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Articles

Use of environmental isotopes of water and geochemistry to determine nitrate sources in south of Cuernavaca aquifer

Uso de isótopos ambientales del agua y geoquímica para determinar fuentes de nitrato en el sur del acuífero Cuernavaca

Luis González-Hita[1]

Miguel A. Mejía-González[2], ORCID: https://orcid.org/0000-0001-8921-1100

Vicente Ortega-Lara[3]

Blanca Carteño-Martinez[4]

Juan C. Aparicio-González[5]

Dustin S. Mañon-Flores[6]

[1]Instituto Mexicano de Tecnología del Agua, Hydrology Coordination, Groundwater Hydrology Subcoordination, Jiutepec, Morelos, Mexico, lghita@tlaloc.imta.mx

[2]Instituto Mexicano de Tecnología del Agua, Hydrology Coordination, Groundwater Hydrology Subcoordination, Jiutepec, Morelos, Mexico, mamejia@tlaloc.imta.mx

[3] Independent researcher, México, vortegalara@gmail.com

[4]Instituto Mexicano de Tecnología del Agua, Hydrology Coordination, Groundwater Hydrology Subcoordination, Jiutepec, Morelos, Mexico, blancartmart@hotmail.com

[5]Instituto Mexicano de Tecnología del Agua, Hydrology Coordination, Groundwater Hydrology Subcoordination, Jiutepec, Morelos, Mexico, jcapariciog@gmail.com

[6]Instituto Mexicano de Tecnología del Agua, Hydrology Coordination, Groundwater Hydrology Subcoordination, Jiutepec, Morelos, Mexico, pistolpiolo@gmail.com

Corresponding author: Miguel A. Mejía-González, mamejia@tlaloc.imta.mx

Abstract

The Cuernavaca aquifer is the main source of permanent fresh water that supports economic development and supply to the population of the capital of the state of Morelos. During the last thirty years in the south of the Cuernavaca aquifer nitrates have increased degrading groundwater quality. The identification of the origin of the nitrate is essential to implement public policies that minimize the contamination. Taking into account the characteristics of the region such as soil type and geology, population growth, economic activities such as agriculture, and recreational activities including golf, four possible sources of nitrate are proposed in the study area: 1) Natural sources (soil organic nitrogen), 2) infiltration of wastewater, 3) infiltration of agricultural water, and 4) infiltration of water used in the irrigation of golf courses. In the southern portion of the Cuernavaca aquifer, the geochemistry and environmental isotopes of water ($\delta^{18}O$, $\delta^{2}H$, $\delta^{3}H$) indicate that the main nitrate contaminant source is sewage disposal. The data also indicate that the arsenic detected in the wells comes from the deepest strata of the aquifer. The results also show correlation between nitrate and uranium concentration. The data obtained allowed to determine the meteoric water line of the Cuernavaca Aquifer.

Keywords: Cuernavaca aquifer, nitrate, sewage water, geochemistry, environmental isotopes of water, arsenic, uranium, meteoric water line of Cuernavaca aquifer.

Resumen

El acuífero Cuernavaca es la principal fuente de agua dulce permanente que sustenta el desarrollo económico y el suministro a la población de la capital del estado de Morelos. En los últimos 30 años, los nitratos en el sur del acuífero Cuernavaca se han incrementado, degradando la calidad del agua subterránea. La identificación del origen del nitrato es fundamental para implementar políticas públicas que controlen y reviertan dicha contaminación. Con base en las características propias de la región, como son tipo de suelo y geología, incremento acelerado de la población, y actividades industriales, agrícolas y recreativas, entre ellas la práctica del golf, se plantean cuatro posibles fuentes de nitrato en la zona de estudio: 1) nitrato del suelo, 2) infiltración de aguas residuales, 3) fertilizantes provenientes de zonas agrícolas, y 4) fertilizantes provenientes de campos de golf. En la porción sur del acuífero Cuernavaca, los datos geoquímicos y los isótopos ambientales del agua ($\delta^{18}O$, $\delta^{2}H$, $\delta^{3}H$) indican que la contaminación por nitrato proviene principalmente de aguas residuales. Asimismo, indican que el flujo profundo aporta arsénico a algunos aprovechamientos subterráneos. Los resultados también muestran correlación entre las concentraciones del nitrato y del uranio en el acuífero. El registro periódico de los datos isotópicos permitió también determinar la línea de agua meteórica de la zona del acuífero Cuernavaca.

