)**. 2018. Dissertação (Mestrado), Universidade Federal da Bahia, Salvador, 2018. 53 f. Disponível em: < <https://repositorio.ufba.br/handle/ri/28608>>. Acesso em: 15.04.2022.
- COUTINHO, C.A.M. **A fluorose dentária na região cárstica do município de Santana-BA: definição de áreas de risco para consumo humano das águas subterrâneas com base nos dados hidroquímicos e epidemiológicos**. 2014. 106 f.Dissertação (Mestrado), Universidade Federal da Bahia, Salvador, 2014. Disponível em: < <https://repositorio.ufba.br/handle/ri/21533>>. Acesso em: 15.04.2022.
- CRUZ, M. J. M.; COUTINHO, C. A. M.; GONÇALVES, M. V. P. The Dental fluorosis on Santana karst region, Bahia State, Brazil. **Journal of Geography**, v.3, n.2, p.51-67, 2015. DOI: 10.15640/jges.v3n2a3
- DEAN, H.T.; JAY, P.; ARNOLD Jr, F. A. ELVOVE, E. Domestic water and dental caries. II. A study of 2,832 white children, aged 12 to 14 years, of 8 suburban Chicago communities, including Lactobacillus acidophilus studies of 1,761 children. **Public Health Rep.**, v.56, n.15, p. 761-9, 1941. DOI: 10.2307/4583693
- DEHBANDI, R.; MOORE, F.; KESHAVARZI, B. Geochemical sources, hydrogeochemical behavior, and health risk assessment of fluoride in an endemic fluorosis area, central Iran. **Chemosphere**, v. 193, p. 763-776, 2018. DOI: 10.1016/j.chemosphere.2017.11.021
- ENALOU, H. B.; MOORE, F.; KESHAVARZI, B.; ZAREI, M. Source apportionment and health risk assessment of fluoride in water resources, south of Fars province, Iran: Stable isotopes ( $\delta^{18}O$  and  $\delta D$ ) and geochemical modeling approaches. **Applied Geochemistry**, v.98, p.197-205, 2018. DOI: 10.1016/j.apgeochem.2018.09.019
- FAIRCHILD, I. J.; BORSATO, A.; TOOTH, A. F.; FRISIA, S.; HAWKESWORTH, C. J.; HUANG, Y.; MCDERMOTT, F.; SPIRO, B. Controls on trace element (Sr-Mg) compositions of carbonate cave waters: implications for speleothem climatic records. **Chemical geology**, v.166, n.3-4, p.255-269, 2000. DOI: 10.1016/S0009-2541(99)00216-8
- FERREIRA, E.F.; VARGAS, A.M.D.; CASTILHO, L.S.; VELÁSQUEZ, L.N.M.; FANTINEL, L.M.; ABREU, M.H.N.G. Factors Associated to Endemic Dental Fluorosis in Brazilian Rural Communities. **Int J Environ Res Public Health**; v.7, n.8, p. 3115-3128, 2010. DOI: 10.3390/ijerph7083115
- FRAZÃO, P.; Ely, H. C.; NORO, L. R. A.; PINHEIRO, H. H. C.; CURY, J. A. The surveillance framework of water and the reporting of fluoride concentration indicators. **Saúde em Debate**, v.42, n.116, p. 274-286, 2018. DOI: 10.1590/0103-1104201811622
- FEJERSKOV, O.; MANJI, F.; BÆLUM, V.; MØLLER, I. J. **Fluorose dentária: um manual para profissionais da saúde**. São Paulo: Editora Santos, 1994. 122p.
- GALAGAN, D.J.; VERMILION, J.R. Determining optimum fluoride concentrations. **Public health reports**, v.72, n.6, p.491-493, 1957. DOI: 10.2307/4589807
- GOMES, A. S. R. **Modelagem metalogenética das mineralizações de Pb-Zn Hospedadas em carbonatos Neoproterozóicos de Irecê (BA), Serra do Ramalho (BA) e Montalvânia (MG)**. 2005. 200 f. Tese (Doutorado), Universidade Federal da Bahia, Salvador, 2005. Disponível em: < <https://repositorio.ufba.br/handle/ri/22036>>. Acesso em: 15.04.2022.
- GONÇALVES, M.V.P. **Flúor no Aquífero Bambuí no Sudoeste da Bahia (Brasil)**. 2014. 193 f. Tese (Doutorado), Universidade Federal da Bahia, Salvador, 2014. Disponível em: < <https://repositorio.ufba.br/handle/ri/21531>>. Acesso em: 15.04.2022.
- GONÇALVES, M.V.P.; CRUZ, M.J.M.; SANTOS, R. A.; RAMOS JUNIOR, A.B. S.; COUTINHO, C.A.M. Flúor na água do Aquífero Bambuí no Oeste da Bahia (Brasil). **Brazilian Journal of aquatic Science and technology**, v.22, n.1, p.10-21, 2018. DOI:10.14210/bjast.v22n1.9654

