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Quality Assessment of fluoridation of public water supply in 88 municipalities in the Northeast region of the state of São Paulo (Brazil)

Avaliação da qualidade da fluoretação de águas de abastecimento público em 88 municípios da região Nordeste do estado de São Paulo (Brasil)

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ABSTRACT

Introduction: During 1 year, the Núcleo de Ciências Químicas e Bromatológicas do Centro de Laboratório Regional do Instituto Adolfo Lutz de Ribeirão Preto (SP, Brasil) has increased the number of public water supply samples analyzed for fluoride concentration in relation to what is usually requested by 88 Municipal Sanitary Surveillance of Northeast State of São Paulo (Brazil). **Objective:** To verify if fluoridation indicators values will vary when increasing the number of samples analyzed and to assess the quality of fluoridation in the region. **Method:** Fluoride was determined by ion chromatography in samples collected by Sanitary Surveillances. **Results:** Indicators values of fluoridation obtained in the two sampling (Sanitary Surveillances and expanded) were similar for most cases. The mapping indicated a greater number of municipalities with values greater than 80% in Regional Departments of Health of Barretos and Franca, while in Araraquara and Ribeirão Preto prevail indicator values lower than 40%. **Conclusions:** Investment and technical assistance are insufficient to increase the values above 80% in municipalities with small populations. For the cases in which the values are smaller than 40% an approach based on the similarity between municipalities is suggested to increase these values.

KEYWORDS: Fluoride; Public Water Supply; Quality of Fluoridation; Sanitary Surveillance

RESUMO

Introdução: Durante um ano, o Núcleo de Ciências Químicas e Bromatológicas do Centro de Laboratório Regional do Instituto Adolfo Lutz de Ribeirão Preto (SP, Brasil) aumentou o número de amostras de águas de abastecimento público analisadas para concentração de fluoreto em relação ao que é normalmente solicitado por 88 Vigilâncias Sanitárias Municipais do Nordeste do estado de São Paulo (Brasil). **Objetivo:** Verificar se os valores dos Indicadores de Fluoretação variam quando se aumenta o número de amostras analisadas e avaliar a qualidade da fluoretação na região. **Método:** Fluoreto foi determinado por cromatografia de íons em amostras coletadas pelas Vigilâncias Sanitárias. **Resultados:** Os valores dos Indicadores de Fluoretação obtidos nas duas amostragens (das Vigilâncias e as ampliadas) foram similares para a maioria dos casos. O mapeamento indicou um maior número de municípios com indicadores maiores que 80% nos Departamentos Regionais de Saúde de Barretos e Franca, enquanto nos de Araraquara e Ribeirão Preto prevalecem indicadores menores que 40%. **Conclusões:** Investimento e assessoria técnica são insuficientes para elevar os indicadores a valores acima de 80% em municípios com populações pequenas. Para os casos em que os valores são menores que 40% sugere-se uma abordagem baseada na similaridade entre municípios para se aumentar esses valores.

PALAVRAS-CHAVE: Fluoreto; Água de Abastecimento Público; Qualidade da Fluoretação; Vigilância Sanitária

INTRODUCTION

In chemical terms, fluorine (F_2) is a highly reactive halogen that is not found in the environment. Instead, what we find is its ion, whether combined with other elements forming minerals, or in the simple ionic form called fluoride (F^-) which is predominant in aqueous media: seawater contains approximately 1 mg L^{-1} of fluoride, while the water of rivers and lakes has concentrations lower than 0.5 mg L^{-1}. Low or high concentrations may occur in groundwater depending on the nature of the soil and the presence of fluoride-containing minerals in its composition[1].

In terms of Public Health, excessive intake of fluoride may lead to dental fluorosis (characterized by the occurrence of yellowish or orange striations or spots on the enamel) and, in more severe cases, bone fluorosis (osteofluorosis), which presents various degrees of severity and occurs in several places, especially in India, China and Africa. Prolonged exposure to high levels of fluoride may lead individuals to conditions of disabling bone deformation. Bone fluorosis is mainly attributed to the consumption of water containing high levels of fluoride, but exposure to other sources such as air (in China, certain provinces burn fluoride-rich coal for cooking and curing food). Some foods (the brick tea in Asia, for example) are also important. In the case of water with high levels of fluoride, there are several defluoridation processes, such as those involving the use of activated alumina or bone charcoal, but the first option to mitigate the fluoride contamination of an affected community is to provide an alternative source of supply[1].

On the other hand, the beneficial action of fluoride in controlling the progression of dental caries[2] is recognized by the World Health Organization, mainly through its topical use in toothpaste and its systemic use in water intended for human consumption (fluoridation) – in which case the concentration of the ion in the water should be maintained within a certain range, generally between 0.5 and 1.0 mg L^{-1}. Caries is a condition caused by enzymes released by certain bacteria that ferment sugar residues present in the mouth causing acids that demineralize the tooth enamel, forming cavities - fluoride increases the resistance of the tooth enamel, additionally leading to the remineralization of the areas that suffered the action of acids. In Brazil, caries is a serious problem in oral health, affecting a large share of the population. To face this problem, the National Health Foundation (Funasa) admits that "access to treated and fluoridated water is fundamental for the health conditions of the population. Enabling public policies that ensure the implementation of water fluoridation is the most comprehensive and socially fair way of having access to fluoride."[3]

For the beneficial action of fluoride with regard to oral health, there is an optimal amount of ingestion: above this amount there is the risk of fluorosis, and below there would be loss of efficiency in the reduction of dental caries. In this sense, the Ministry of Health through Decree n. 635/Bsb of December 26, 1975[4], which sets out norms and standards on water fluoridation, recommends thresholds for fluoride concentration as a function of average daily maximum air temperatures ($\bar{T}_{máx}$) - e.g. in locations where $\bar{T}_{máx}$ are between 14.7 and 17.7° C, it is recommended that the minimum and maximum concentrations should be 0.8 and 1.3 mgL^{-1}, respectively. The fluoride concentration of 1.0 mgL^{-1} is considered optimal. In particular, in the state of São Paulo, Brazil, the current legislation[5] establishes that the concentration of the fluoride ion should be in the range of 0.6 to 0.8 mg L^{-1}, and 0.7 mg L^{-1} is considered the ideal content.

