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SCIENTIFIC ARTICLE

Fluoride concentration in private artesian wells of different depths

Concentração de fluoreto em poços artesianos particulares de diferentes profundidades

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ABSTRACT: The groundwater presents a concentration of fluoride ranging from values below 0.05 and up to 35 $mgF^{-} L^{-1}$, depending on rocks composition. The concentrations of fluoride above 1.45 $mgF^{-} L^{-1}$ are associated with harmful effects, such as dental fluorosis, when the constant intake of high concentrations occurs during the tooth formation period. Considering that variations in fluoride concentrations in groundwater can be influenced by the depth of wells and high concentrations represent risk, it is necessary to monitor the groundwater. In this context, the objective of this study was to evaluate fluoride concentration in artesian wells water at different depths in Maringá region, PR. Water samples were collected from 166 artesian wells, with depths ranging from 9 to 400 m and information about depths and location of the wells were obtained from Water Institute of Maringá. The samples analysis were carried out by specific ion electrode to fluoride identification. The results showed that the average concentration of fluoride was 0.09 $\pm$ 0.15 $mgF^{-} L^{-1}$, ranging from 0.03 to 0.92 $mgF^{-} L^{-1}$. No correlation was observed between fluoride concentrations with wells depths. It was concluded that the majority samples evaluated in this study presented low concentration of natural fluoride, except for one water sample.

Keywords: Groundwater; Fluoride; Water Wells.

RESUMO: A concentração de fluoreto (F^{-}) em águas subterrâneas pode variar entre valores abaixo de 0,05 até acima de 35 $mgF^{-} L^{-1}$, dependendo da composição das rochas. Concentrações acima de 1,45 $mgF^{-} L^{-1}$ associam-se a efeitos nocivos, tais como a fluorose dental, quando o consumo de altas doses de fluoretos ocorre durante a formação do germe dentário. Considerando que variações nas concentrações de F podem ser influenciadas pela profundidade dos poços e as altas concentrações representam riscos, o constante monitoramento das águas provenientes dos poços artesianos torna-se necessário. Assim, o objetivo deste estudo foi avaliar a concentração de F^{-} em águas de poços artesianos de diferentes profundidades na região de Maringá, PR. As amostras de água foram coletadas de 166 poços artesianos, com profundidades de 9 a 400 m e as informações quanto à profundidade e localização foram obtidas do Instituto de Águas de Maringá. A análise das amostras foi realizada com eletrodo específico para a identificação de fluoreto. Os resultados demonstraram que a concentração média de F^{-} foi de 0,09 $\pm$ 0,15 $mgF^{-} L^{-1}$, variando de 0,03 a 0,92 $mgF^{-} L^{-1}$. Nenhuma correlação foi observada entre as concentrações de F^{-} e profundidade dos poços. Conclui-se que a maioria das amostras apresentou baixa concentração natural de F^{-}, exceto para uma amostra.

Palavras-chave: Água Subterrânea; Fluoretos; Poços de Água.

INTRODUCTION

Access to drinking water was declared a fundamental human right by the United Nations and is one of the global targets included in the 6th Sustainable Development Goals (GDSs) agenda for 2030 to ensure the availability and sustainable management of water supply and sanitation for all (Un, 2017). Population growth and increased water human consumption have resulted in higher demands for natural sources of groundwater (Barros & Amin, 2008). Because human health depends on the quality of drinking water, constant monitoring is required to ensure its safety. The presence of ions of

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naturally occurring elements, such as fluoride, may be either beneficial or toxic for human health, depending on their concentrations (Aoun et al., 2018).

Fluoride has been recognized worldwide for its beneficial effect on dental caries control (Spencer et al., 2018, Cruz & Narvai, 2018). However, dental fluorosis from chronic exposure to fluoride, characterized by enamel hypomineralization, could negatively affect smile aesthetics and the quality of life of people, especially children (Cury et al., 2019). Generally, when high concentrations of fluoride are present in drinking water, or when fluoride in toothpastes, food, and beverages is ingested, fluorosis can be highly prevalent. Some studies conducted in Brazil have observed a wide variability in the prevalence and severity of fluorosis in the population, ranging from 0 (Campos et al., 1998) to 97.6% (Capella et al., 1989), depending on the region studied, according to a critical review in 42 cities in the period from 1970 to 2000 (Cangussu et al., 2002). Although most findings have shown low levels of fluorosis, cases of moderate to severe of the fluorosis degrees have been observed due to the presence of high levels of fluoride naturally occurring in groundwater sources (Lima et al., 2019).

