FLUORIDE OCCURRENCE IN TAP WATER AT "LOS ALTOS DE JALISCO" IN THE CENTRAL MEXICO REGION

[1]R. Hurtado, [2]J. Gardea-Torresdey, and [2]K. J. Tiemann

[1]Environmental Science and Engineering Ph. D. Program, Center for Environmental Resources Management, The University of Texas at El Paso, 505 W. University Ave, El Paso, TX 79902; Phone: (915) 747-8626; Fax: (905) 747-5145; e-mail: rhurtado@miners.utep.edu.
[2]Department of Chemistry & Environmental Science and Engineering, The University of Texas at El Paso, 505 W. University Ave, El Paso, TX 79902; Phone: (915) 747-5359; Fax: (905) 747-5748; e-mail: jgardea@miners.utep.edu.

ABSTRACT

In Mexico, like most countries in the world, the maximum contaminant level (MCL) of fluoride in drinking water is 1.5 mg/L. The toxic effects of fluorides, other than dental and skeletal *fluorosis*, include enzyme damages, which result in a wide range of chronic diseases such as genetic damage, premature aging, mental retardation, cancer, bone pathology, and others. In order to characterize the content of fluoride in the drinking water at *Los Altos de Jalisco* region, tap water and well water samples from 23 different sites were electrochemically analyzed using the approved U.S. EPA ion-selective method (Method 340.2). *Los Altos de Jalisco* is located in the Trans-Mexican Volcanic Belt (TMVB), also known as Neo-Volcanic Axes, where the many groundwater wells have been found to have fluoride concentrations over the MCL. Based on the results of this study, the content of fluoride in the water samples ranged from 0.14 to 12.97 mg/L. The cities where we found reticulated water with fluoride concentration over the MCL are Temacapulín (11.25-12.97), Mexticacán-1 (6.64-7.50), Lagos de Moreno (4.77-4.96), Encarnación de Díaz (4.25-4.40), and Tepatitlán-1 (1.83-6.79). While everyone may not be affected, an important fraction of the total population (approximately 200,000) of these cities is under serious health risk and should be of a major concern.

Key words: *fluoride, fluorosis, fluorite, tap water, Los Altos de Jalisco*

INTRODUCTION

The *Los Altos de Jalisco* region, with 16,410 km^2 of surface area and a population over 600,000 people, is located in the northeastern part of the state of Jalisco, Mexico (see Figures 1 and 2). *Los Altos de Jalisco* is conformed by 20 counties or municipalities with populations ranging from 4,907 to 124,927 inhabitants. Table 1 shows the surface area and its corresponding population for each county in the *Los Altos de Jalisco* region (INEGI, 1995; ITESM, 1995).

Drinking water, in *Los Altos de Jalisco*, has two origins: (1) surface water, and (2) groundwater wells. In the case of surface water, the mineral content is low and is mainly due to rainwater, which is stored in surface reservoirs. The mineral content in rivers, lakes, and reservoirs is mainly due to the discharge of spring waters into the main streams. Nevertheless, it is possible to find polluted waters from anthropogenic sources. The use of groundwater for drinking purposes should be decided after chemical characterization, because there are many hydrothermal zones where the mineral content is over the Mexican national standards.

Our interest to evaluate the quality of the drinking water in *Los Altos de Jalisco* is because the region is located in the Trans-Mexican Volcanic Belt (TMVB), also known as Neo-Volcanic Axes (*Eje Neovolcánico*, in Spanish language). The TMVB is a Pliocene-

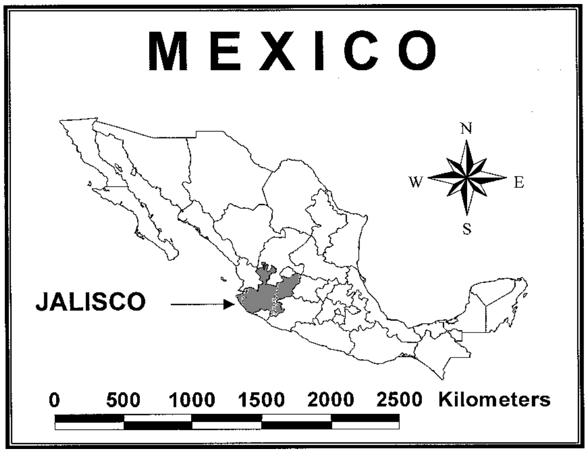

Figure 1. Location of the state of Jalisco in the country of Mexico.