The previous paragraph implies that a given sample of water supply can be classified into two categories, based on the classification of fluoride concentration and human consumption: (i) adequate (concentration within the range of the minimum and maximum concentrations and (ii) inadequate (concentration outside the range between minimum and maximum concentrations). To this dichotomous classification, in 2011 the Collaborating Center of the Ministry of Health in Oral Health Surveillance (CECOL) proposed a new classification relating the concentration of fluoride to the preventive benefit of dental caries and, simultaneously, the risk of dental fluorosis[6]. This new classification is indicated in Table 1, in which we can notice that $\bar{T}_{máx}$ is also considered.

If non-fluoridation (or its discontinuation) eliminates the benefit of caries prevention, the insufficient addition of fluoride makes the procedure innocuous, and the addition of excessive amounts may cause dental fluorosis, thus constant monitoring of fluoride levels in public water supply is key to promote the quality of oral health without deleterious effects. In this sense, it is up to the municipality to determine the basic characteristics of this monitoring in accordance with the "National Guideline of the Sampling Plan for the Environmental Surveillance Related to Water

Table 1. Water classification of CECOL according to fluoride content and benefit-to-risk commitment[6].

$\bar{T}_{max}$(°C)	Fluoride concentration in water (mg L^{-1})	Benefit (preventing caries)	Risk (causing dental fluorosis)
Below 26.3	0.00 to 0.44	Insignificant	Insignificant
	0.45 to 0.54	Minimum	Low
	0.55 to 0.64	Moderate	Low
	0.65 to 0.94	Maximum	Low
	0.95 to 1.24	Maximum	Moderate
	1.25 to 1.44	Questionable	High
	Higher than 1.45	Harm	Very high
Between 26.3 and 32.5	0.00 to 0.44	Insignificant	Insignificant
	0.45 to 0.54	Minimum	Low
	0.55 to 0.84	Maximum	Low
	0.85 to 1.14	Maximum	Moderate
	1.15 to 1.44	Questionable	High
	Higher than 1.45	Harm	Very high
Above 32.5	0.00 to 0.34	Insignificant	Insignificant
	0.35 to 0.44	Minimum	Low
	0.45 to 0.74	Maximum	Low
	0.75 to 0.84	Maximum	Moderate
	0.85 to 1.44	Questionable	High
	Higher than 1.45	Harm	Very high

Source: CECOL/USP, 2011.

Quality for Human Consumption"[7] which establishes not only the number of samples (n) to be collected for analysis, but also when, how and where to collect them. As regards n, it is established at a national level that the minimum number of samples for the hetero-control[8] of the fluoride concentration in public water supply should be calculated on the basis of the total population of the municipality for the various forms of supply – for example, for municipalities with up to 50,000 inhabitants, the minimum monthly number of samples is five. At the state level, the Water Quality Monitoring Program for Human Consumption of the State of São Paulo (Proagua)[9], under the coordination of the Health Surveillance Center (CVS), includes the determination of the fluoride concentration in the public water supply in the State, although less frequently than established for other physico-chemical (pH, content of free residual chlorine, water temperature, apparent color and turbidity) and microbiological (total coliforms and Escherichia coli) parameters.This lower frequency for fluoride analysis is explained by the fact that it may be sufficient to analyze samples of each treatment system on a monthly basis, regardless of the demographic size of the territory covered by the system[6]. In order to determine the fluoride concentration and to evaluate the other parameters in Proagua, the monthly sample size (n) is previously defined in the Program of Actions of Sanitary Surveillance (Pavisa); this program is elaborated by the Sanitary Surveillance team because it is the holder of the knowledge of the universe of its operation and of its operational capacity - nevertheless, it is recommended that this team articulate with other sectors of the Health Department and collaborators who assist in the technical and political feasibility of surveillance actions[10]. Normally, the number of samples analyzed for fluoride through Proagua is lower than that established by the national guidelines.

To evaluate the fluoridation quality of public water supply, Proagua uses the Fluoridation Indicator (IFLU)[11]; this indicator results from the ratio (expressed as a percentage) of the number of samples showing fluoride concentrations in accordance with the drinking standard established by Resolution SS-250 of 15 August 1995[5], (i.e. concentrations between 0.6 and 0.8 mg L^{-1}) and the total number of samples analyzed for a given period, as indicated in the equation below:

$$IFLU = \frac{n_d}{n_t} * 100$$

where: n_d = number of samples according to Resolution SS-250 per period and

n_t = total number of samples analyzed for fluoride in the same period.

The monitoring carried out by Proagua has promoted initiatives to improve the IFLU, especially in small municipalities where challenges in the process of fluoridation of public water supply were detected. Thus, the "Promotion and Quality of Life - Fluoridation of Public Water Supply" project, promoted by the State Health Department (SES), began in 2003. In its first phase, it covered 117 municipalities in São Paulo where the responsibility for the supply was municipal and with a population below 30,000 inhabitants, for which IFLU values were only 9.2%. Resources were made available to the municipalities for the acquisition of equipment and inputs for fluoridation, including equipment for the determination of fluoride ion. At the end of this stage, in 2007 the IFLU reached 42%. Despite the significant increase, this figure was still well below the initial target (IFLU = 80%). In view of this result, a second phase began in 2009 (with an expected end in 2014) involving 107 small municipalities. This strategy involved not only new investments in equipment, but also a series of actions among which the partnership with the Basic Sanitation Company of the State of São Paulo (Sabesp). The partnership included training and education of supply system operators in 2011 provided by Sabesp and CVS technicians[11]. According to information obtained on Jan/18/2017 on the CVS website (www.cvs.saude.sp.gov.br), "the project is in the result evaluation phase".

