

# Fluorine in Water and Dental Fluorosis in a Community of Queretaro State Mexico

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Received May 11<sup>th</sup>, 2011; revised June 18<sup>th</sup>, 2011; accepted July 23<sup>rd</sup>, 2011.

## ABSTRACT

The community La Llave, Queretaro State, Mexico, has not been identified by the sanitary authorities as living in a hazard area related to dental fluorosis. However, a high concentration of fluoride is found in their drinking water causing them dental fluorosis. Physical-chemical analysis was carried out in the groundwater sources to determine the prevalence of dental fluorosis risk and caries accordingly to criteria of The World Health Organization, and 154 school children of ages 10 to 13 years were examined. As a result, 1.9 mg/L of fluorine concentration in drinking water was obtained; dental fluorosis presence was detected in the school children with an incidence of about 98%, in 47% of cases severe fluorosis with a dental caries index of 3.06 was traced. The groundwater sources in La Llave community contains fluorine above the limits dictated by Mexican regulations, producing serious repercussions on the health of school children, with an unnaturally high incidence of dental fluorosis.

**Keywords:** Fluorosis, DEAN Index, Groundwater, La Llave Querétaro, Mexico

## 1. Introduction

The Community of La Llave, Queretaro State, where this work was carried out, is located at 1900 m above mean sea level (amsl) on the southern part of the plain that extends from El Zamorano volcano, north of Queretaro City, to San Juan del Rio City, located in the central part of Mexico (**Figure 1**). The plain contains some volcanic reliefs hardly 100 m heights above the plain surface, such as Cerro La Cruz and La Carbonera, at whose base La Llave is located.

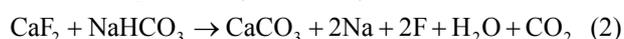
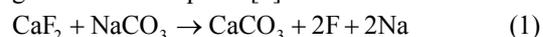
The geological framework of the region is constituted by rocks of chemical composition ranging from intermediate to acid, represented by interstratified ignimbrites with pumice tuffs, both expelled through the Caldera of Amealco. These materials are covered on the plain by alluvial deposits. Alluvial deposits consist of sediments of several sizes ranging from clay and sand to rounded blocks of different composition derived from pre-existing nearby rocks. These deposits form the most surface layer of the plain and have a thickness from 5 to 30 m.

Comisión Estatal de Agua [1] shows the existence at La Llave's zone of an aquifer formed jointly by fractured rocks and granular sediments that reaches a thickness of

200 m, settlements of the region obtain their water supply from this aquifer. The geological model shows the existence of volcanic bodies of Olocene Age, suggesting the presence of high temperature in the rocks beneath, this feature is considered in determining the temperature (26°C) of tapped groundwater by wells.

The La Cruz, La Carbonera, and Viejo hills are constituted by ignimbrites, rhyolites, and tuffs rocks. Containing abundant fluorine where groundwater dissolves accessory minerals contained in rocks such as, mica, amphibole and apatite, the later releases fluorine to groundwater.

On the other hand, groundwater rich in HCO<sub>3</sub>, during the interaction between water and mineral (fluorite) the following reaction takes place [2]



According to this reaction high contents of NaHCO<sub>3</sub> in water increase the CaF<sub>2</sub> dissolution in the rock releasing fluoride in the groundwater. The dissociation of fluorine-rich minerals, is also favored by the existence of rich mineral calcite (CaCO<sub>3</sub>), as shown in the following reactions [3].