Palabras clave: acuífero Cuernavaca, nitrato, aguas residuales, geoquímica, isótopos ambientales del agua, arsénico, uranio, línea de agua meteórica del acuífero Cuernavaca.

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Introduction

Nitrate (NO_3^-) is one of the main causes of groundwater contamination (Zhao, 2015; Canter, 1997). Various sources of nitrate contribute to this contamination, including agricultural and livestock activities, and wastewater (Haller, McCarthy, O'Brien, Riehle, & Stuhldreher, 2013; Viers, Liptzin, Rosenstock, Jensen, & Hollander, 2012).

The consumption of drinking water with high concentrations of nitrate is a risk factor for several types of cancer, including gastric, colorectal and brain tumors (CDPH, 2013). The World Health Organization (WHO) established a maximum permissible limit of 50 mg/l for nitrates (WHO, 2007) to prevent damage to the health of the population. For Mexico, the permissible limit (Modification to the Official Mexican Standard NOM-127-SSA1-1994) (SSA, 1994) for drinking water is 10 mg/l of nitrogen (as $N\text{-}NO_3^-$), which is equivalent to 44.3 mg/l of nitrate.

Aquifers contaminated with nitrates have been reported in Mexico, both from agricultural activities and wastewater (Pacheco & Cabrera, 1997; Cardona, 2004; Torres-Martínez *et al*., 2021). In the study carried out by Suarez-Barragán and Ureno-Luna (1988) about the quality of groundwater in the Cuernavaca Valley, the presence of nitrates was detected in wells in some municipalities; among them, the Tetecalita well in Emiliano Zapata with 6.2 mg/l, a spring in Temixco with 4.43 mg/l, a spring in Cuernavaca with 6.64 mg/l, the Melchor Ocampo well with 5.75 mg/l and the Hotel Casino de la Selva well with 5.85 mg/l, both in downtown Cuernavaca. By 1995 (Conagua, 2002), nitrate concentrations in the western portion of the Cuernavaca aquifer (municipalities of Temixco and Acatlipa), already reached values of 20 mg/l (Figure 1).