The support to the municipalities through the training mentioned in the previous paragraph brings to the discussion the technical aspect of fluoridation as at least one of the obstacles to producing adequately fluoridated water. In 1977, Sabesp, at the beginning of its current successful fluoridation program in the municipalities where it is a concessionaire of the supply services, innovated when it replaced the use of sodium fluorosilicate salt (Na_2SiF_6) by the aqueous solution of fluorosilicic acid (H_2SiF_6), lowering costs and simplifying the operational processes[12]. However, we can find a number of alternatives involving both the H_2SiF_6 and the Na_2SiF_6 in the "Manual for the Fluoridation of Water for Human Consumption", published in 2012 by Funasa[3]. In the case of H_2SiF_6, two types of dispensers may be used: dosing pumps and constant level dispensers (in this case, the dispensers may be simple by gravity, by gravity and transport, by gravity aided by water under pressure for transport and application of the mixture, and by constant level under pressure). In the case of Na_2SiF_6, the most commonly used devices are: saturation cone, saturation cylinder, saturation pipes, multi-chamber saturation meter and dosing pump. Obviously, this range of options in the manual of Funasa intends to make it easier for suppliers to choose the most appropriate option for the reality of their municipality; however, for small municipalities with limited resources for the implementation of fluoridation, it may seem attractive to adopt operationally simpler techniques, but whose efficiency lacks proof, such as those that use the so-called "fluoride tablets" available in the market.

In this context, the present study sought to make a detailed and updated diagnosis of the fluoridation quality of public water supply in 88 municipalities in the Northeast region of the state of São Paulo, Brazil. This region comprises the Regional Health Care Network 13 (RRAS 13), which includes four Regional Health Departments (DRS of Ribeirão Preto, Araraquara, Barretos and Franca) of SES (Figure 1), to which the respective Health Surveillance Groups (GVS) report. Each Municipal Sanitary Surveillance body (VISA-M) is linked to a certain GVS - thus, we have: the GVS of Ribeirão Preto consists of 26 VISA-M; the GVS of Araraquara, 24 VISA-M; the GVS of Barreto, 18 VISA-M and the GVS of Franca, 22 VISA-M. In 24 municipalities in the region, Sabesp is responsible for the public water supply.

As urbanization and increasing industrialization provoke an increase of anthropic pressure in the springs and, consequently, change the characteristics of the water used for human consumption, in 2014 our laboratory began a study that for 12 consecutive months provided for the analysis of samples from RRAS 13 public water supply, a region considered as being in process of industrialization[13]. The objective of this project was to characterize the quality of public water supply by increasing the number of investigated parameters: besides those normally required by Proagua (pH, free residual chlorine content, water temperature, apparent color, turbidity, fluoride, total coliforms and *Escherichia coli*), concentrations of lithium, sodium, ammonium, potassium, calcium, magnesium, chlorite, bromate, chloride, bromide, chlorate, nitrate, phosphate and sulfate ions and conductivity measurements were added. The acquisition of the experimental data began in May 2015 and ended in April 2016. The determination of the 15 ions mentioned (including fluoride) was possible due to the ion chromatography introduced by the project[14]. The chromatography also enabled us to increase the number of samples analyzed for the fluoride parameter in relation to the number of analyses required by the VISA-M for hetero-control of the fluoridation quality (in the case of fluoride, the chromatography replaced the potentiometric method with ion electrode selective, used in this laboratory until April/2015). Thus, in addition to presenting a diagnosis of the fluoridation quality of the public water supply of RRAS 13, the present study also draws a comparison between the IFLU values calculated based on the hetero-control conducted by Proagua and the IFLU values calculated from the samples of this study (which include the samples of the hetero-control). Therefore, this comparison can be understood as the verification of the influence of the increase of n (number of samples) on the IFLU value.

METHOD

Sampling

Sampling was described in the previous paper[14], but it can be summarized as follows: the VISA-M bodies are responsible for collecting the samples, for measurements of temperature, free residual chlorine content and pH, and for sending the samples to the laboratory for the complementary analyses. We tried not to interfere in the sampling plans of the municipalities, based on the "National Guideline for the Sampling Plan of the Environmental Surveillance related to the quality of water for human consumption"[7] and on strategies established by Proagua[9,10]. Thus, this laboratory received from the VISA-M 7,587 samples for analysis in the period (from May/2015 to April/2016). Of this total, the VISA-M requested the analysis of fluoride in 2,346 samples. As it would not be possible to analyze all the 7,587 samples received due to the time required for the chromatographic analysis (approximately 38 minutes per sample), we decided to increase the number of samples to be analyzed for fluoride (and for the other 14 ions) based on some conditions: (i) the minimum number of samples per municipality per collection was two; (ii) we sought to maintain a minimum ratio of 1/10,000 between the number of samples per month (n_m) and the number of inhabitants (n_h) of the municipality ($n_m/n_h \geq 1/10{,}000$ or $n_m/n_h \geq 1.10^{-4}$); (iii) whenever possible, samples were selected from sites in the municipality with the greatest distance between them. In this way, we could analyze 2,001 samples in addition to the 2,346 requested, making a total of 4,347 samples analyzed. It should be pointed out that of the total of 90 municipalities in the region, only the municipalities of Franca and Motuca were not included in the study. Franca does not send water samples to this laboratory once it performs the potability analyzes in its own laboratory, and the VISA-M of Motuca did not send water samples for analysis in the period investigated. The remaining 88 municipalities correspond to a population of approximately 3 million inhabitants.