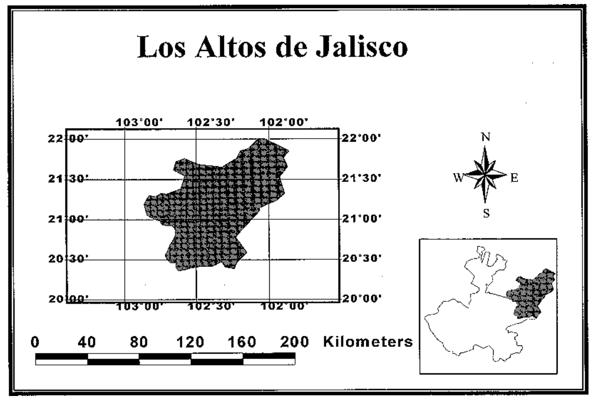

Figure 2. Location of the *Los Altos de Jalisco* region.

Quaternary calc-alkaline province, which crosses Mexico between 19° and 21° N latitude, and is characterized by hydrothermal activity (Campos-Enriquez and Garduño-Monroy, 1995).

In the TMVB province, the chemical composition of the geological materials indicates that groundwater wells might have considerable amounts of toxic elements, such as copper, strontium, zinc, arsenic, lead, chromium, selenium, aluminum, and fluoride.

Our chief concern is that the large amount of people living in *Los Altos de Jalisco* are under high health risk because they are consuming groundwater without any treatment or support from the water and health agencies. However, we found information in the regional newspapers where the health authorities mention that the people in *Los Altos de Jalisco* are consuming safe water.

The result of our evaluation, which is shown below, indicates that several cities are consuming water with levels of fluoride over the Mexican national standard of 1.5 mg/L. Nevertheless, most people in the region are consuming bottled water and are cooking food with boiled water.

The highest health risk corresponds to children in the schools, mainly public schools, where they consume considerable amounts of tap water directly from the faucets.

The objective of this manuscript presentation is the chemical characterization of the tap water in *Los Altos de Jalisco* in order to help in the determination of a practical solution for each site where the quality of the water is not satisfying national and international standards.

GEOCHEMISTRY OF FLUORIDE

Fluorine is found in the environment as fluorides, mainly because it is the most electronegative and reactive of all chemical elements.

Practically all natural waters have fluoride, ranging from trace levels to several dozen milligrams per liter. Exceptionally higher values can be found, such as in the case of some lakes in Kenya where the content of fluoride is over 2,000 mg/L (Gaciri and Davies, 1993). The average concentration of fluoride in seawater is 1-1.3 mg/L; however, rivers, lakes, and groundwater generally have concentrations of less than 0.5 mg/L. In contrast, high fluoride concentrations are frequently associated with hydrothermal waters resulting from volcanic activity and fumarolic gases (WHO, 1996). Temperature, pH, presence or absence of complexing or precipitating ions and colloids, solubility of fluorine-bearing minerals, anion exchange capacity of aquifer materials (OH^- for F^-), and the size and type of geological formations traversed by water are the most important factors controlling the concentration of fluoride in natural waters (Apambire et al., 1997).

