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Research Paper

Increase in the concentration of fluorine compounds as indicator of the decrease in the dynamic level in groundwater in a population of western Mexico

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ABSTRACT: Fluorides are compounds that contain the fluorine ion and are found in most soils, in highly variable concentrations. In the present work, the concentration of fluorides in deep wells that supply drinking water to an average city of approximately 110,000 inhabitants, located in the upper region of the state of Jalisco, was related with the dynamic level of extraction (depth), finding that of the 49 deep wells that the city regularly operates, 25 % have fluoride concentrations that exceed the maximum permissible limit for drinking water, reporting values of up to 11.44 mg / L. The highest concentrations of fluorides are located mainly in wells located to the West, Northwest and Southeast of the city center, some possible causes of the high concentration of fluorides can be: the number of wells operating, the high extraction volumes and the hours that operate daily, consequently dynamic levels of average extraction are observed in the study area of 185 meters.
KEYWORDS: Dynamic Level Decrease, Fluoride Concentration, Groundwater, Western Mexico

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I. INTRODUCTION

For decades it has been suggested that fluorine is a constituent considered one of the most volatile and characteristic in igneous rocks of felsic character [1], in the earth's crust its average concentration is 625 mg / L. Fluorine is a very important component in magma, which favors its increase in water; its solubility within magma is mainly associated with cations where sodium and potassium influence more than calcium and magnesium [2].

Gunnar and collaborators document that for a particular flow system (local, intermediate or regional) the concentration of fluorides increases from the recharge zone reaching maximum values in the discharge zone. Therefore, it is important to highlight the role of hydrogeological functioning in the knowledge and evaluation of different chemical responses of water, in this case fluoride, to understand its incidence in groundwater sources and how it affects the health of the population that consumes it.

The main factors that intervene to increase the concentration of fluorides in groundwater are: the type of minerals of origin, the residence time of the water with the rock, the temperature due to the depth of circulation and the pH [3], as well as the activity and solubility of the minerals themselves, the porosity of the soil and the presence of elements that induce fluoride to conjugate, also vary significantly over time and the distance between two groundwater wells when it is equal to or greater than 500 meters [4].

According to the Stockholm International Water Institute the constant consumption of water with a composition not suitable for health, is the possible origin of various diseases in the skin, teeth, bones and internal organs such as the liver. Therefore, the presence of many compounds and chemical elements dissolved in water should not exceed the adequate levels allowed for its adequate and safe human consumption [5].

The ionic strength of fluorides also influences the solubility of the candidate mineral to dissolve and ion exchange reactions [6]. Thus, the minerals that commonly provide fluoride are few or moderately soluble and release the fluoride ion to the water slowly [7], although the rate of weathering of micas and amphiboles is low, the fluoride ion is released from them from their position. hydroxyl, which increases the levels of dissolved fluoride compounds [8].

Among the most common minerals that provide fluorides to groundwater are: halides, phosphates, silicates, rhyolites, granites, amphiboles, micas and some clays where fluoride replaces the hydroxyl radical (OH-) within of the mineral structure [9]. These compounds are abundant in some igneous and metamorphic rocks and in the sediments derived from the erosion of these rocks (Fig. 1), in the same way, fluorides can form strong complexes with aluminum, boron, beryl, iron, uranium and vanadium. pendulum is a weight suspended from a pivot so that it can swing freely.