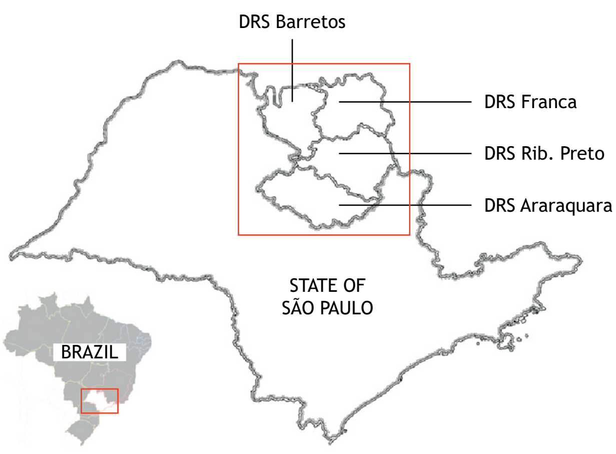

Figure 1. The Regional Health Care Network 13 (RRAS 13) in the state of São Paulo, including the DRS of Araraquara, Barretos, Franca and Ribeirão Preto. DRS = Regional Health Department

Materials and reagents

The reagents used in the analyses had analytical grade (Sigma-Aldrich and Merck) and all aqueous solutions were prepared with type I water obtained from Milli-Q Direct 8 model Millipore purification system.

The 4,347 samples from Proagua analyzed in the period were collected in clean plastic vials (with capacity varying between 250 and 1,000 mL) by agents of the Municipal Sanitary Surveillance body and sent to this laboratory, where they were analyzed on the same date of collection. Prior to the chromatographic analysis, the samples were filtered on Millipore 0.45 µm porous syringe filters, Millex HV model: 33 mm, non-sterile PVDF (polyvinylidene fluoride).

Analysis method and equipment

Fluoride was determined by ion chromatography (Metrohm equipment, model 930 Compact IC Flex Oven/SeS/PP/Deg) using validated method described in prior publication[14].

Data analysis

In the treatment and analysis of the data we used Microsoft Excel® 2013 and Origin® 9.1Pro.

RESULTS AND DISCUSSION

General analysis

Based on the state legislation for the amount of fluoride in public water supply (Resolution SS-250/1995)[5], we observed that of the total of 4,347 samples analyzed, 2,617 samples (60.2%) had fluoride content within the range of 0.6 to 0.8 mg L^{-1} (i.e. within the potability standard), whereas 1,313 samples (30.2%) had a fluoride content of less than 0.6 mg L^{-1} and 417 samples (9.6%) had a fluoride content higher than 0.8 mg L^{-1}. Thus, according to state legislation, 60.2% of the samples were classified as adequate for human consumption and 39.8% were classified as inadequate. The application of the classification of CECOL[6] results in Table 2, in which we can see that 2,760 samples (63.5% of the total) presented the maximum benefit (caries prevention) with a low risk (occurrence of dental fluorosis). These results are lower than those published in 2015, in a study conducted by the State Council of Dentistry of São Paulo (Crosp) for the entire state, in partnership with CECOL and the University of Campinas[15]. Back then, from the 11,715 samples analyzed in 642 municipalities, it was reported that 71.5% of the samples were adequate, according to Resolution SS-250/1995 (71.1% with maximum benefit/low risk, according to CECOL).

Comparison between the IFLU indicators calculated from the Proagua sampling and the sampling performed in this study

In this study, the increase in the frequency of analysis of water samples compared to that normally requested by the VISA-M through Proagua for the determination of fluoride concentration complied with the conditions mentioned before (sub-item "Sampling", under "Method"). For example, if in a given collection a given municipality sent ten samples, of which three required fluoride analysis, the additional samples would be chosen among the remaining seven samples, always following those conditions. With this procedure, we tried not to interfere with the sampling of the VISA-M, except for the value of n. These samplings are guided by the National Guideline[7] not only as to the size of n, but also as to when (how often), where and how to perform the collections, in order to address different characteristics that may exist in the same municipality, different water sources, different treatment and distribution systems, priority areas for surveillance etc. Thus, during the period covered by the experimental part of the project (May 2015 to April 2016), in addition to the 2,346 samples for which VISA-M requested fluoride analysis, it was possible to analyze 2,001 additional samples (resulting in a total of 4,347 samples), which means an increase of 85.3% in the number of samples analyzed for fluoride. In fact, this substantial increase in total samples is the result of very different contributions from each municipality. Table 3 shows that, while the municipalities of Cássia dos Coqueiros, Jeriquara, Santa Cruz da Esperança and Cristais Paulista (lines 3, 5, 23 and 53 of Table 3, respectively) had an increase of only 8.3% in n as indicated in column G ("Increase in n"), for the municipalities of Jaboticabal (line 21), São Joaquim da Barra (line 59), Monte Azul Paulista (line 66) and São Carlos (line 27) increases in n were 188.9%; 195.5%; 204.0%; and 223.5%, respectively. This heterogeneity of n for each municipality is a function not only of the respective number of inhabitants, but also of the number of monthly collections and the particular interest in a given municipality (Monte Azul Paulista water, for example, has challenges due to nitrate[16] contamination – thus, in this case, it was important to increase n with consequent increase in the likelihood of a probable inference to be made from the data, not necessarily related to the quality of the fluoridation).

When changing n, the immediate question to be answered concerns the IFLU: what was the impact on this indicator when n increased? With that in mind, IFLU values were calculated on both samplings: those requested by the VISA-M through Proagua (column H of Table 3) and the sampling of this study (column I - Table 3 was built so that the IFLUs in that column were in descending order). In a first analysis, the Wilcoxon signed-ranks test between the two series of indicators (Proagua sampling vs. study sampling), with significance level $\alpha = 0.05$, showed that there were no statistically significant differences between them ($p = 0.51$), that is, there were no systematic differences between the pairs of data (or, in other words: increasing n did not systematically change IFLU values).