- GONÇALVES, M. V. P.; SANTOS, R. A.; JUNIOR, A. B. D. S. R.; COUTINHO, C. A. M.; CRUZ, M. J. M.; DA CUNHA, M. P. Flúor na Água Subterrânea de Feira da Mata e Santana (BA): comparação dos teores do fluoreto e saúde. **Geochimica Brasiliensis**, v.33, n.2, p. 175-175, 2019.
- GONÇALVES, M. V. P.; SANTOS, R. A.; COUTINHO, C. A. M.; CRUZ, M. J. M. Fluoride Levels in the Groundwater and Prevalence of Dental Fluorosis in the Municipality of Santana, in Region Karstic of West Bahia, Brazil. In: **Groundwater Hydrology**. London, UK: IntechOpen, 2020. DOI: 10.5772/intechopen.85007
- GOPALAKRISHNAN, S.B.; VISWANATHAN, G. Assessment of fluoride-induced changes on physicochemical and structural properties of bone and the impact of calcium on its control in rabbits. **J. Bone Miner Metab.**, v.30, n.2, p.154-163, 2012. DOI: 10.1007/s00774-011-0312-6.
- HAN, J.; KISS, L.; MEI, H.; REMETE, A. M.; PONIKVAR-SVET, M.; SEDGWICK, D. M.; ANDREW TOMKIN; SOLOSHONOK, V. A. Chemical aspects of human and environmental overload with fluorine. **Chemical Reviews**, v.121, n.8, p.4678-4742, 2021. DOI: 10.1021/acs.chemrev.0c01263
- HANDA, B. K. Geochemistry and genesis of Fluoride-Containing ground waters in india. **Groundwater**, v.13, n.3, p. 275-281, 1975.
- IBGE – Instituto Brasileiro de Geografia e Estatística. 2010. **Dados do Censo demográfico 2010**. Diário Oficial da União do dia 24/11/2010.
- INMET - Instituto Nacional de Meteorologia. Balanço hídrico e dados climatológicos. **Mapa Climatológico de Precipitação Pluviométrica Acumulada Anual (1931-1990), 2011 e 2012**. Disponível em: <www.inmet.gov.br>. Acesso em: 10 mar. 2016.
- KOMATI, S. H.; FIGUEIREDO, B. R. Flúor em água e prevalência de fluorose em Amparo (SP). **Geociências (São Paulo)**, v.32, n.3, p. 547-559, 2013.
- KRAUSKOPF, K.; BIRD, D. **Introduction to geochemistry**. 3ed. New York: Mc Graw-Hill, 1995. 227 p.
- LANDIM, P. M. B. **Análise estatística de dados geológicos multivariados**. 1 ed. São Paulo: Editora Oficina de Textos; 2011. 208 p.
- LI, P.; HE, X.; LI, Y.; XIANG, G. Occurrence and health implication of fluoride in groundwater of loess aquifer in the Chinese loess plateau: a case study of Tongchuan, Northwest China. **Exposure and Health**, v.11, n.2, p.95-107, 2019. DOI: 10.1007/s12403-018-0278-x
- LIMA, I. F. P.; NÓBREGA, D. F.; CERICATO, G. O.; ZIEGELMANN, P. K.; PARANHOS, L. R. Prevalência de fluorose dental em regiões abastecidas com água sem suplementação de flúor no território brasileiro: uma revisão sistemática e metanálise. **Ciência & Saúde Coletiva**, v. 24, p. 2909-2922, 2019. DOI: 10.1590/1413-81232018248.19172017
- LOGAN, J. **Interpretação de Análises Químicas da Água**. U.S. Agency for International Development, Recife, 1995. 75p.
- MARIMON, M. P. C. **O Flúor nas Águas Subterrâneas da Formação Santa Maria, na Região de Santa Cruz e Venâncio Aires, RS, Brasil**. 2006. 228 f. Tese (Doutorado), Universidade Federal do Rio Grande do Sul, Porto Alegre, 2006. Disponível em: <https://lume.ufrgs.br/handle/10183/7289>. Acesso em: 15.04.2022.
- MARTHALER, T. M. Changes in dental caries 1953–2003. **Caries research**, v. 38, n. 3, p. 173-181, 2004. DOI: 10.1159/000077752
- MARTINS, V. S. **Estudos de inclusões fluidas e de isótopos de estrôncio dos depósitos de fluorita da Serra do Ramalho (Bahia) e Montalvânia (Minas Gerais)**. 2001. 89 f. Dissertação (Mestrado), Universidade Federal da Bahia, Salvador, 2001. Disponível em: <https://rigeo.cprm.gov.br/xmlui/handle/doc/395>. Acesso em: 15.04.2022.
- MCDONAGH, M. S.; WHITING, P. F.; WILSON, P. M.; SUTTON, A. J.; CHESTNUTT, I.; COOPER, J.; MISSO, K.; BRADLEY, M.; TREASURE, E.; KLEIJNEN, J. Systematic review of water fluoridation. **Bmj**, v. 321, n. 7265, p. 855-859, 2000. DOI: 10.1136/bmj.321.7265.855
- MERKEL, B. J., PLANER-FRIEDRICH, B. **Geoquímica de águas subterrâneas: um guia prática de modelagem de sistemas aquíferos naturais e contaminados**. Campinas (SP): Editora da Unicamp; 2012. 244 p.