Compound type	**Description**	**Mineral**
Halides, salts	Binary chemical compounds, made up of a fluorine atom and a lower electronegativity radical.	NaF (Villiaumite) KF CaF_2 (Fluorite)
Halides with fluoro-anions	Salts formed by several atoms of Fluorine and anions.	$Na(BF_4)$ $K(BF_4)$ $Na_3(AlF_6)$ (Cryolite)
Phosphates	Salts or esters of phosphoric acid, with a phosphorous atom surrounded by four oxygen atoms in tetrahedral form.	$Ca_5(PO_4)_3F$ (Fluorapatite)
Silicates	Union of silicon and oxygen, also called salts of silicic acid, are the most abundant minerals in the earth's crust.	$Al_2F_2(SiO_4)$ (Topaz) $KAl_2F_2(AlSi_3O_{10})$ (Muscovite) $KMg_3F_2(AlSi_3O_{10})$ (Phlogopite)
Rhyolites	Extruded igneous rock (volcanic felsic), with a chemical composition very similar to that of granite, which is why it is considered the volcanic equivalent of granite.	Quartz, potassium feldspar, oligoclase, biotite, amphibole, pyroxene.
Pimples	Intrusive igneous rock of felsic composition consisting essentially of quartz, alkali feldspar, plagioclase and mica.	

Table 1: Main compounds containing fluorine in mineral form. Source: [10] and [11].

Infiltration (percolation) is the phenomenon of when water passes through the soil and rock formations that contain minerals such as fluorite, cryolite and fluorapatite, in this process the fluoride dissolves, entering the natural underground sources of water, the above is part of the biogeochemical cycle of chemical elements present in nature.

Fluorine is the first element of the halogen family and the most reactive chemical element, the term "fluoride" refers to compounds that contain the fluorine ion (F-) such as the salt of hydrofluoric acid, instead the term "Fluorides" refers to compounds that contain fluoride, whether organic or inorganic, generally colorless.

The different fluorine compounds are mostly soluble in water and can be solid, liquid, or gas. Fluoride as such cannot be found in nature, however, fluorides are everywhere: in soil, air, water, as well as in plants and animals [12].

Fluorides are released into the environment naturally through the weathering and dissolution of minerals, emissions from volcanoes, and marine aerosols. They are also released through the combustion of coal, industrial sewage, and the wastes of many manufacturing and transformation processes, in particular steelmaking, primary production of aluminum, copper and nickel, processing of ores from phosphate, the production and use of phosphate fertilizers, the manufacture of glass, bricks, ceramics, semiconductors and the production of glue and adhesives, the transformation of oil, glass [13].

It also comes from the incineration of municipal waste such as fluorinated textiles and plastics, or decomposition of calcium difluoride, in waste sludge, due to the formation of hydrofluoric acid emissions, due to processes in the soil or in water such as fluoride ions [14].

The natural concentration of fluorides depends on the geological, chemical and physical characteristics of the aquifer, the porosity and acidity of the soil and stones, the temperature, the action of other chemical elements and the depth of the extraction wells.

In Mexico, 17 states have problems due to natural contamination of groundwater by fluorine, this contamination is of geological origin, the result of the natural interaction that groundwater has with some volcanic rocks that are widely scattered in the Sierra Madre Occidental and that constitute some of the main aquifers with which the population is supplied with water: in the states of Baja California Norte, Durango, Aguascalientes, Zacatecas and Guanajuato, groundwater contamination is located in most of the state, while in Sonora, Chihuahua , Coahuila, Nuevo León, Sinaloa, San Luis Potosí, Jalisco, Michoacán, Querétaro, State of Mexico, Hidalgo and Puebla, the presence of pollutants is observed only in some localities [15] and [16].

Likewise, according to the Mexico National Water Commission (CONAGUA, by its acronym in Spanish), most of the aquifers in the state of Jalisco are unavailable (Fig. 1).

Figure 1: Geo-hydrological condition of the aquifers of the state of Jalisco, Mexico (2018). Source: [17].

The main source for the supply of drinking water in the Highlands of Jalisco region are underground aquifers, which in many cases are of hydrothermal origin, which are characterized by the presence of potentially harmful chemical compounds for human and animal consumption [18].

The objective of this work is to relate the concentration of fluoride compounds (Fluorides) with the decrease in the dynamic levels at which drinking water is extracted from the deep wells of the city of Tepatitlan de Morelos in the state of Jalisco, Mexico.