Fluoride concentration levels are an important parameter to evaluate drinking water quality, either due to its beneficial effect in preventing dental caries, or because of its negative effect in causing dental fluorosis. In 2011, the Brazilian Ministry of Health (ordinance number 2914) defined the maximum permitted value of fluoride in drinking water to be 1.5 $mgF^{-} L^{-1}$; the Health Ministry and local Health Secretaries being responsible for its monitoring (Brasil, 2001). According to a new fluoride concentration classification proposed by the Brazilian Ministry of Health Oral Health Surveillance Cooperation Center (CECOL, in Portuguese), fluoride concentrations in the drinking water supply between 0.45 to 0.84 $mgF^{-} L^{-1}$ offer low risk of fluorosis. However, when fluoride concentrations are above 1.45 $mgF^{-} L^{-1}$, the risk of fluorosis is considered very high, in locations where the maximum temperature is between 26.3^{0}C and 32.5^{0}C (Centro Colaborador do Ministério da Saúde em Vigilância da Saúde Bucal, 2011). The Ordinance 635/1975 that established the calculation formula to fluoride concentration in public drinking water, according to air maximum temperature range as in higher temperature regions, people will need to drink more water, then, fluoride concentrations should be lower (Brasil, 1975).

Some previous findings of fluoride concentrations, ranging from 3.03 to 6.95 $mgF^{-} L^{-1}$, found in 19 deep groundwater wells in the region of Salto-Indaiatuba, SP, Brazil, are worrisome (Hypolito et al., 2010). In Porto Alegre, RS, Brazil, high levels of fluoride were also found in 90 wells, which showed a mean concentration of approximately 6 $mgF^{-} L^{-1}$ (Roisenberg et al., 2003). In other countries, such as the United States, high concentrations of fluoride (32 $mgF^{-} L^{-1}$) in groundwater have also been reported (Hem, 1985). In the hot springs in the Rift Valley in Ethiopia, anomalous values of fluoride (26 $mgF^{-} L^{-1}$) have also been observed (Ashley & Burley, 1995). A recent systematic review revealed higher prevalence of dental fluorosis in populations in Brazil supplied with drinking water from groundwater or artesians wells, when compared to Brazilian cities supplied with non-fluoridated water. These findings indicated that the presence of high concentrations of naturally occurring fluoride can represent a risk factor for the development of dental fluorosis (Lima et al., 2019).

There are some reasons why groundwater could present a higher amount of fluoride: rock porosity, fluoridated compound solubility, and the soils to which it is related (Kumar et al., 2014). Considering that variations in fluoride concentrations in groundwater can be influenced by the depth of wells (Panagoulias Theodoros & Silva Filho, 2006) and the type of rocks water passes through, the content of naturally occurring fluoride in wells requires investigation in order to ensure the safety of drinking water supply to the general population. Moreover, as the demand for drinking water grows, there is an increased demand for more private water wells. According to Brunt et al. (2004), there is little information on fluoride naturally occurring in groundwater and its relation with Paraná State geology. A multilayered aquifer system can act as a source of fluoride at various depths, which could result in areas being contaminated.

Therefore, the objective of this study was to investigate the association between natural fluoride concentration levels and the different depths of artesian private wells supplied by the Serra Geral groundwater aquifer in the region of Maringá.

MATERIAL AND METHODS

This is an observational study that used data about depths and locations of the wells obtained from the records of the Water Institute of Maringá, which is part of the Paraná State Environment and Water Resources Secretary.

Water sampling points were established after researching the number of water wells implanted in the region of Maringá from 2005 to 2015, totaling 918 wells. Once the ID number of each well was obtained, registered wells were identified on Paraná government website (http://www.aguasparana.pr.gov.br/), resulting in 343 wells . Subsequently, data from each well (depth and location) were collected from the records. Only wells with information about depths and addresses were selected, resulting in 196 wells, however, during the collection procedure, 30 wells produced no samples due to the following reasons: deactivated wells, addresses not found, abandoned sites, and collection not authorized by the well owner. Thus, 166 wells were included in the analysis, with depths ranging from 9 to 400 m, that were divided into six groups, according to their depth: A, wells ≤ 50 m (n = 14); B, 51-100 m (n = 56); C, 101-150 m (n = 83); D, 151-200 m (n = 7); E, 201-250 m (n = 5); and F, > 250 m (n = 1). Afterwards, the wells were plotted on the map of the region investigated to show their location (Figure 1).