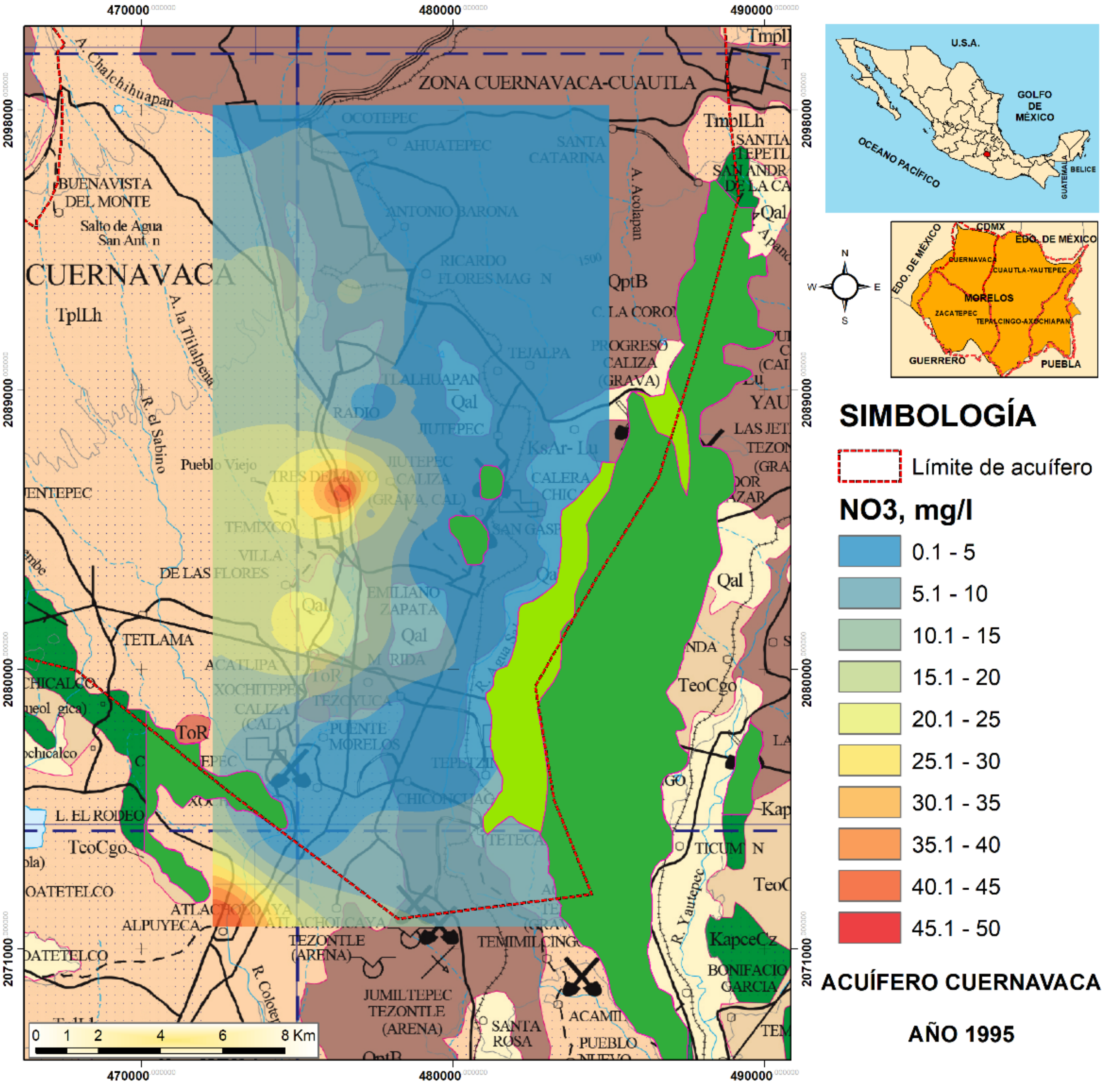

Figure 1. Nitrate concentrations in 1995 in the Cuernavaca Aquifer.

About 89 Mm^3 of wastewater is generated annually in Morelos, of which 66 % is discharged without treatment to septic tanks, absorption wells, or directly into streams, ravines and rivers (Conagua, 2010). Discharges are mainly public-urban with 46 % and services with 21 %. The municipal wastewater flow is approximately 4 585 l/s, of which 2 820 l/s are collected and 959 l/s are treated (Conagua, 2010).

58 % of Morelos' surface area is used for agricultural and pasture production. The entity has 204,441 hectares dedicated to the agricultural sector (INEGI, 1991). In the study area, the productive crop areas are located in Jiutepec, in the ejidos of Acapantzingo, Chapultepec and Atlacomulco; to the northeast, in the limits of the municipalities of Cuernavaca and Tepoztlán; to the south, in the ejidos of Temixco, Acatlipa, Zapata and Tezoyuca (POZCI, 2009). The main crops grown in this region are tomatoes, green tomatoes, corn, rice and ornamental horticulture (Sagarpa, 2003). Pesticides, herbicides and fertilizers used in agricultural areas, when they infiltrate into the subsoil, can contaminate groundwater (Conagua, 2010).

Four golf courses are located in the study area. Some of the nutrients applied to the grass are (Del-Campo-Delgado, 2016): a) Nitrogen, an essential element for grass growth and giving green color; b) phosphorus, related to root development, and c) potassium, which acts at many levels of plant metabolism. When water and nutrients are applied in excess, fertilizers infiltrate in the form of leachates.

Nitrates in the southern Cuernavaca aquifer have increased over the last 30 years, degrading groundwater quality. The identification of the origin of nitrate is fundamental to implement public policies to control and revert such contamination.

The objective of this study was to determine nitrate concentrations in the southern portion of the Cuernavaca aquifer, and identify the sources that produce nitrate. To do so, hydrogeological tools, groundwater chemistry, and environmental water isotopes ($\delta^{18}O$, $\delta^{2}H$, $\delta^{3}H$) were used.

Study area

The Cuernavaca aquifer is located in the physiographic province of the Mexican Volcanic Belt, subprovince Balsas-Mexcala Basin (Raisz, 1964), between parallels 18º 40' and 19º 10' North latitude, and 99° 00' and 99° 20' West longitude (Figure 2).