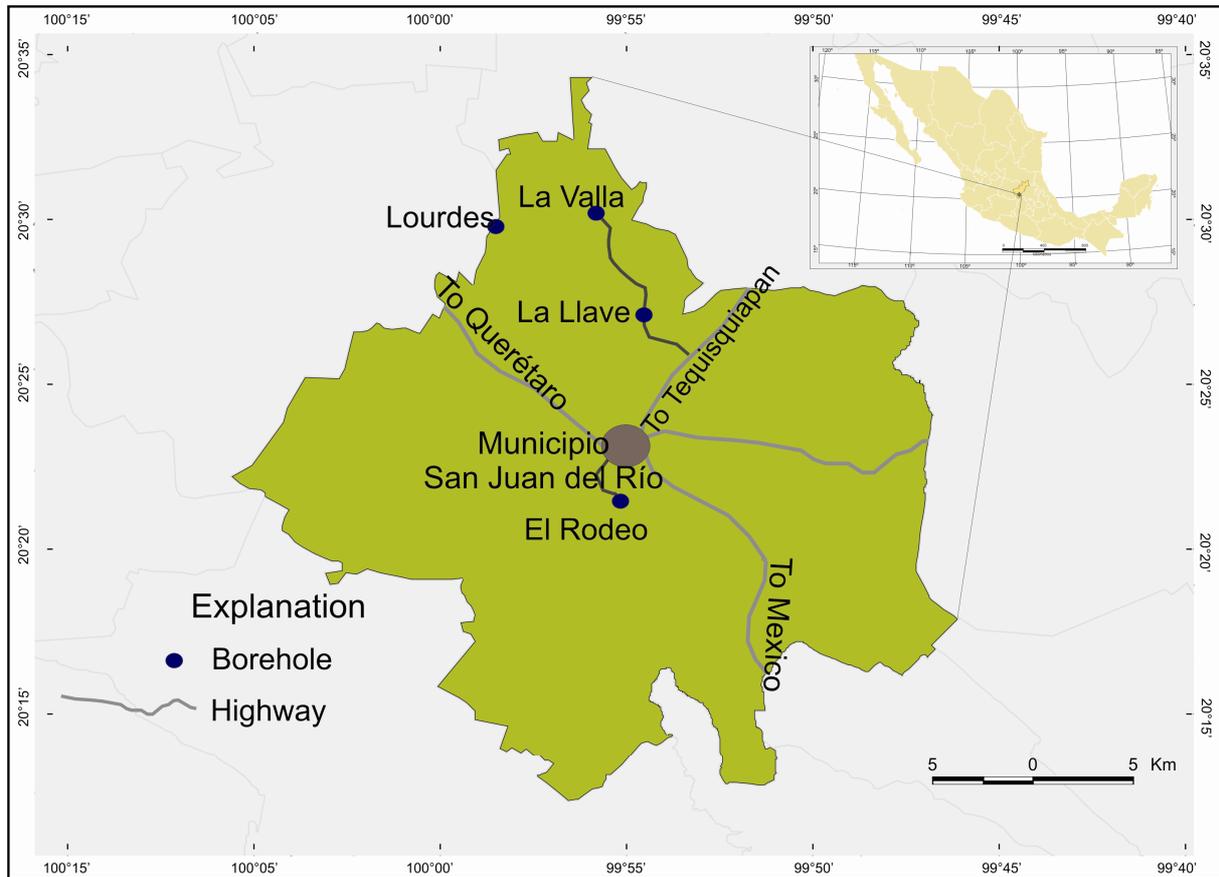
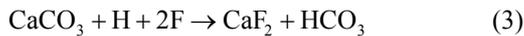


Figure 1. Location of groundwater sample borehole.



$$K = \frac{\{a(\text{HCO}_3)\}}{\{a(\text{H}) * a(\text{F})\}} \quad (5)$$



$$K_{\text{fluorita}} = (\text{Ca}^{2+})(\text{F}^-)^2 = 10^{-10.57} \text{ at } 25^\circ\text{C} \quad (7)$$

$$\text{or, } \log K_{\text{fluorita}} = (\text{Ca}^{2+}) + 2\log(\text{F}^-) = -10.57 \quad (8)$$

where K is the equilibrium constant, a is the activity. If pH is constant, the activity of fluorine is directly proportional to HCO<sub>3</sub>. This relationship is independent of Ca<sup>2+</sup> produced by the low solubility of CaF<sub>2</sub>.

Fluorine content in groundwater is positively correlated with Na<sup>+</sup> and pH, the presence of F<sup>-</sup>; is associated with sodium bicarbonate type water with a high pH. However, it has a negative correlation with Ca<sup>2+</sup>, this indicates that the fluorine concentration is associated with reactions where Na<sup>+</sup> and pH increase, and Ca<sup>2+</sup> decreases as a result of cation exchange, or precipitation of calcite. This suggests that F<sup>-</sup> concentration is controlled by the Fluorite (CaF<sub>2</sub>) equilibrium, *i.e.*, the limits of fluorine are determined by the calcium ion [4-8].

According to Edmunds [9], the upper limit on fluoride activity in aqueous solutions is controlled by the solution of the products, K<sub>fluorite</sub>

Those equations show that in the presence of fluoride, the fluorine contents are inversely proportional to Ca<sup>2+</sup>. The silicates dissolution in alkaline rocks, where oligoclase is plagioclase dominant, rich in sodium and calcium deficient, provides sodium and fluorine enrichment in groundwater [6,10,11]. Based on the foregoing, F<sup>-</sup> can be removed by precipitation when there is a high concentration of Ca<sup>2+</sup> [7].