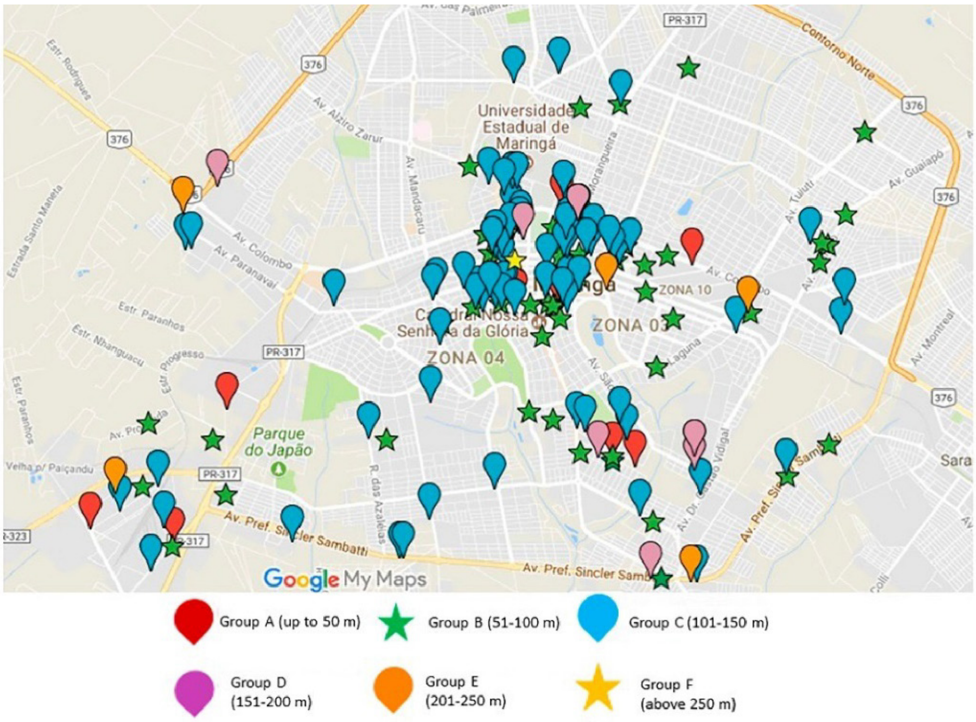

Figure 1 - Distribution of water sampling points in Maringá

Water sampling took place from January to February 2016 in 60 ml polyethylene flasks, which had been previously rinsed 3 times with the same collected water, and identified with the name of the well owner, location and depth. All samples were stored at 5-8 degree Celsius, and fluoride content was determined immediately after all 166 wells samples were collected. The water samples were analyzed at the Biochemistry Laboratory of Piracicaba School of Dentistry, State University of Campinas (FOP-UNICAMP).

Fluoride concentration was determined using an ion analyzer (Orion model Star A214; Orion Research Inc., United States), coupled to an ion specific electrode for fluoride identification (Orion model 9609 BN; Orion Research Inc., USA). The calibration curve was performed in triplicate with six standard solutions, ranging from 0.2 to 2.0 mgF^- L^{-1}, and by adding the same volume of total ionic strength adjustment buffer (TISAB II). Subsequently, the samples were analyzed in duplicate, also with the addition of TISAB II (ratio 1:1) and the data transformed into mgF^- L^{-1}. The mean from the duplicate was calculated for each well. Then, the wells were grouped according to the depth of each group and the group mean was calculated.

The average variation coefficient of the duplicate analyses was 0.043% (n=166). Pearson's correlation coefficient was used to verify if an association between fluoride concentration and the depth of the wells existed. The correlation analysis was undertaken with the total samples of the groups and using RStudio (Version 1.3.1073).

RESULTS

Most of the samples presented low fluoride concentration levels. Mean concentrations of fluoride in the studied wells ranged from 0.05 to 0.26 mgF$^-$ L^{-1} (Table 1) among all groups. The lowest means fluoride concentration (0.05 mgF$^-$ L^{-1}) was found in group A (≤ 50 m) and the highest (0.26 mgF$^-$ L^{-1}) was found in group E (201 - 250 m), as demonstrated in Figure 2. In groups B (51-100 m), C (101-150 m) and D (151-200 m), mean fluoride concentration was the same (0.07 mgF$^-$ L^{-1}), offering no risk for dental fluorosis.