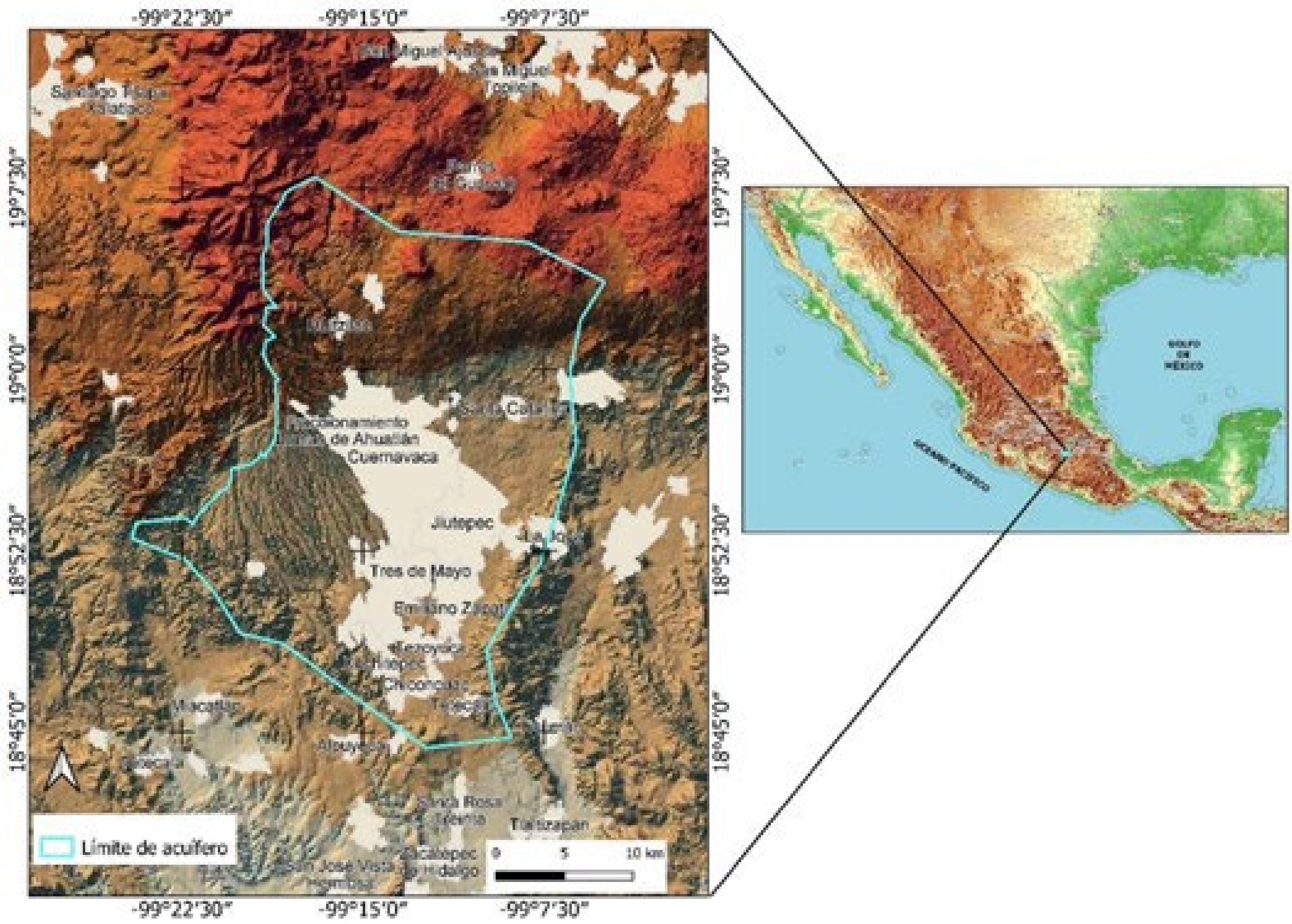

Figure 2. Location of Cuernavaca aquifer.

The geomorphological limits of the Cuernavaca aquifer are: to the north the Zempoala, Ajusco and Chichinautzin mountains ranges, to the south the Xochicalco mountain range, to the west the extension of the Zempoala mountain range and to the east the Tepoztlán-Tlaltizapán mountain range (Conagua, 2020). The Zempoala (3 650 masl) and Chichinautzin (3 690 masl) mountain ranges are the highest elevations (Conagua, 2020).