Fluorosis is a disturbance caused by excessive ingestion of fluorine during the odontogenic stage [12]. In Mexico, the prevalence and incidence of fluorosis have increased during the past years, as a result of ingestion of fluorine during dental formation by means of water, food prepared with fluorated salt, and inadvertent ingestion of this element during dental brushing with fluorated toothpaste, as well as hidden fluorine in beverages [13,14].

In light of dental spots observed to children of this

community and the lack of reports related to the fluorine concentration in water in this area, it was considered to be important to carry out the present study. The goal of this study is to determine the fluorine concentration in drinkable water, as well as to analyze the effects of fluorosis and dental caries in school children residents in La Llave town.

## 2. Methodology

### 2.1. Water Sample

Samples of water were taken for physical chemical analysis in polyethylene bottles prewashing in lab with hydrochloric acid (5%) and distilled water solution. Air bubbles were avoided to limit any potential change in the original chemical water composition. A 100 ml bottle was taken without adding any preservation agent or filtered, this sample was used in anions determination. The sample for cations and trace elements was taken in a bottle of 125 ml; it was filtered using a cellulose acetate membrane of 0.45 $\mu$ m and was then acidified (to a pH  $\approx$  2) by means of nitric acid of high purity (in order to avoid precipitation and absorption of elements on the walls of the bottle). Analyses were carried out by a mass spectrometer with inductive plasma coupling (ICP-MS). The detection limits obtained with this equipment is very low (less than 0.001 mg/l). The analyses were performed by Actlabs of Canada.

### 2.2. Clinical Examination

For convenience, the 154 subjects, between 10 to 13 years old, were checked; all of them are residents of the community La Llave, Municipality of San Juan del Rio, Queretaro. Only children with explicit permission from their parents participated in this study, their residence greater than 10 years in the zone was also demonstrated. Children with background of metabolic disease or those under orthodontic treatment were excluded from the study.

The clinical examination was carried out with natural light without desiccation, using dental plain mirrors, #5, without amplification. Results were recorded in individual files. The exam was performed by two qualified and standardized observers accordingly to the WHO criteria [15]. Among the Dean Fluorosis Index (ID) and dental caries index (DMF-T and DMF-S), when the Kappa test was applied, congruence was observed in the diagnosis criteria of 87% and 95% for fluorosis caries index respectively.

## 3. Results and Discussion

### 3.1. Water Quality

According with the prevailing geological framework, the

chemical quality of the water that the population drinks depends of the type of rock material it circulates through. Consequently, fluorine concentration in the aquifer is associated to rock-water interaction and is determined on the one hand by the abundance in the minerals forming the rocks [16,17], as well as by the solubility of the minerals, and the water subsaturation as related to calcite. In as much as the calcium limits the solute or dissolved fluorine concentration [18], **Table 1** contains the chemical analytical results for this paper, where it can be observed that the greater calcium content the lower content of fluorine, and vice-versa. Based on the solubility control of fluoride it may be removed by precipitating it by maintaining a high concentration of Ca<sup>2+</sup> [7], these researchers propose that fluorine dispersion and transport are determined by pH and water temperature.

In a laboratory experiment, the quantity of solute or dissolved fluorine in acidic rocks was found to be almost double the quantity to that in basic rocks; this also showed the close relationship between fluorine content in groundwater and the nature of strata it flows through [19].

In most of the towns of Mexico, the water supply is derived from groundwater sources, whose natural mineralization has increased in some elements such as fluorine [20], In many cases, the result depended on the way the water is extracted. *i.e.*, an intense extraction of ground

**Table 1. Groundwater chemical composition of the wells analyzed in this study (Lourdes, La Valla, and La Llave), values in mg/L.**

Parameter	Lourdes	Llave	Valla	WHO (g/v)
pH	8.05	8.10	8.07	n-h
Water temp	31.0°C	30.0°C	25.9°C	n-h
Alcalinity	140.00	107.00	150.00	n-h
Ca <sup>2+</sup>	22.00	12.10	21.00	n-h
Mg <sup>2+</sup>	5.61	3.16	7.92	n-h
K <sup>+</sup>	10.50	10.10	13.70	n-h
Na <sup>+</sup>	40.00	45.00	42.00	n-h
Cl <sup>-</sup>	3.79	4.84	3.95	n-h
SO <sub>4</sub> <sup>2-</sup>	6.91	7.35	10.40	n-h
Si	37.00	43.50	39.20	n-h
Mn	0.05	0.01	0.01	0.4
Fe	0.10	0.10	0.10	N-H
F <sup>-</sup>	<b>0.52</b>	<b>1.94</b>	<b>0.51</b>	<b>1.5</b>

WHO (g/v), Guideline value; n-h = Not of health concern at levels found; in drinking-water.

water causes the drawdown of the water-table in wells and induces water flow from the deeper levels [21], where their mineral content might contribute with fluorine in higher concentration than those considered, as optimal for fluorosis prevention. However, in wells, where water is extracted from low depth obtaining local or intermediate flows with traces of fluoride can reduce significantly the levels of fluoride in the obtained water.