In fact, when we look at the first 28 rows of Table 3, we note that the difference between the calculated values is small (column J), above and below zero, coinciding with the municipalities where fluoridation is apparently well established (high values of IFLU in any of the two samples). In the following lines, low values remain for the differences, with some exceptions involving municipalities with less than 40,000 inhabitants. At least part of these larger differences can be justified by taking examples, such as the municipality of Morro Agudo (line 64), which presented the largest difference among the 88 investigated (-25.5 percentage points): in the period, this municipality sent (through two monthly collections) 108 samples of water to this laboratory, of which in 24 the VISA-M requested fluoride analysis. None of these 24 samples was collected at addresses located in the district named "Center". On the other hand, this study allowed the additional analysis of 30 samples, of which 15 had addresses located in that district – of these 15 samples with addresses located in the "Center", only one presented the adequate concentration of

Table 2. Classification of the supply water of the Regional Health Care Network 13 (RRAS) according to the classification of the Collaborating Center in Oral Health Surveillance of the Ministry of Health (CECOL).

Fluoride content (mg L^{-1})	Benefit	Risk	N. samples	%
Below 0.44	Insignificant	Insignificant	821	18.9
Between 0.45 and 0.54	Minimum	Low	370	8.5
Between 0.55 and 0.84	Maximum	Low	2760	63.5
Between 0.85 and 1.14	Maximum	Moderate	323	7.4
Between 1.15 and 1.44	Questionable	High	40	0.9
Above 1.45	Harm	Very high	33	0.8

Table 3. Calculation of the IFLU indicators from the Proagua sampling and the sampling performed in this study.

	A	B	C	D	E	F	G	H	I	J
	Municipality	Responsible	Water source(a)	Habits(b)	Number of samples (n)		Increase in n (%)	Calculated IFLU (%)		Difference
					Proagua	This study		Proagua	This study	(I - H)
1	Barretos	SAM(c)	SUB + SUP(d)	112101	62	145	133.9	100.0	100.0	0.0
2	Cajuru	Sabesp	SUP	23371	27	48	77.8	100.0	100.0	0.0
3	Cássia dos Coqu	Sabesp	SUP	2634	24	26	8.3	100.0	100.0	0.0
4	Dourado	Sabesp	SUB	8609	20	26	30.0	100.0	100.0	0.0
5	Jeriquara	Sabesp	SUB	3160	24	26	8.3	100.0	100.0	0.0
6	Miguelópolis	Sabesp	SUB	20451	19	43	126.3	100.0	100.0	0.0
7	Rib Corrente	Sabesp	SUB	4273	24	28	16.7	100.0	100.0	0.0
8	Sta Ernestina	Sabesp	SUB	5568	24	30	25.0	100.0	100.0	0.0
9	Sta Rosa Viterbo	Sabesp	SUP	23862	24	39	62.5	100.0	100.0	0.0
10	Terra Roxa	Sabesp	SUB	8505	24	30	25.0	100.0	100.0	0.0
11	Jaborandi	Sabesp	SUB	6592	23	31	34.8	95.7	96.8	1.1
12	Colômbia	Sabesp	SUP	5994	24	28	16.7	95.8	96.4	0.6
13	Guaíra	SAM	SUP	37404	36	52	44.4	97.2	96.2	-1.0
14	Colina	SAM	SUB	17371	27	38	40.7	92.6	94.7	2.1
15	Igarapava	Sabesp	SUB	27952	24	57	137.5	95.8	94.7	-1.1
16	Pedregulho	Sabesp	SUB + SUP	15700	24	37	54.2	95.8	94.6	-1.2
17	Restinga	Sabesp	SUP	6587	15	28	86.7	100.0	92.9	-7.1
18	Rifaina	Sabesp	SUB	3436	16	26	62.5	93.8	92.3	-1.5
19	Guariba	Sabesp	SUB	35486	22	53	140.9	86.4	90.7	4.3
20	Buritizal	Sabesp	SUB	4053	24	42	75.0	91.7	90.5	-1.2
21	Jaboticabal	SAM	SUB + SUP	71662	36	104	188.9	94.4	90.4	-4.0
22	Porto Ferreira	Odebrecht	SUP	51400	42	78	85.7	88.1	89.7	1.6
23	Sta Cruz Esper	Sabesp	SUB	1953	24	26	8.3	91.7	88.5	-3.2
24	Itirapuã	Sabesp	SUB	5914	20	24	20.0	90.0	86.4	-3.6
25	Viradouro	SAM	SUB + SUP	17297	20	28	40.0	90.0	85.7	-4.3
26	Ipuã	SAM	SUB + SUP	14148	24	49	104.2	87.5	83.7	-3.8
27	São Carlos	SAM	SUB + SUP	221950	51	165	223.5	84.3	83.6	-0.7
28	Taquaral	SAM	SUB	2726	25	36	44.0	80.0	83.3	3.3
29	**Pitangueiras**	**SAM**	**SUB + SUP**	**35307**	**24**	**52**	**116.7**	**70.8**	**82.7**	**11.9**
30	Când Rodrigues	Sabesp	SUB	2668	13	26	100.0	84.6	80.8	-3.8
31	Ituverava	SAM	SUP	38695	24	52	116.7	70.8	78.8	8.0
32	Patr Paulista	SAM	SUB + SUP	13000	24	42	75.0	75.0	78.6	3.6
33	Guará	SAM	SUB	19858	24	39	62.5	75.0	76.9	1.9
34	Monte Alto	Sabesp	SUB	46642	24	64	166.7	75.0	76.6	1.6
35	Bebedouro	SAM	SUB + SUP	75035	51	104	103.9	76.5	76.0	-0.5
36	Rib Preto	SAM	SUB	604682	62	144	132.3	75.8	75.7	-0.1
37	Ibitinga	SAM	SUB + SUP	53158	42	79	88.1	78.6	74.7	-3.9
38	Serra Azul	Sabesp	SUB	11256	26	35	34.6	76.9	74.3	-2.6
39	Altair	Sabesp	SUB	3815	21	33	57.1	76.2	72.7	-3.5
40	Batatais	SAM	SUB + SUP	56476	36	80	122.2	63.9	72.5	8.6
41	**Dumont**	**SAM**	**SUB**	**8143**	**24**	**29**	**20.8**	**83.3**	**72.4**	**-10.9**
42	Taquaritinga	SAM	SUB + SUP	53988	41	78	90.2	75.6	71.8	-3.8
43	Sta Rita P Quatr	SAM	SUP	26478	24	43	79.2	70.8	69.8	-1.0
44	Nuporanga	SAM	SUB	6817	20	26	30.0	70.0	69.2	-0.8
45	Araraquara	SAM	SUB + SUP	208662	52	148	184.6	65.4	68.2	2.8
46	Descalvado	SAM	SUB + SUP	31056	24	52	116.7	75.0	67.3	-7.7
47	Sertãozinho	SAM	SUB	110074	72	156	116.7	72.2	67.3	-4.9
48	Vista Alegr Alto	SAM	SUB	6886	23	34	47.8	65.2	64.7	-0.5
49	Ibaté	SAM	SUB	30734	19	52	173.7	63.2	61.5	-1.7
50	Matão	SAM	SUB	76786	40	104	160.0	57.5	59.6	2.1
51	Pontal	SAM	SUB	40244	23	64	178.3	56.5	59.4	2.9
52	Cravinhos	SAM	SUB + SUP	31691	24	52	116.7	62.5	57.7	-4.8
53	Cristais Paulista	SAM	SUB + SUP	7588	24	26	8.3	58.3	57.7	-0.6
54	Olímpia	SAM	SUB + SUP	50024	51	78	52.9	52.9	53.8	0.9
55	Cajobi	SAM	SUB	9768	20	25	25.0	50.0	52.0	2.0
56	**Guaraci**	**SAM**	**SUP**	**9976**	**20**	**27**	**35.0**	**55.0**	**44.4**	**-10.6**
57	Pradópolis	SAM	SUB	17377	24	31	29.2	45.8	41.9	-3.9
58	**Trabiju**	**SAM**	**SUB**	**1544**	**16**	**31**	**93.8**	**31.3**	**41.9**	**10.6**