- MENDES, B.; OLIVEIRA, J.F.S. **Qualidade da água para o consumo humano**. Lidel, Edições Técnicas, Lisboa, 2004. 640 p.
- MESSAITFA, A. Fluoride contents in groundwaters and the main consumed foods (dates and tea) in Southern Algeria region. **Environ. Geol.**, v.55, n. 2, p.377-383, 2008. DOI: 10.1007/s00254-007-0983-4.
- MIKKONEN, H. G.; VAN DE GRAAFF, R.; MIKKONEN, A. T.; CLARKE, B. O.; DASIKA, R.; WALLIS, C. J.; REICHMAN, S. M. Environmental and anthropogenic influences on ambient background concentrations of fluoride in soil. **Environmental Pollution**, v. 242, p. 1838-1849, 2018. DOI: 10.1016/j.envpol.2018.07.083
- MIRANDA, L. D.; MONTEIRO, M. D.; CAVALCANTI, J. C. C.; VALLE, C. R. O.; SILVA, J. C. Projeto Fluorita da Serra do Ramalho. **Salvador-Ba, SME: CBPM, Convênio SME/CBPM**; 92 p., 1976.
- MIRANDA, L. L. F.; MONTEIRO, M. D.; CAVALCANTI, J. C. C.; VALLE, C. R. O.; SILVA, J. C. Projeto Fluorita da Serra do Ramalho. **Salvador-Ba, SME: CBPM, Convênio SME/CBPM**, 1976. 92p
- MISI, A.; IYER, S.S.; COELHO, C.E.S.; TASSINARI, C.C.; FRANCA- ROCHA, W.J.; GOMES, A.S.R.; CUNHA, I.A.; TOULKERIDIS, T.; SANCHES, A. L.A. Metalogenic evolution model for the lead-zinc deposits of the Meso and Neoproterozoic sedimentary basins of the São Francisco Cráton, Bahia and Minas Gerais, Brazil. **Revista Brasileira de Geociências**, v.30, n.2, p.302-305, 2000.
- MISI, A.; KAUFMAN, A. J.; VEIZER, J.; POWIS, K.; AZMY, K.; BOGGIANI, P. C.; CLAUDIO, G.; TEIXEIRA, J. B. G.; SANCHES, A. L.; IYER, S. S. Chemostratigraphic Correlation of Neoproterozoic successions in South America. **Chemical Geology**, v.237, n.1-2, p.161-185, 2007. DOI: 10.1016/j.chemgeo.2006.06.019
- MISI, A.; KAUFMAN, A. J.; AZMY, K.; DARDENNE, M. A.; SIAL, A. N.; DE OLIVEIRA, T. F. Neoproterozoic successions of the Sao Francisco Craton, Brazil: the Bambui, Una, Vazante and Vaza Barris/Miaba groups and their glaciogenic deposits. **Geological Society. Memoirs (London)**, v.36, p. 509-522, 2011. DOI:10.1144/M36.48
- NARVAI, P. C. Fluorose dentária iatrogênica endêmica. **Revista Brasileira de Epidemiologia**, v. 5, p. 387-387, 2002. DOI: 10.1590/S0102-311X2002000100002
- NASEEM, S.; RAFIQUE, T.; BASHIR, E.; BHANGER, M. I.; LAGHARI, A.; USMANI, T. H. Lithological influences on occurrence of high-fluoride groundwater in Nagar Parkar area, Thar Desert, Pakistan. **Chemosphere**, v.78, n.11, p.1313-1321, 2010. DOI: 10.1016/j.chemosphere.2010.01.010
- NIRGUDE, A. S.; SAIPRASAD, G. S.; NAIK, P. R.; MOHANTY, S. An epidemiological study on fluorosis in an urban slum area of Nalgonda, Andhra Pradesh, India. **Indian journal of public health**, v.54, n.4, p.194-196, 2010. DOI: 10.4103/0019-557X.77259
- RAJU, N. J. Prevalence of fluorosis in the fluoride enriched groundwater in semi-arid parts of eastern India: Geochemistry and health implications. **Quaternary International**,v.443, p.265-278, 2017. DOI: 10.1016/j.quaint.2016.05.028
- RAO, N. S. High-fluoride groundwater. **Environmental monitoring and assessment**, v.176, n.1, p. 637-645, 2011.
- ROISENBERG, C.; VIERO, A. P.; ROISENBERG, A.; SCHWARZBACH, M. S.; MORANTE, I. C. Caracterização geoquímica e gênese dos principais íons das águas Subterrâneas de Porto Alegre, RS. **Revista Brasileira de Recursos Hídricos**, v.8, n.4, p.137-147, 2003.
- SAEED, M.; MALIK, R. N.; KAMAL, A. Fluorosis and cognitive development among children (6–14 years of age) in the endemic areas of the world: A review and critical analysis. **Environmental Science and Pollution Research**, v. 27, n. 3, p. 2566-2579, 2020. DOI: 10.1007/s11356-019-06938-6
- SANTOS, R. A. **Hidrogeoquímica dos Domínios Cársticos de Irecê, Bahia-Brasil**. 2017. 82 f. Tese (Doutorado), Universidade Federal da Bahia, Salvador, 2017. Disponível em: <<https://repositorio.ufba.br/handle/ri/25830l>>. Acesso em: 15.04.2022.
- SINGH, S.; SAHA, S.; SINGH, S.; SHUKLA, N.; REDDY, V. K. Oral health-related quality of life among 12–15-year children suffering from dental fluorosis residing at endemic fluoride belt of Uttar Pradesh, India. **Journal of Indian Association of Public Health Dentistry**, v.16, n.1, p.54-57, 2018. DOI: 10.4103/jiaphd.jiaphd\_139\_16