Table 1 - Mean (standard-deviation) of fluoride concentrations (mgF$^-$ L^{-1}) and range in groundwater of private wells in the region of Maringá

Groups (well depth)	*N*	*F±SD (range) mgF$^-$ L^{-1}*	*Median mgF$^-$ L^{-1}*
A (0-50 m)	14	0.05 ± 0.01 (0.04 - 0.08)	0.06
B (51-100 m)	56	0.07 ± 0.09 (0.03 - 0.72)	0.11
C (101-150 m)	83	0.07 ± 0.05 (0.04 - 0.35)	0.19
D (151-200 m)	7	0.07 ± 0.04 (0.04 - 0.16)	0.10
E (201-250 m)	5	0.26 ± 0.37 (0.05 - 0.92)	0.48
F (> 250 m)	1	0.14 (0.14)	0.14
Total	**166**	**0.09 ± 0.15 (0.03 - 0.92)**	**0.29**

Source: Elaborated by author

However, one sample from Group E (201-250 m) was classified as presenting moderate risk of fluorosis, according to CECOL classification (0.92 mgF$^-$L^{-1}). However, this particular well was in the industrial area and the water used for industrial purposes only (Figure 2).

Figure 3 shows the scatter plot of all data. There was no correlation between fluoride concentrations and the depths of the wells (r^2 = 0.19, p<0.01).

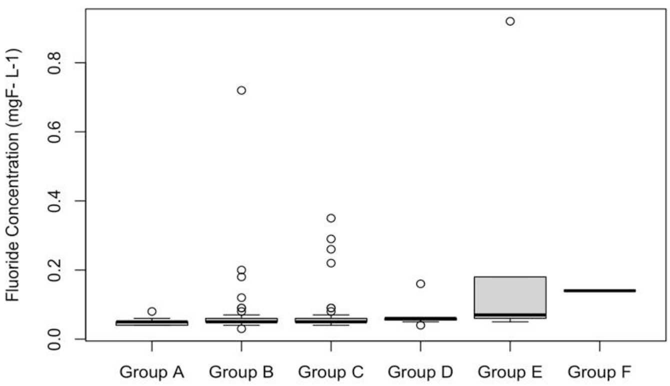

Figure 2 - Concentration of fluoride naturally occurring in the groundwater of private wells in the region of Maringá

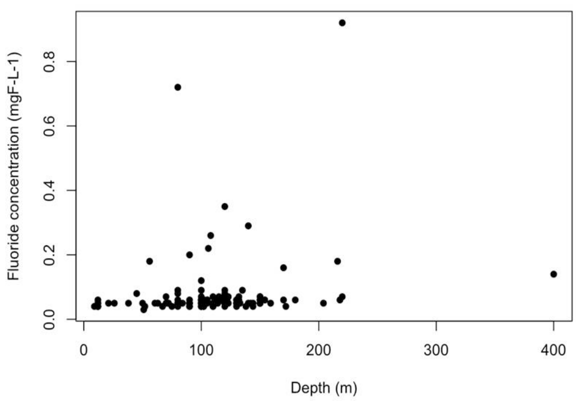

Figure 3 - Scatter plot of fluoride concentration (mgF$^-$ L^{-1}) and depth of wells (m)

DISCUSSION

This study evaluated fluoride concentration levels occurring naturally in artesian private wells in the region of Maringá, with depths ranging from 9 to 400 m. The findings reported in this study demonstrated that the vast majority of the studied wells presented low mean fluoride concentrations, ranging from 0.03 to 0.92 mgF$^-$ L^{-1}. Twelve samples from all groups showed fluoride concentration above 0.1 mgF$^-$ L^{-1}, demonstrating the presence of any type of rocks and soil with fluoridated compounds and rock porosity (Sajil Kumar et al., 2014).