II. MATERIALS AND METHODS

According to the Standard Methods for the Examination of Water and Wastewater [19], fluoride concentrations in water samples can be determined by different analytical methods, the most suitable for groundwater are: the ion-selective electrode and the spectrophotometric method (SPANDS) [20].

In the present investigation, the second method was used to quantify the concentration of fluorides in the groundwater samples, using the reagent that contains: Hydrochloric acid, Zirconium oxychloride and Sodium arsenite [21], and the use of the equipment brand HACH model DR 2800, which consists of a visible spectrum spectrophotometer, with a wavelength range of 340 to 900 nanometers (Fig. 2), previously calibrated according to the manufacturer's specifications with the program for the determination of fluorides with the measurement range: 0.1 to 2.5 mg / L, making dilutions (1:10, 1:20) when necessary. Standard deionized water was used both for the calibration of the equipment and for the dilutions.

Figure 2: DR 2800 spectrophotometer (HACH). Source: [22].

In addition, when taking the water samples from the wells and making the fluoride determinations, the Mexican Standard NMX-AA-077-SCFI-2001 (Analysis of water - determination of fluorides in natural, waste and treated wastewater) was taken into consideration.

Tepatitlan is a municipality located in the South Highlands region in the state of Jalisco (Fig. 3). Its regional location with respect to the state is to the center and with respect to the region to the southeast, at coordinates 20º 54 '50' 'and 21º 01' 30 " North latitude and 102º 33 '10' 'at 1021 56 '15' 'West longitude at a height of 1,800 meters above sea level.

Figure 3: Location of the study area, city of Tepatitlan, Jalisco Mexico.
Source: [23].

It is limited to the north with Yahualica de González Gallo and Valle de Guadalupe; to the south with Tototlán and Atotonilco el Alto; to the east with San Miguel el Alto and Arandas; and to the west with Acatic, Cuquío and Zapotlanejo, its territorial extension is 1532.78 square kilometers (Km²).

The climate of the municipality is semi-dry with dry winter and spring, and semi-warm with mild winter. The average annual temperature is 19 ºC, and it has an average annual rainfall of 874.7 millimeters with rainfall in the months of June, July and August. The prevailing winds are from the southeast.

The municipality has the rivers: Tepatitlán, Verde, Calderón and Los Arcos; It also has the streams: Laborcilla, Milpillas, Juanacasco, San Pablo, El Tecolote, Jesús María, Perón, Mezcala, Guayabo, La Vieja, El Jihuite and El Ocote. There are the Carretas, Jihuite, La Red, Calderón, La Vieja and El Pantano dams.

The dominant soils belong to the ferric luvisol type, eutric flatol and haplic feozem; and as associated soils are Pelic Vertisol and Molic Planesol. Most of the relief has agricultural and livestock use, with private property being the predominant form of land tenure, the region is part of the Trans-Mexican Volcanic Belt, which is a volcanic region of approximately 1000 kilometers that extends from the Gulf of Mexico to the Pacific Ocean, crossing the center of the country, a region where there are many underground thermal water deposits and where more than 40 % of the population lives [24].

The natural wealth that the municipality has is represented by more than 4,000 hectares of forest where species of white oak, pine, oak, mesquite, ash and sweet wood predominate, mainly. Its mineral resources are deposits of manganese, sand and gravel [25].

The population in the municipality of Tepatitlan, Jalisco in 2015, according to the Intercensal Survey was 141,322 people, comparing this population amount with that of 2010 (table 2), it is obtained that the municipal population increased by 3.8 % in five years, official estimates establish that by 2020 this population will increase to 153,678 inhabitants, maintaining 1.8 % of the total population of the state. The municipal seat of Tepatitlan is the most populated locality, it is estimated that by 2020 it will concentrate approximately 108,089 inhabitants [26].