Table 1. Municipality surface and population in *Los Altos de Jalisco.*

Municipality	Surface (km^2)	Population (1995)
Acatic	362	17,906
Arandas	1,238	70,901
Cañadas de Obregón	471	4,907
Cuquío	880	17,034
Encarnación de Díaz	1,297	43,875
Jalostotitlán	481	26,297
Jesús María	570	20,356
Lagos de Moreno	2,849	124,927
Mexticacán	177	7,328
Ojuelos de Jalisco	1,317	25,743
San Diego de Alejandría	432	6,389
San Juan de los Lagos	874	53,366
San Julián	268	13,700
San Miguel el Alto	511	27,237
Teocaltiche	914	37,164
Tepatitlán de Morelos	1,533	109,300
Unión de San Antonio	688	15,172
Valle de Guadalupe	516	5,663
Villa Hidalgo	511	13,715
Yahualica de González Gallo	521	23,539
TOTAL	**16,410**	**664,519**

Sources: (ITESM, 1995), and (INEGI, 1995)

Some of the minerals where fluoride can be found in significant fractions are fluorite (CaF_2), apatite, villiaumite (NaF), cryolite ($Na_3 AlF_6$), topaz ($Al_2SiO_4(F,OH)_3$), mica, amphiboles, and rock phosphate. The mineral that predominantly determines the concentration of fluoride found in natural waters is fluorite. Solubility product (k_{sp}) of fluorite, at 20 °C, is 3.9×10^{-11}. This low solubility value implicates that waters with low content of calcium should have high fluoride concentration.

The apatite group plays an important role in determining the level of toxicity of waters due to the additional dissolution of elements such as arsenic and lead. The general formula of the apatite group is

$$A_5(BO_4)_3(OH,F,Cl)$$

where

A= Ca, Ba, Na, Pb, Sr, La, and Ce

B= P, V, and As

BO_4 = CO_3 and SiO_4

One important member of this group is fluorapatite, $Ca_{10}(PO_4)_6F_2$.

HEALTH EFFECTS

There is a big controversy on the usefulness of fluoride in protecting tooth decay. In 1984, the World Health Organization (WHO) recommended keeping the concentration of fluoride in the range of 0.5 to 1.5 mg/L (WHO, 1996) in order to prevent dental caries, especially in children. The ingestion of low doses of fluoride could protect tooth enamel from dissolution. Tooth enamel consists of *hydroxyapatite* ($Ca_{10}(PO_4)_6(OH)_2$). If a reasonable level of fluoride ion is present in the diet during the growing phase of teeth, a significant amount of *fluorapatite*, ($Ca_{10}(PO_4)_6F_2$, is incorporated in the enamel in place of the *hydroxyapatite*. *Fluorapatite* is less soluble in mouth acids; hence fluoride-containing teeth are less susceptible to decay.

After the WHO recommendation, an important number of countries legislated to set 1.5 mg/L as the maximum contaminant level (MCL) for fluoride in drinking waters. Nevertheless, the USEPA, after an evaluation by the National Research Council (NRC, 1993), kept 4.0 mg/L of fluoride as MCL or *primary standard* for drinking water and 2.0 mg/L as MCLG (maximum contaminant level goal) or *secondary standard*. Instead of a health problem, the USEPA considers mild dental fluorosis as an aesthetical one.

An important number of epidemiological studies have shown possible adverse effects of the long-term ingestion of fluoride. Concentrations above the MCL value carry an increasing risk of dental fluorosis, and much higher concentrations lead to skeletal fluorosis, bone cancer, premature aging, mental retardation, and other health concerns.

In several districts of Rajasthan, India, the population is consuming water with fluoride concentration up to 44 mg/L. Permanent deformities, joints pains, general debility, and misery has been the consequence of consuming water with such high mentioned levels of fluoride (Agarwal et al., 1999).

Endemic fluorosis has been reported in several regions of Mexico, where people are consuming water with fluoride contents over the MCL of 1.5 mg/L, which is the Mexican national standard for drinking water. Not only tap water, but also bottled waters with fluoride ranges between 0.33 to 6.97 mg/L were found in the city of San Luis Potosi, Mexico (Grimaldo et al., 1995). Similar values, between 0.7 and 5.6 mg/L, were detected in the city of Durango, Mexico (Ortiz et al., 1998). In both cities, San Luis Potosi and Durango, the health risk is higher than normal because the intake of fluoride comes not only by drinking water with high levels of fluoride, but also by eating food cooked with boiled water.