According to mean annual temperatures, the Cuernavaca aquifer zone has four types of climates (Conagua, 2002): Semi-cold (5-12 °C), temperate (12-18 °C), semi-warm (18-22 °C) and warm (> 22 °C). The semi-cold and temperate climates are located in the northern portion, while the semi-warm and warm climates are located in the central and southern portions, respectively.

The orography and relief of the terrain (Conagua 2002) influence rainfall that affects the Cuernavaca aquifer. In the upper portion of the region, precipitation is in the order of 1 500 to 1 100 mm per year, while in the southern portion it is in the order of 950 to 850 mm per year. In general, precipitation is more abundant from June to October.

In the northern portion, various types of temperate and cold climate forests predominate, including: Pine forest, oyamel forest, cedar-tascate forest, oak forest, pine-oak forest and mountain mesophyll forest (Secretaría de Desarrollo Sustentable de Morelos, 2009). In the central and southern portions, low deciduous forest, low subtropical scrub and pasture predominate (Secretaría de Desarrollo Sustentable de Morelos, 2009). Agricultural crops are also a type of non-natural vegetation present in the plain zone.

The stratigraphic column of the Cuernavaca aquifer is composed of thirteen types of rock units, which range from Lower Cretaceous to recent (Conagua, 2020). These units are: Xochicalco Formation, Morelos Formation, Cuautla Formation, Mexcala Formation, Balsas Group, Tilzapotla Rhyolite, Colotepec Granite, Tepoztlan Formation, Zempoala Andesite, Cuernavaca Formation, Chichinautzin Formation, Continental Clastic Deposits and Quaternary Alluvial Deposits.

Hydrogeology

Based on geomorphological and geohydrological aspects, it is possible to identify three hydrogeological units in the Cuernavaca aquifer (Figure 3) (Conagua, 2002; Morales-Casique, Guinzberg-Belmont, & Ortega-Guerrero, 2016; Ortega, Garcia, & Pelayo, 2003).