Year	Population	Percentage with respect to the population of the municipality
1980	41,813	53.35
1990	54,036	58.48

1995	65,930	60.32
2000	74,262	62.30
2010	91,959	67.55

Table 2: Population growth in the city of Tepatitlan and its percentage with respect to the total population in the municipality. Source: [27].

With information provided by the potable water operating agency of the municipality of Tepatitlan (ASTEPA), field trips and geographical information, the deep wells that supply potable water to the municipal seat were located, accounting for 65 wells of which 49 operate regularly. were sampled at least twice during the months from May to October 2020.

The groundwater samples were collected in 500 ml plastic containers, previously identified, later the samples were stored at 4 ºC for transportation to the Laboratory for Water Analysis of the Los Altos University Center of the University of Guadalajara.

In Mexico, the Official Mexican Standard NOM-127-SSA1-1994, "Environmental Health. Water for human use and consumption. Permissible limits of quality and treatments to which the water must be subjected for its purification" establishes the maximum permissible limit in water for human use and consumption at 1.5 mg / L of fluorides, which coincides with that established by the World Health Organization [28].

However, since 2011, the Department of Health and Human Services (HHS), as well as the Environmental Protection Agency of the US [29], announced important measures to ensure that the norms and guidelines regarding fluorides in drinking water will continue to provide maximum protection to their citizens, so the department's proposed recommendation was the maximum limit of 0.7 mg / L of fluorides, seeking to balance the benefits of preventing dental caries and setting the limit of non-toxic effects. desired in health.

Likewise, the Agency for Toxic Substances and Disease Registry of the US [30], has calculated the minimum reference for fluoride consumption, which is 0.05 mg / kg / day for chronic oral exposure; that is, the recommended doses in the USs and Canada are 4 mg / day for men, 3 mg / day for women, and between 2 and 3 mg / day for children and adolescents [30].

III. RESULTS

The quality of groundwater extracted for human consumption in the city of Tepatitlan normally complies with the corresponding official regulations in force (NOM 127 SSA1 -1994), so the only treatment they receive consists of supplying sodium hypochlorite to 13 %, using diagrammatic dosing pumps programmed so that residual chlorine is between 0.5 and 1.5 mg / L, each deep well has a dosing system, currently there are no systems for the control of ions or compounds such as fluorides [31].

Approximately 70 % of the population of the city of Tepatitlan (75,663 inhabitants) is supplied by the network of deep wells (2020), the remaining 30 % (32,426 inhabitants), which basically correspond to the downtown area of the city, does so through the purification of surface sources (two dams). The main uses of drinking water, supplied by the operating body, both underground and surface, are domestic and commercial, including personal hygiene and food preparation.

The summary of the results of the determination of fluorides with the SPANDS method in the deep wells of the city of Tepatitlan Jalisco is presented below (table 3).

Water Well NO.	Condition operative	Name / Location	Average fluorides (mg / L)
1	Active	Front of Chevrolet	0.06
2	Intermittent	San Gabriel	0.32
3	Active	Viveros inside	0.89
4	Active	Del Monte well	1.23
5	Active	Che Campestre	**1.67**
6	Active	Aguilillas	0.13
7	Active	Jardines de Tepa	0.95
8	Active	Jardines de la Rivera	**4.30**
9	Out of service	Colonia del Carmen	-------
10	Active	Unidad deportiva (Hidalgo)	**11.44**
11	Active	Ojedas	**9.50**
12	Active	Las Cruces	**13.27**