The water extracted from the well in La Llave contained more fluorine than those in neighboring towns as Lourdes and La Valla (**Table 1**). However, that content is low related to those reported in other regions in the country also of volcanic origin, such as San Luis Potosí (4 - 7 mg/L), [22], Tesisitan Jal. (4.7 mg/L), [23], and Durango (5.7 mg/L) [24]. In agree to groundwater flow systems model [25], the records of fluorine content and temperature in the first three wells, led to assumption that the obtained water is related to a groundwater flow system of local type; whereas, for the other regions, it is more related to an intermediate flow.

### 3.2. Dental Fluorosis

The prevalence of students affected by dental fluorosis was 98%, this dental defect is regarded as a biomarker of chronic toxicity caused by continuous exposure to fluorine. The Community Dean Index (CDI) was  $3.06 \pm 1$ . Moreover, approximately 48% of those children showed severe fluorosis (**Table 2**) with loss of dental enamel structure requiring prosthetic and anti-aesthetic actions.

The **Table 3** shows the mean of the different index by gender, and the percentage of children with dental caries was 70% with a DMFT of  $2.7 \pm 2.7$  and a DMFS of  $3.6 \pm 3.7$ .

The prevalence of caries was greater in those children with high severe fluorosis (**Figure 1**). There is information about low levels of fluorosis preventing caries, but severe levels constitute a risk factor for diseases since the structural defects due to fluorosis alterations cause spots where the bacterial plaque can remain [26].

Present study coincides in the general objective regarding the communities of La Fuente, Santillan, and San José, all located close to San Juan del Rio Queretaro, where a prevalence of 87% of fluorosis in children from 11 to 13 years of age, has been reported [27].

In the village of La Llave, the dental fluorosis could be related not only with the fluoridated drinking water above optimal limit level, but also by preparing meals and milk formula, in addition fluoride is also ingested through foods prepared with fluoridated salt.

Some authors report that the more fluorosis affected populations are those located at high altitude [28]. At 1500 m high amsl renal filtration and filtration of some substances are modified to favor an increase of fluorine

**Table 2. Severity of fluorosis according to the DEAN index affecting scholars of 10 - 13 years old at the Community La Llave.**

Severity of Fluorosis*	F	%
None Fluorosis	3	2.0
Very slight	13	8.0
Slight	29	19.0
Moderate	36	23.5
Severe	73	47.5
Total	154	100.0

\*Criteria of Dean.

**Table 3. Dental fluorosis and caries index by gender in scholars of 10 to 13 years old at the Community La Llave.**

	IDC		DFMT		DMFS	
	X	DE	X	DE	X	DE
Men	3.05 - 3.04	1.02	2.75	2.8	3.7	3.8
Women	3.11 - 3.08	1.06	2.82	2.6	3.55	3.5

X = Mean. DE = Standard deviation. IDC = Community Dean Index. DFM-T = Decayed Filled Missed Teeth. DMFS = Decayed Filled Missed Teeth by Surface

in the blood, which eventually will be concentrated in teeth and bones [29]

### 4. Conclusions

Fluorine concentration in water supplied to La Llave village had a 1.9 mg/L concentration. Continuous chemical monitoring of water quality is considered relevant; it has to be performed in the wells and water supply plants of nearby zones to geographically determine an acceptable and required fluorine characterization.

The results obtained in this paper highlights differences of dental fluorosis showing that the maternal and offspring population are exposed to various sources of fluoride that cause fluorosis; these sources are:

- Boil drinking water
- Food prepared with water having high fluoride content
- Cooking with fluoridated salt

Boiling water consumption rises from 60% to 70% the original concentration of fluoride, which puts at risk for developing fluorosis in permanent teeth [14,30,31].

A high fluorosis prevalence was observed, the IDC points a severe public health problem, so it is important to consider that well-water purification systems will be installed in La Llave village or that other sources for supplying potable water will be needed. It appears a valid

proposal that the inhabitants receive information related to the fluoride sources related to dental fluorosis risks.

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