In 1996, the WHO concluded that, in setting national standards for fluoride, it is important to consider climatic conditions, water intake, and intake of fluoride from other sources such as food and air (WHO, 1996). In tropical countries, where people consume considerable amounts of water, the health risk is higher. In addition, high levels of fluoride in the air have been reported in some regions of China from the combustion of high fluoride-bearing coal (Wang et al., 1999). Therefore it is important to understand the total possible exposure to fluoride before assessing the potential public health risk.

EXPERIMENTAL

Sampling

Tap water samples were collected in clean fluoride-free plastic bottles, directly from home faucets in *Los Altos de Jalisco*. Trace-grade nitric acid was added in order to preserve the samples for future ICP-MS analysis. The samples were then stored at approximately 5 °C. The same procedure was used for the one river sample obtained.

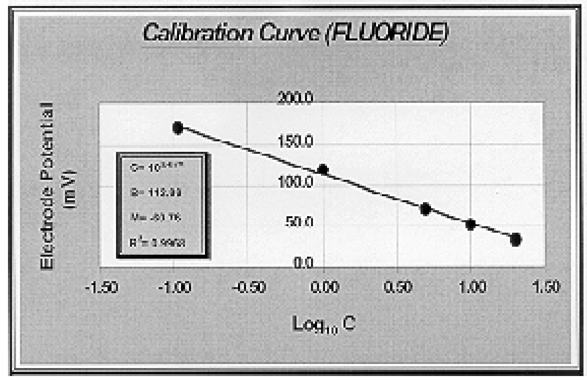

Figure 3. Calibration curve for determination of fluoride in tap waters.

METHOD OF ANALYSIS

Fluoride concentrations were determined electrochemically, using the approved USEPA ion-selective method (Method 340.2). This method is applicable to the measurement of fluoride in drinking water, surface and saline waters, and domestic and industrial wastes in a range of concentration from 0.1 up to 1000 mg/L (USEPA, 1991). A detailed explanation of this analytical method has been reported before (G. Rum et. al., 2000). The electrode used was a Fluoride/Combination Fluoride Electrode – Orion 96-09, which was coupled to an Orion 420A Electrometer (pH meter). Standards were prepared from a stock solution (100 mg/L) of sodium fluoride. Figure 3 shows the calibration curve prepared from the measurement of the potential (mV) of five standard solutions (0.1, 1.0, 5.0, 10.0 and 20 mg/L) where the R^2 value was 0.9968. Three replicates of each sample were analyzed and the average is reported herein.

RESULTS AND DISCUSSION

The total number of sites where water samples were collected was 23; one corresponds to a river sample (Rio Verde river);

three correspond to samples directly collected from well bore (Cañadas-2, Mezcala-1, and Jalostotitlán); and 19 were tap water, collected directly from home faucets. Tepatitlán de Morelos was the municipality where the highest number of samples was taken (six samples, corresponding to three different cities). The Cañadas de Obregón municipality was where the highest concentration of fluoride was found, which consisted of four samples and one river sample. The sample from Temacapulín (a small village with a population of around 400 people) had the greatest fluoride concentration of 12.97 mg/L.

Table 2 shows the fluoride concentration of each site sampled across the *Los Altos de Jalisco* region. The fluoride content in the water samples ranged from 0.14 to 12.97 mg/L. These values indicate not only the differences in the source of water (surface or groundwater) used in the region, but also different mineralogical characteristics in the geological structures of the area.

The cities where fluoride concentrations were found to be over the MCL are Temacapulín (11.25-12.97 mg/L), Mexticacán-1 (6.64-7.50 mg/L), Lagos de Moreno (4.77-4.96 mg/L), Encarnación de Díaz (4.25-4.40 mg/L), and Tepatitlán-1 (1.83-6.79 mg/L). The total population of these cities is around 200,000 people. While the entire population is not at risk, a large portion of this population is exposed to a serious potential health risk and should be of a major concern.