13	Active	El Aguacate	1.21
14	Out of service	Bus station	-------
15	Active	Los Sauces de Abajo	0.41
16	Dry	Fracc. Los Sauces	-------
17	Active	Popotes	0.49
18	Dry	Prol. Betulias	-------
19	Off	Viveros cerca	------
20	Active	Hacienda Popotes	**3.86**
21	Active	New slaughterhouse	**3.12**
22	Dry	Regional hospital	--------
23	Intermittent	Lomas del Real	**7.80**
24	Active	La Villa	0.16
25	Active	Jihuite	0.35
26	Intermittent	Bosques del Lago	0.32
27	Active	Rehabilitation center (Sauces)	0.49
28	Active	La Loma	0.44
29	Out of service	Adobes	------
30	Active	San Pablo	**1.68**
31	Active	El Vivorero	1.23
32	Active	Paso de Carretas	0.50
33	Active	Fracc. Guadalupe	0.80
34	Off	Fracc. Guadalupe	------
35	Active	Aguilillas	0.15
36	Active	Sauces de arriba	0.75
37	Active	Pochote	0.52
38	Active	Buganbilias	0.35
39	Out of service	Lomas de San Miguel	-------
40	Active	La Guayabera	**9.32**
41	*I*ntermittent	Bosques de la Gloria	0.39
42	Active	Preparatoria nueva	**9.41**
43	Active	San Alfonso	0.13
44	Active	Valle Real	0.31
45	Active	San Ángel	**9.12**
46	Active	Juan Pablo II	0.27
47	Active	Santa Bárbara	0.23
48	Out of service	Mariano Jiménez	------
49	Active	Cotos del rey	0.86
50	Active	El Colonial	0.73
51	Active	Ojo de Agua de Becerra	1.12
52	Off	Prados del Roble	------
53	Active	Rinconada San Pablo	0.49
54	Active	Fracc. San Jorge	0.86
55	Dry	Loma Dorada	-------
56	Re-pumping	Constelación	-------
57	Active	Tablón II	0.96
58	Active	Los Arroyos	1.12
59	Re-pumping	Santa Teresita	-------
60	Active	La Esperanza	0.02
61	Off	Cerrito Tablón	------
62	Active	La Cuesta	0.06
63	Off	Hacienda La Cruz	------
64	Active	Royal Park	0.69
65	Active	CUAltos-UdG	**9.13**

Table 3: Fluoride content in water from deep wells in Tepatitlan Jalisco. Source: Own elaboration.

Of the 65 deep wells registered in the operating agency for the supply of drinking water for the city of Tepatitlan Jalisco, 45 are reported as active, 4 in intermittent operation, 5 shut down, 5 out of service, 4 dry and 2 for re-operation. -pumping, therefore, of the 49 regular operating wells (active and intermittent), 13 have fluoride levels that exceed the maximum permissible limits for drinking water (25.53 %), reaching values of 11.44 mg / L, the remaining 39 present values within the permissible limits in the current official regulations (maximum of 1.5 mg / L).

Table 3 contains some operational characteristics of the wells, such as the flow rate in liters per second (lps), the depth at which the water is extracted in meters (dynamic level) and the hours they work per day.

Water well NO.	**Spending (lps)**	**Hours of operation per day**	**Dynamic level (m)**
1	8	24	140
2	9	12	182
3	11	24	193
4	5	24	140
5	12	18	151
6	5	24	180
7	5	24	138
8	8	24	190
10	17	24	180
11	17	24	163
12	12	24	220
13	7	24	208
15	18	24	163
17	6	24	205
19	7	6	145
20	12	24	188
21	5	18	200
23	5	4	242
24	7	14	220
25	5	4	218
26	5	4	163
27	16	24	191
28	7	24	193
30	11	14	182
31	12	24	183
32	12	24	181
34	12	8	180
35	8	18	184
36	18	16	236
37	8	12	153
38	12	12	204
40	15	24	172
41	5	8	175
42	30	24	171
43	8	12	35
44	5	12	205
45	11	18	238
46	11	16	166
47	7	20	152
49	4	24	209
50	6	24	235
51	16	24	125
53	5	16	231
54	11	12	203
57	22	20	173

58	11	24	203
60	5	12	176
62	6	4	236
64	6	6	184
65	12	16	240

Table 4: Main characteristics of deep wells operating regularly in Tepatitlan Jalisco. Source: [31].