Continues

Table 3. Calculation of the IFLU indicators from the Proagua sampling and the sampling performed in this study. Continuation

59	S Joaquim Barra	SAM	SUB + SUP	46512	22	65	195.5	40.9	41.5	0.6
60	S José Bela Vista	SAM	SUB + SUP	8406	25	29	16.0	40.0	41.4	1.4
61	Borborema	SAM	SUB	14529	24	39	62.5	33.3	41.0	7.7
62	Sales Oliveira	SAM	SUB	10568	24	28	16.7	41.7	39.3	-2.4
63	Taiuva	SAM	SUB	5447	24	32	33.3	33.3	37.5	4.2
64	**Morro Agudo**	**SAM**	**SUB**	**20116**	**24**	**54**	**125.0**	**62.5**	**37.0**	**-25.5**
65	Dobrada	SAM	SUB	7939	17	28	64.7	35.3	35.7	0.4
66	Mte Azul Pta	SAM	SUB	18931	25	76	204.0	28.0	34.2	6.2
67	Nova Europa	SAM	SUB	9300	18	27	50.0	38.9	33.3	-5.6
68	Barrinha	SAM	SUB	28496	24	41	70.8	29.2	31.7	2.5
69	São Simão	SAM	SUB + SUP	14346	24	32	33.3	37.5	31.3	-6.2
70	Altinópolis	SAM	SUB + SUP	15607	25	31	24.0	28.0	25.8	-2.2
71	Itápolis	SAM	SUB + SUP	40051	26	65	150.0	15.4	23.1	7.7
72	**Severínia**	**SAM**	**SUB**	**15501**	**23**	**60**	**160.9**	**34.8**	**21.7**	**-13.1**
73	Brodowski	SAM	SUB	21107	23	51	121.7	61.7	21.6	-0.1
74	Taiaçu	SAM	SUB	5894	24	33	37.5	25.0	21.2	-3.8
75	Am Brasiliense	SAM	SUB	34478	24	52	116.7	29.2	19.2	-10.0
76	Tabatinga	SAM	SUB	14686	24	29	20.8	16.7	17.2	0.5
77	Aramina	SAM	SUB	5152	24	42	75.0	12.5	16.7	4.2
78	Rib Bonito	SAM	SUB + SUP	12135	24	33	37.5	12.5	15.2	2.7
79	Gavião Peixoto	SAM	SUB	4419	18	29	61.1	11.1	13.8	2.7
80	Luís Antônio	SAM	SUB	11286	22	29	31.8	9.1	13.3	4.2
81	Jardinópolis	SAM	SUB + SUP	37661	24	52	116.7	8.3	11.5	3.2
82	Guatapará	SAM	SUB	6966	23	29	26.1	13.0	10.3	-2.7
83	Sta Lúcia	SAM	SUB	8248	22	26	18.2	9.1	7.7	-1.4
84	Sto Ant Alegria	SAM	SUB	6304	24	27	12.5	8.3	7.4	-0.9
85	Serrana	SAM	SUB	38878	25	55	120	0.0	5.5	5.5
86	Rincão	SAM	SUB	10414	16	26	62.5	0.0	3.8	3.8
87	B Esperança Sul	SAM	SUB	13645	18	26	44.4	0.0	0.0	0.0
88	Orlândia	SAM	SUB + SUP	39781	25	52	108.0	0.0	0.0	0.0

(a) Information obtained from the Urban Water Supply Atlas of the Brazilian National Water Agency17; (b) The number of inhabitants of each municipality was obtained in the 2010 census of the Brazilian Institute of Geography and Statistics (www.ibge.gov.br); (c) SAM = Municipal Supply Service (means that the municipality is responsible for its own public water supply); (d) SUB = use of groundwater for supply; SUP = use of superficial water for supply; SUB + SUP indicates that the municipality uses both sources of water.

fluoride (therefore an IFLU for this district, the "Center", would have the extremely low value of 6.7%). This fact suggests that, in the case of Morro Agudo, the sampling performed by the VISA-M should include samples that adequately represent the entire municipality so that the municipal IFLU reflects the quality of the fluoridation more accurately. From the technical point of view, it seems that the part of the distribution system that supplies the "Center" district needs extensive adaptation, since the median fluoride concentration of the 15 samples located therein is only 0.08 mg L^{-1}.