There were few studies that evaluated fluoride in groundwater from artesian wells in the literature. A recent systematic review showed that fluoride concentrations between 1.2 and 4.8 mgF$^-$ L^{-1} have been found in groundwater in Brazil (Lima et al., 2019). In one of the included studies, high concentrations of fluoride (1.4 and 4.8 mgF$^-$ L^{-1}) were found in two rural communities in the State of Minas Gerais, which demonstrated a high prevalence of fluorosis (80.4%) in the local population (Ferreira et al., 2010). In another included study, fluorosis affected all the 27 children in a small city in the State of Espírito Santo supplied with drinking water from wells with a mean fluoride concentration of 2.4 mgF$^-$ L^{-1} (0.4 to 7.1 mgF$^-$ L^{-1}) (Carvalho et al., 2011).It is likely that all included studies in this systematic review had higher fluoride concentration in drinking water compared to our results because the aim of their study was to estimate the prevalence of dental fluorosis in Brazilian cities supplied with non-fluoridated water and in locations that used groundwater.

Of the 166 wells analyzed in the present study, only one sample showed a fluoride concentration (0.92 mgF$^-$ L^{-1}) classified as having a moderate risk of fluorosis (Centro Colaborador do Ministério da Saúde em Vigilância da Saúde Bucal, 2011). This location was in the industrial area of Maringá and the water was exclusively used for industrial purposes and not for human consumption. This finding may be related to the mineral composition of the soil, which changes according to the location of the well. The high fluoride values in groundwater of the Serra Geral Aquifer may be associated with the weathering of igneous rocks of the Serra Geral Formation, in which the natural sources of fluoride are related to the occurrences of fumaroles and magmatic gases, hydrothermal vents, volcanic glass and accessory minerals that replace hydroxyl ions (OH^-) during and after the crystallization of minerals, such as apatite, micas (especially biotite) and amphiboles and other pegmatitic minerals (topaz, tourmaline, fluorite, etc.) (Andreazzini et al., 2006).

The lack of correlation between fluoride concentrations and the depths of the wells ($r^2 = 0.19$) found in the present study is in disagreement with a previous study (Panagoulias Theodoros & Silva Filho, 2006). According to the authors, shallower wells presented significantly lower concentrations of fluoride than deeper wells, and attributed their findings to groundwater being diluted by the infiltration of rainwater and/or surface water. Although such a correlation was not found in the present study, the lowest mean concentration of fluoride (0.05 $\pm$ 0.01 mgF$^-$ L^{-1}) was also found in shallower wells, with depths up to 50 m (group A). Following this rationale, it would be expected that the deepest well (400 m – group F) in the present study presented the highest concentration of fluoride, which was not the case. Other factors such as temperature, pH (Sajil Kumar et al., 2015), presence of mineral complexes, precipitated ions and colloids, mineral solubility, ionic exchange capacity of minerals, granulometry, type of lithology and water resistance time, may all influence the concentration of natural fluoride in groundwater (Apambire et al., 1997). In addition, the number of samples in each group was not similar because this was the total available wells in the Maringá region according to the inclusion criteria of this study.

CONCLUSION

Based on the results and limitations of the present study, it may be concluded that the groundwater of private wells in the region of Maringá presented low concentrations of naturally occurring fluoride. Nonetheless, one of the studied wells demonstrated a concentration of fluoride indicative of moderate risk of fluorosis. No correlation was found between the concentrations of fluoride and the depth of the studied wells. The present findings reinforce the importance of constant water quality monitoring by local professionals and authorities.

Only 18% of all private wells registered at the Water Institute of Maringá were included for analysis in this study. This low level of inclusion was due to limitations such as, the lack of authorization by well owners, incomplete records, as well as problems with water sample collection in sites that had been abandoned. Future studies should map the concentrations of fluoride and other ions in private wells, associating results with the geological data of each location to build a database of groundwater which is safe for human consumption. Thus, more studies involving multidisciplinary teams of geologists and health professionals are required to ensure the quality of drinking water supplied to the population.

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Josely Emiko Umeda: responsible for analysis of data, revising the article critically for important intellectual content, and for final approval of the version to be published;

Jaime Aparecido Cury: responsible for the conception of the study, revision of the article critically for important intellectual content and for final approval of the version to be published;

Livia Maria Andaló Tenuta: responsible for the design of the study, revising the article critically for important intellectual content, and for final approval of the version to be published;

Mitsue Fujimaki: responsible for conception and design of the study, agreement to be accountable for all aspects of the work in ensuring that questions related to the accuracy or integrity of any part of the work are appropriately investigated and resolved and revising the article critically for important intellectual content, and for final approval of the version to be published.