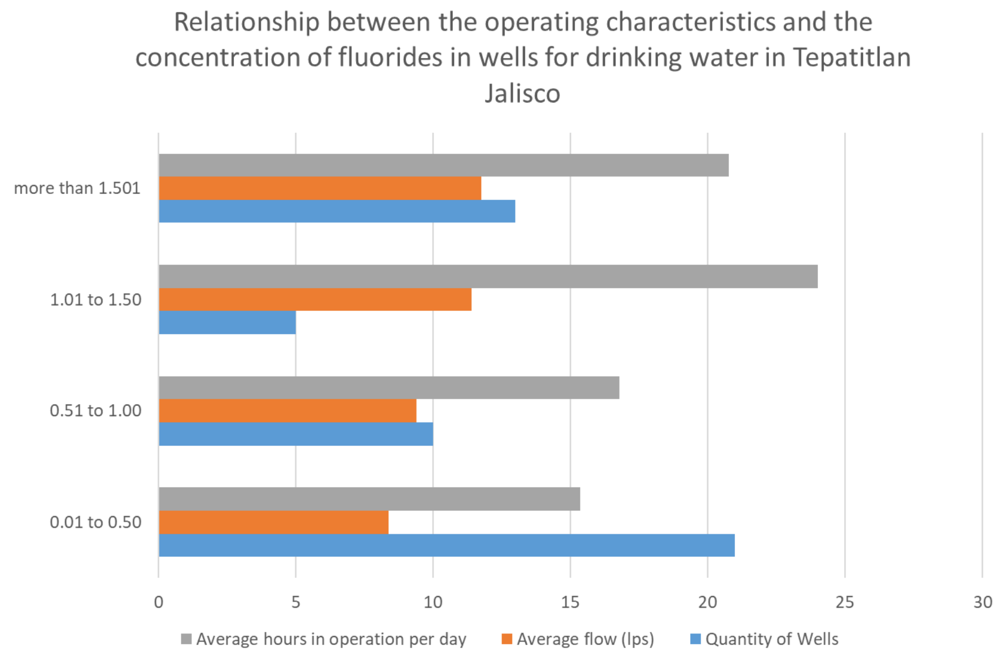
By grouping the operating wells by the range in fluoride concentration, it is possible to generate a classification with 4 strata, as well as averaging the values of the operating characteristics of the wells (table 5);

Fluoride Range (mg / L)	Quantity of Wells	Average spending (lps)	Average hours of operation per day	Average dynamic level (m)
0.01 to 0.50	21	8.38	15.37	178.33
0.51 to 1.00	10	9.40	16.80	193.61
1.00 to 1.50	5	11.39	24.00	171.80
More than 1.50	13	11.75	20.77	195.15

Table 5: Comparison between fluoride content and operating characteristics of deep wells in Tepatitlan Jalisco. Source: [31].

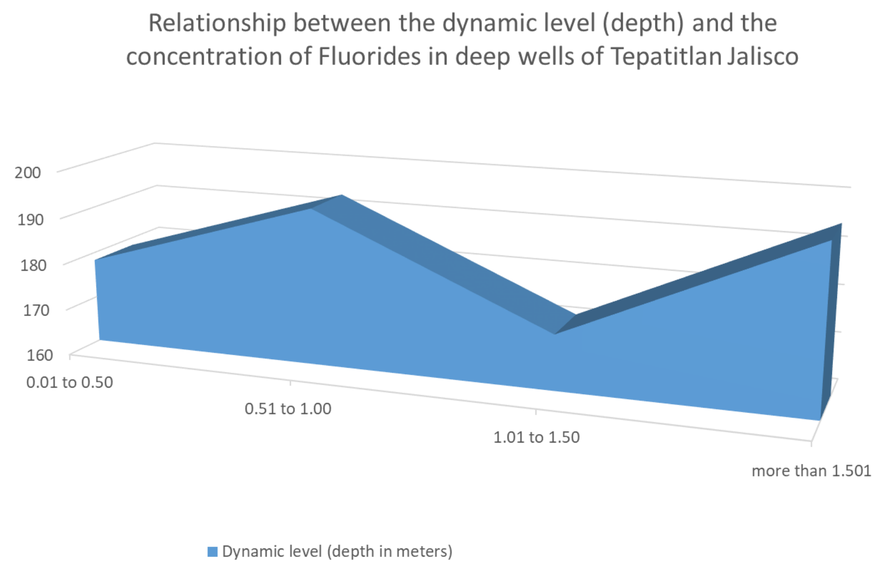
The following correlations can be observed:

1) The group of wells with the highest concentration of fluorides are those that, on average, handle the highest flow and those that operate the highest number of hours per day on average (graph 1).



Graph 1: Relationship between the operating characteristics and the concentration of fluorides in wells for drinking water in Tepatitlan Jalisco. Source: Own construction.

2) Regarding the average dynamic level at which the water is extracted, it should be noted that the strata vary; however, the wells with the highest concentration of fluorides are those with the greatest average depth (graph 2).



Graph 2: Relationship between the dynamic level (depth) and the concentration of Fluorides in deep wells of Tepatitlan Jalisco. Own construction.

Another important observation is that the highest concentrations of fluorides are located in wells located mainly to the West, Northwest and Southeast of the city center (Figure 5), so that the intake of water from these areas represents a greater risk for people's health, compared to the rest of the study region.

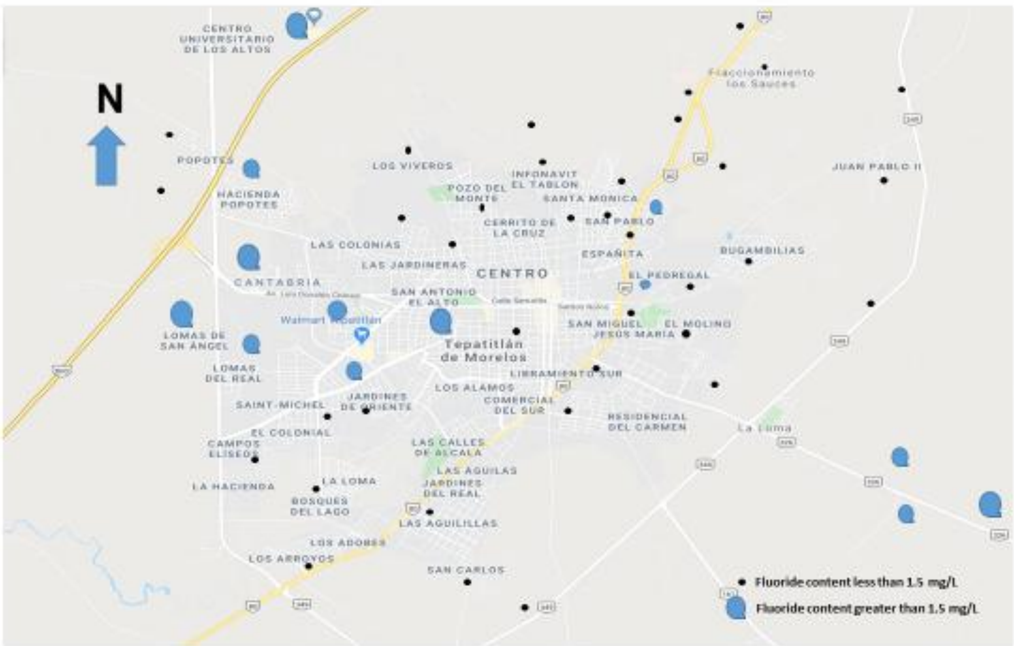


Figure 5: Location of active wells in the city of Tepatitlan Jalisco. Source: Own construction.