SILVA, C. R.; VIGLIO, E.; QUINTARELLI, J. M. Geochemical megaprovince of fluorine and endemic fluorosis in the middle São Francisco river, Minas Gerais-Bahia, Brazil. **Journal of the Geological Survey of Brazil**, v. 3, n. 3, p. 211-224, 2020. DOI: 10.29396/jgsb.2020.v3.n3.5

SOUZA, C. F. M.; LIMA, J. F.; ADRIANO, M. S. P. F.; DE CARVALHO, F. G.; FORTE, F. D. S.; DE FARIAS OLIVEIRA, R., SILVA, A. P.; SAMPAIO, F. C. Assessment of groundwater quality in a region of endemic fluorosis in the northeast of Brazil. **Environmental monitoring and assessment**, v.85, n.6, p.4735-4743, 2013. DOI 10.1007/s10661-012-2900-x

ŠTEPEC, D.; PONIKVAR-SVET, M. Fluoride in human health and nutrition. **Acta Chimica Slovenica**, v. 66, n. 2, p. 255-275, 2019. DOI: 10.17344/acsi.2019.4932

TERRA, L. G.; BORBA, W. F.; FERNANDO, G. D.; TROMBETA, H. W.; SILVA, J. L. S. Caracterização hidroquímica e vulnerabilidade natural à contaminação das águas subterrâneas no município de Ametista do Sul-RS. **Revista Monografias Ambientais**, v.15, n.1, p.94-104, 2016. DOI: <https://doi.org/10.5902/2236130820033>

VELÁSQUEZ, L. N. M.; FANTINEL, L. M.; FERREIRA, E. F.; CASTILHO, L. S.; UHLEIN, A.; VARGAS, A. M. D.; ARANHA, P. R. A. Fluorose dentária e anomalias de flúor na água subterrânea no município de São Francisco, Minas Gerais. *In*: SILVA, C. R.; FIGUEIREDO, B. R.; DE CAPITANI, E. M.; CUNHA, F. G. (Org). **Geologia Médica no Brasil**: efeitos dos materiais e fatores geológicos na saúde humana e meio ambiente. Rio de Janeiro: CPRM - Serviço Geológico do Brasil, p.110-117, 2006.

ZUO, H.; CHEN, L.; KONG, M.; QIU, L.; LÜ, P.; WU, P.; YANG, Y.; CHEN, K. Toxic effects of fluoride on organisms. **Life Sciences**, v.198, p.18-24, 2018.

W.H.O. - World Health Organization. **Calibration of examiners for oral health epidemiological surveys**. Geneva: ORH/EIS/EPID; 1993.

W.H.O. - World Health Organization. **Oral health surveys: basic methods**. 4 ed. Geneva: World Health Organization: ORH/EPID; 1997.

W.H.O. - World Health Organization. **Guidelines for drinking-water quality**. Geneva, World Health Organization. Geneva: World Health Organization, 2006.