Another notorious example is Pitangueiras (line 29), which sent (through a monthly collection) 108 water samples, of which in 24 the VISA-M requested fluoride analysis. We could analyze another 28 additional samples. In this case, where the difference was positive (+11.9 percentage points), there were repetitions of collection points in the sampling performed with regard to districts (central urban area and suburbs). For example, on July 13, 2015, all nine samples collected in Pitangueiras came from the "Center" district; in the collection of November 23, 2015, eight samples were collected in the "Center" and only one in a suburb (Jardim Santa Rita); this sampling was repeated on November 6, 2016 (eight samples in the "Center" and one in Jardim Santa Rita). These replicates, mostly in districts of adequate fluoridation, led to a bias in the calculation of the IFLU from the data of this study, causing an increase in its value when compared with the value calculated only with the Proagua samples. It is interesting to note that fluoride levels in the Pitangueiras water supply seemed to become more suitable to the range established by Resolution SS-250/1995[5], in relation to the period prior to this study, notably after July 2013; in fact, the IFLU values calculated for the two immediately preceding periods (May 2013 to April 2014, and May 2014 to April 2015) showed an upward trend (55.6 and 76%, respectively).

At first, the differences between the IFLU indicators calculated for the other municipalities highlighted in Table 3 (Dumont, line 41; Guaraci, line 56; Trabiju, line 58; and Severínia, line 72) could be interpreted similarly to the cases of Morro Agudo and Pitangueiras (Proagua sampling that was not representative for negative differences or bias caused by repetitions in areas of adequate fluoridation in the sampling of this study). If the positive difference observed for Trabiju can be justified by the simplicity of the distribution system of the municipality with fewer inhabitants in the whole state (one well and one reservoir), which caused repetitions in the sampling of this study, the causes of the negative differences of the other three municipalities are less evident, especially in the cases of Dumont and Guaraci, in which only five and seven samples were added to this study, respectively. Nevertheless, it is interesting to note that the variability of fluoride concentrations, represented by the range from the lowest to the highest concentration determined in the period from May/2015 to April/2016, was relatively large for these municipalities (Guaraci, from 0.009 to 1.972 mg L^{-1}; Severínia, 0.05 to 1.782 mg L^{-1}; Dumont, 0.303 to 1.053 mg L^{-1}) and higher than those observed in the Proagua sampling, contrasting, for example, with the range of concentrations determined in Pitangueiras (from 0.443 to 0.873 mg L^{-1}). This fact suggests that an increase in the value of n can better evaluate the variability of fluoride concentrations in municipalities whose fluoridation process needs more substantial improvement. In practical terms, this would mean more accurately diagnosing the difficulties of each municipality in maintaining fluoride levels within the range established by the legislation.

Mapping the quality of fluoridation in the municipalities

In order to visualize the distribution of municipalities with similar fluoridation quality levels, aiming at possible future actions, either under RRAS 13 or within each of the four DRS in the region, the municipalities were classified into four groups, as a function of the calculated IFLU values in this study (column I of Table 3):

- Group 1: the IFLU value of 30 municipalities (26.7% of the total population of the municipalities studied) was higher than 80.0%. In this group, the supply of 20 municipalities is the responsibility of Sabesp. Highlights to the municipalities of Barretos, Santa Rosa de Viterbo, Cajuru, Miguelópolis, Dourado, Terra Roxa, Santa Ernestina, Ribeirão Corrente, Jeriquara and Cássia dos Coqueiros, all with IFLU = 100.0 %. São Carlos (IFLU = 83.6%), the second municipality in population among the 88 studied, also belongs to this group, as well as Pitangueiras (the latter with the exception that sampling produced bias, as discussed in the previous item). Sabesp's expertise is evident, either in the fluoridation of waters from surface water sources (involving water treatment plants – ETAs) or in the fluoridation of groundwater springs (or when both are used, as in the case of Pedregulho). Nonetheless, it should be pointed out that the municipalities responsible for their own supply are also capable of fluoridating water in these three catchment situations (groundwater, surface water or both), including those of more complex distribution systems due to the larger population (like Barretos and São Carlos), and those that require less complex systems (like Taquaral and Ipuã).
- Group 2: in a second group of 12 municipalities (32.9% of the population), the IFLU value was between 70 and 80%. At first, municipalities in this group need minor technical adjustments in fluoridation processes in order to achieve the minimum goal of 80%. In particular, three municipalities in this group have Sabesp responsible for their supply, but their present classification in Group 2 is apparently the result of specific events (not methodological shortcomings in the fluoridation process), since the IFLUs calculated for the two periods immediately before (May 2013 to April 2014, and May 2014 to April 2015) presented values equal to or higher than 90.0%. Ribeirão Preto (municipality with the largest population in the region) increased its IFLU in this study by more than 20% in comparison with previous periods (IFLU = 55.6% from May 2013 to April 2014 and IFLU = 52.4% from May 2014 to April 2015).
- Group 3: IFLU values are between 40.0% and 70.0%. This group consists of 19 municipalities (24.6% of the population), including Araraquara (3rd in population) and Sertãozinho (5th in population); in the case of this group, we suggest that significant changes must be made in the fluoridation process in order to enhance its indicators. Araraquara experienced a growing trend in the IFLU value in the three mentioned periods: from 51.6% (May 2013 to April 2014), the index increased to 60.0% in the following period, finally reaching 68.2% in this study). A reverse trend is observed in Sertãozinho: 78.3%, 68.1% and 67.3%, respectively. If in the first case the actions should be for improvement, in the second the actions should be corrective to reverse the downward trend in the IFLU value.
- Group 4: 27 municipalities with up to 40,000 inhabitants (corresponding to 15.7% of the total population covered by this study) presented IFLU below 40.0%. For this group we suggest that the fluoridation process be fully revised since the indicators are well below the 80% target. Highlights to Boa Esperança do Sul and Orlândia, both with 0% IFLU (these indicators were also obtained in the two immediately previous periods). It is important to note that in 2013 the Regional Chemistry Council of the IV Region (SP) - CRQ-IV summoned about 70 head technicians in ETAs in the state of São Paulo to provide clarification regarding the inadequacy of fluoridation in their respective municipalities. This intervention was motivated by the results of a Crosp study, in partnership with the School of Dentistry of Piracicaba (Unicamp)[18]. It is also worth noting that Rincão, Serrana, Santo Antônio da Alegria and Santa Lúcia presented values of IFLU below 10%. In the case of Santa Lúcia, it should be noted that in 2003 the CRQ-IV had to appeal to the court for the municipality to hire a chemist to work on the treatment of public water supply (drinking water treatment is to be done only by chemists, as established in the third paragraph of article 2 of Decree n. 85877 of April 7, 1981[19]). The lawsuit was judged in 2009 with favorable decision to CRQ-IV[20]. Symptomatically, this municipality still finds difficulties not only to adequately fluoride its water supply, but also to conduct efficient processes of disinfection as described in recently published communication[21].