The fluoride concentration of the Rio Verde River (1.94-3.05 mg/L) was over the Mexican national standard of 1.5 mg/L. This river is considered important since it is feeding the *Rio Grande de Santiago* that drains into the Chapala Lake. The city of Guadalajara, one of the most important cities of Mexico, consumes water directly from the Chapala Lake Reservoir.

With the exception of Temacapulín, the levels of fluoride found in the remaining five cities of *Los Altos de Jalisco* (mentioned above) are very similar to the values reported by Grimaldo et al. (1995) in San Luis Potosi, and by Ortiz et al. (1998) in Durango, México. Although there is no data on the percentage of people (at *Los Altos de Jalisco*) who prepare their food with boiled water, representing an additional risk factor for human exposure to fluoride, we can assume that these are similar to the values reported by Ortiz, et al. (1998), which is approximately 90% of the population. This is due to the similarities in culture and culinary costumes. However, one important difference between *Los Altos de Jalisco* and San Luis Potosi and Durango is that the ambient temperature of *Los Altos de Jalisco* is lower than both cities. This would imply that the intake of fluoride could be less at *Los Altos de Jalisco*.

CONCLUSIONS

After obtaining samples from approximately 12,000 km^2, 74% of the *Los Altos de Jalisco* region, we found five cities and one river containing water with fluoride concentrations in the range of 1.83 to 12.97 mg/L (all over the Mexican national standards).

In the case of some cities where there are wells with different concentrations of fluoride

Table 2. Fluoride content (mg/L) of water samples at *Los Altos de Jalisco.*

Municipality	Jun-99	Aug-99	Nov-99	Feb-00
Arandas				
(Arandas)	—	—	0.25	0.14
(Santa María del Valle) *	—	—	—	0.29
Cañadas de Obregón				
(Temacapulín) **	12.97	11.25	11.42	12.29
(Rio Verde)	2.50	1.94	3.05	2.84
(Cañadas-1) *	0.25	0.28	0.44	0.25
(Cañadas-2) *	—	—	0.68	0.29
Cuquío *	—	—	—	0.25
Encarnación de Díaz *	—	—	4.25	4.40
Jalostotitlán *	—	—	1.23	1.10
Lagos de Moreno *	—	—	4.77	4.96
Mexticacán				
(Mexticacán-1) *	—	—	7.50	6.64
(Mexticacán-2) *	—	—	—	6.33
San Diego de Alejandría *	—	—	0.55	0.80
San Juan de los Lagos	—	—	0.64	0.79
San Julián *	—	—	0.49	0.47
San Miguel el Alto	—	—	1.04	0.65
Teocaltiche	—	—	1.18	1.15
Tepatitlán de Morelos				
(Mezcala-1) *	0.51	0.40	—	—
(Mezcala-2)	0.17	0.24	—	—
(Capilla de Guadalupe)	—	—	0.36	0.23
(Tepatitlán-1)	0.15	0.24	Ñ-	Ñ-
(Tepatitlán-2) *	2.23	1.83	6.79	6.75
(Tepatitlán-3) *	—	—	0.64	0.23

* from groundwater well; ** from spring; others from surface reservoir

(such as in Tepatitlán), we recommend one of the following alternatives: (a) close wells that are delivering water with fluoride concentration over the MCL; (b) mix the water of two or more wells in order to deliver water within national standards or; (c) use water wells with fluoride content over the MCL for other applications, such as irrigation of public green areas.

In small towns and villages, such as Mexticacán and Temacapulín, where the population is relatively low, we have to work for the development and/or implementation of non-sophisticated methods of fluoride removal. An additional program of environmental education needs to be developed.

A more complete sampling program will be performed in order to have a better evaluation that will allow recommendations for specific and practical methods of fluoride control for each site. Nevertheless, our attention will be focused on the higher population cities, such as Lagos de Moreno, Tepatitlán, and Encarnación de Díaz, where we are planning a more intensive program of sampling and evaluation.

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