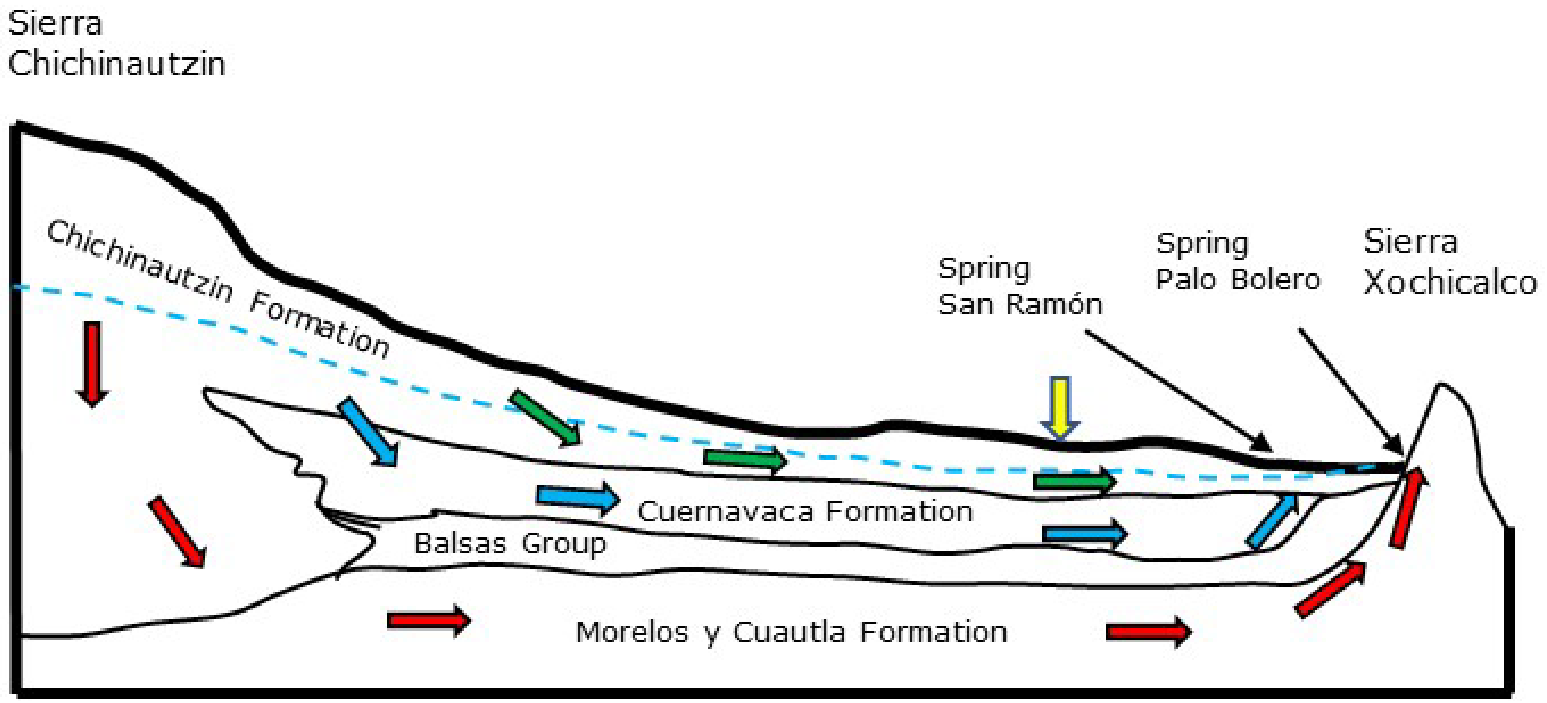

Figure 3. Hydrogeological units of the Cuernavaca aquifer.

The first unit corresponds to a free aquifer hosted in fractured basaltic igneous rocks of the Chichinautzin formation, and continental clastic deposits of the Cuernavaca formation. The second unit, defined as an aquitard, is associated with low permeability materials of the Balsas Group and the Mexcala Formation. It functions as the basement of the free aquifer, and is located at average depths of 100 to 200m in the middle and southern portion of the aquifer. The third unit is a semi-confined regional aquifer, hosted in the limestones of the Morelos and Cuautla formations, and is the basis of the geohydrologic sequence of the Cuernavaca aquifer.

The preferential recharge zones are located in the upper parts of the study area. Sierra Zempoala in the northwest, made up of fractured volcanic rocks from the Tertiary, and Sierra Chichinautzin in the north, made up of fractured and cavernous basalts from the Quaternary (Jaimes-Palomera *et al*., 1989).

Anthropogenic sources of pollution

The main streams and ravines that drain wastewater from the city of Cuernavaca, in a north-south direction, are El Pollo ravine in the western portion, and Barranca Analco in the central and eastern portions of the municipality of Cuernavaca, which flow north of the municipal capital of Temixco into the Apatlaco River. The northeastern part of Cuernavaca discharges into the Puente Blanco ravine, which collects wastewater from Jiutepec and Emiliano Zapata, where it is called Arroyo Dulce, and finally flows into the Apatlaco River south of the town of Xochitepec. In the easternmost portion of the valley drains the Las Fuentes ravine, into which the Huauchiles and Las Fuentes springs discharge, as well as wastewater from Tejalpa, Progreso, Las Fuentes and El Pochotal, finally reaching the agricultural areas of southern Jiutepec and Emiliano Zapata (Figure 4) (IMTA, 2012).