The map shown in Figure 2 indicates the classification of municipalities in each of the four groups. Comparing Figure 2 with Figure 1 we can observe that, while there is a greater number of municipalities from DRS of Barretos and Franca in Group 1, the DRS of Araraquara and Ribeirão Preto present a larger number of municipalities in Group 4. This fact can not be interpreted only by the greater number of municipalities served by Sabesp in the first two DRS (13 municipalities) when compared to the other two (10 municipalities) – for example, the DRS of Barretos

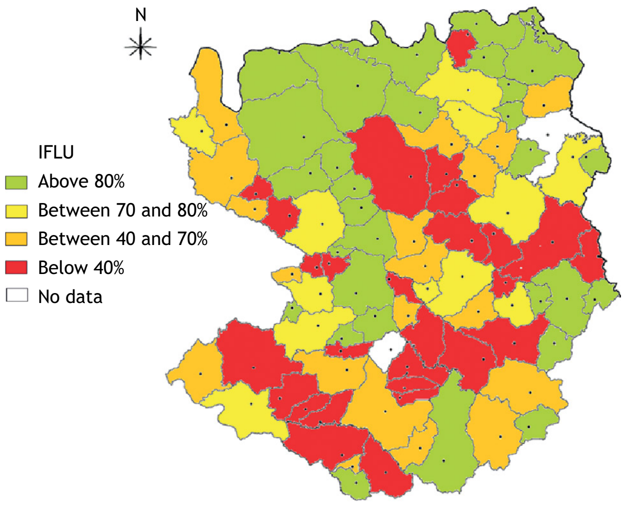

Figure 2. Location of municipalities in the Regional Health Care Network 13 (RRAS 13) with respective classifications regarding the quality of fluoridation

presented eight municipalities classified in Group 1, but only in three Sabesp is responsible for the supply (a fourth municipality of this DRS, served by Sabesp, was classified in Group 2).

It is not the purpose of this study to outline strategies for improving the quality of fluoridation, but a possible approach to the problem in RRAS 13 would be to initially focus attention on the municipalities of Group 4. This approach could involve: (i) intensive community awareness raising (through lectures and handouts, for example) as to the importance of adding fluoride to the water supply - once the population is more aware, they can reason with the municipal managers in order to request their right to access to adequately fluoridated water; (ii) to encourage the exchange of experiences (such as workshops and technical meetings, for example) among those responsible for supplying municipalities with similarities between their realities in drinking water supply. In this second action, one example is the DRS of Barretos, where four municipalities (Monte Azul Paulista, Severínia, Taiaçu and Taiuva) belong to Group 4 and are close to Taquaral and Colina (classified in Group 1); the six municipalities are responsible for their own water supply, they all use groundwater sources, and in terms of population size (which directly influences the complexity of the distribution system), Taiaçu and Taiuva are close to the reality faced by Taquaral, while Monte Azul Paulista and Severínia have similarities with Colina. A workshop (or a series of technical meetings) including these six municipalities could redirect actions to improve the quality of fluoridation in the municipalities classified in Group 4.

CONCLUSIONS

The results obtained in this study suggest that the goal of raising IFLU values to 80% requires not only investments and technical assistance in municipalities with small populations. The situation is complex and must involve other factors, including cultural factors. In most cases, the increase in the number of public water supply samples analyzed for fluoride did not promote significant changes in IFLU values when compared to those normally calculated for Proagua. This implies that this sampling with lower n is satisfactory when the objective is to evaluate the fluoridation process through the IFLU. However, with the increase in n we could detect non-representative sampling, repetitions of collection addresses (causing bias), and higher dispersions in fluoride concentrations in municipalities with a population of less than 40,000 inhabitants (implying serious challenges in the control of the fluoridation process).

The classification of 88 municipalities of RRAS 13 into four groups according to the IFLU levels obtained in this study allowed us to diagnose the presence of serious difficulties in obtaining adequate levels of fluoride in 27 municipalities with up to 40,000 inhabitants, despite the technical and financial support of a project called "Promotion of Quality of Life - Fluoridation of Public Water Supply" started in 2003. A map suggested an alternative approach to the challenge of raising IFLUs to a minimum of 80% in these small municipalities where the municipality itself is in charge of the supply. This approach, based on the combination of experiments with successful fluoridation processes with those that need significant improvements, may allow the exchange of technical information between municipalities that present some similarities, such as the source used and the population to be supplied. The example of the DRS of Barretos involving municipalities of Group 1 (larger IFLU) with municipalities of Group 4 (smaller IFLU) can be understood as a simple transfer of expertise from those that can now obtain better quality fluoridation to those that still need to improve their indicators significantly.

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Conflict of Interest

Authors have no potential conflict of interest to declare, related to this study's political or financial peers and institutions.