IV. DISCUSSION

Hydrodynamics in groundwater mainly depends on the pumping time of deep wells (operating hours per day), necessarily causing a continuous decrease in pressure at the piezometric surface, which induces the rise of deeper water flows, which they have higher concentrations of compounds such as fluorides in reference to the colder and shallower flow systems [3] and [32].

On the other hand, in chemical weathering (chemical decomposition), which is the alteration of rocky materials exposed to air, humidity and the effect of organic matter that alter it, oxidation-reduction reactions, hydration, hydrolysis develop together, and dissolution, according to Buol and collaborators [33], the most important process of chemical weathering is hydrolysis, which causes the complete disintegration or chemical modification of the primary weatherable minerals.

Likewise, during the water-rock interaction, the H + ion that is exchanged comes from the dissociation of carbonic acid that is formed during the natural recharge process of groundwater, and induces that bicarbonate is in direct relation with the consumption of ions. H + during cation exchange [34].

The H + helps in the attack of the feldspar that transforms it into clay minerals, which is exemplified through the dissolution of albite, in whose reaction it consumes H + ions, liberates HCO3- and Na + radicals, consumes CO2- aqueous radicals, increasing the pH [13]. As part of the biogeochemical cycle of chemical elements present in nature, when water passes through rocks and soil with minerals such as fluorite, cryolite, and fluorapatite, fluoride dissolves and enters natural water sources by infiltration and percolation.

In 2002, Hurtado and Gardea determined that for the city of Tepatitlan, with a population of 74,262 inhabitants, the average of fluorides in the first 24 active wells that supplied drinking water was 2.4 mg / L, with a deviation standard of ± 3.8, with a range of 0.1 to 14.4 mg / L, in the present study of the same wells, 5 are depressed (20.83 %) and the average of fluorides in the remaining 19, is 3.101 mg / L, observing an increase of 29.21 % in practically 8 years [35].

The overexploitation of natural resources such as aquifers is causing the contamination of the water, which the majority of the population of our country uses for their use and consumption; This contamination not only corresponds to natural minerals such as fluorides, but also to other elements and harmful chemical compounds, therefore it is of great importance to implement an adequate management of water resources, ensuring the adequate control and distribution of quality water, as well as a active participation of authorities, researchers and society to contribute to the maintenance of this vital resource [36] and [37].

V. CONCLUSION

The increase in the concentration of compounds such as fluorides in the extracted groundwater can be an indicator of dynamic abatement (sensitive decrease in the water level) of deep wells used as sources of drinking water supply, it should be noted that currently the network of deep wells for the supply of drinking water for the city of Tepatitlan; 4 wells are dry, 5 have practically collapsed and another 2 are used only for the re-pumping of water, that is, 16.92 % of the wells in the network do not operate due to a radical decrease in dynamic levels.

Likewise, 25.53 % of the wells present concentrations of fluorides greater than the maximum permissible limit in the current official regulations, so it is advisable to take special precaution with these wells, implementing systems to reduce these values (precipitation, filtration, among others), including mixing with waters that contain lower values from other sources (controlled dilutions). In the same way, it is possible to carry out tests by extracting water at different flow rates (exerting different hydraulic gradients in the well), to obtain the necessary water with the appropriate amount of fluorides, in order to achieve control of this compound, since it includes the underground hydrodynamics of the aquifer. it is possible to properly regulate this important parameter.

Considering the results obtained in the present work, some possible causes of the high concentration of fluorides in deep wells in the city of Tepatitlan, lies in the number of wells operating, the flows that are extracted, as well as the hours that operate daily, various up to 24 hours a day, which causes the over-exploitation of the aquifer, evidenced by the current average dynamic extraction levels of 184.72 meters, with wells operating at 240 meters. Assuming that the volumes of extraction are greater than the volumes of the natural recharge of the aquifer, the waters that are being extracted are increasingly deeper, which puts the sustainability of the aquifer at risk.

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