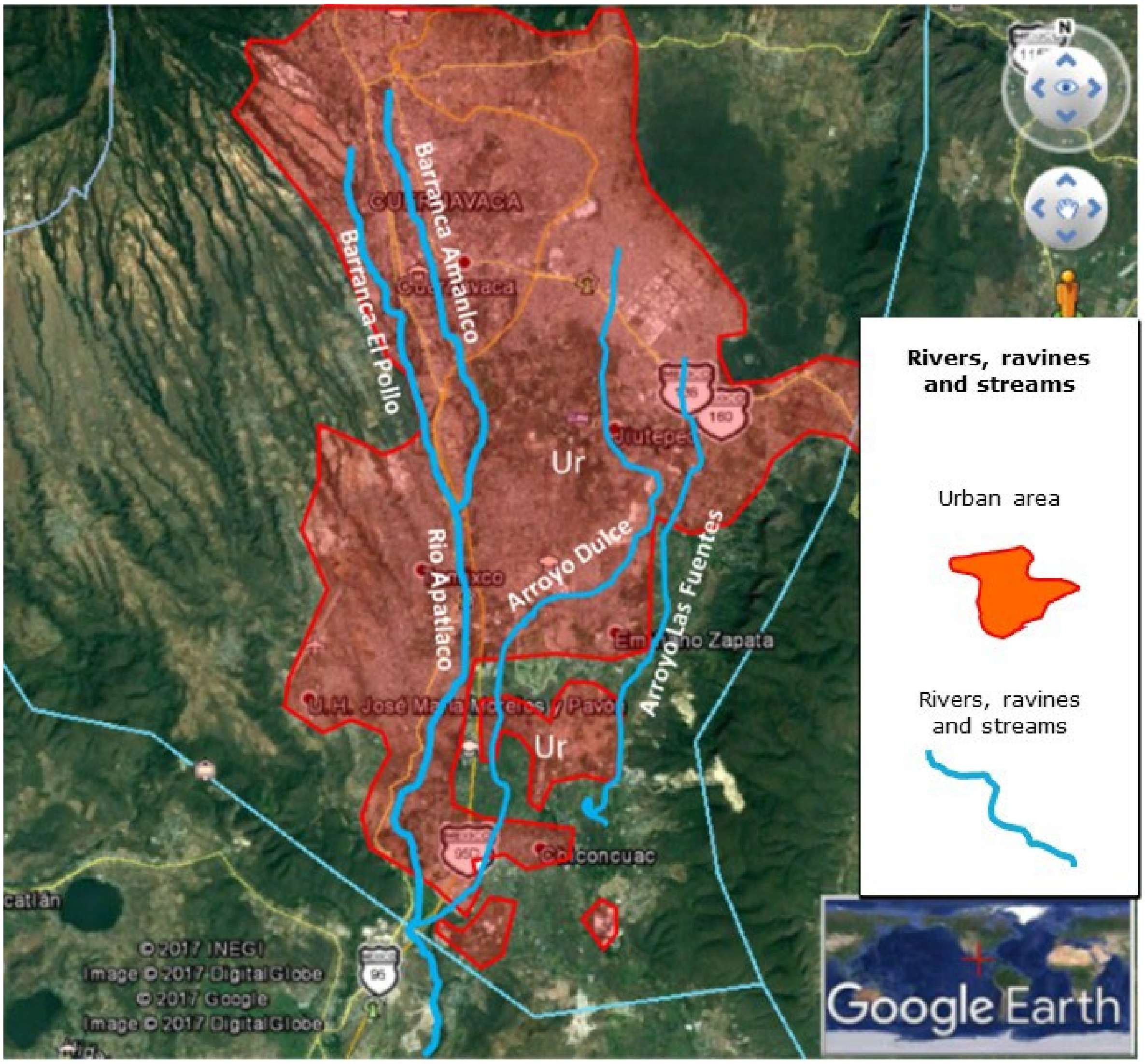

Figure 4. Main rivers, streams and ravines in the Cuernavaca aquifer.

Methodology

Rainfall

Rainwater samples were collected from 2016 to 2022, using RS2 PALMEX rainwater collectors, which allow collecting accumulated samples without evaporation or isotopic fractionation (IAEA, 2002). Samples were filtered using cellulose membranes with 0.45 µm pore size, collected in high-density polyethylene bottles with conical and polyseal insert caps, and preserved at 4 °C.

The collectors are located in the following towns: Tezoyuca, Temixco, Cuernavaca, Huitzilac, Tres Marías and Tres Cumbres. These collectors are part of the National Network of Isotopes in Precipitation (Known as RENIP), which covers the 15 physiographic provinces that make up the national territory. The RENIP is operated by the department of Hydrology of the Mexican Institute of Water Technology (IMTA).

Stable isotope analyses were conducted at the isotope hydrology laboratory of IMTA using laser water isotope analyzers Picarro L2110-i and Los Gatos Research LWIA-45EP. The analytical precision of the measurements was ± 0.5‰ (Vs. VSMOW) for $\delta^{2}H$ and ± 0.13‰ (Vs. VSMOW) for $\delta^{18}O$.

Groundwater

Water samples were collected from 16 groundwater developments during the months of January and February 2014 (Figure 5).

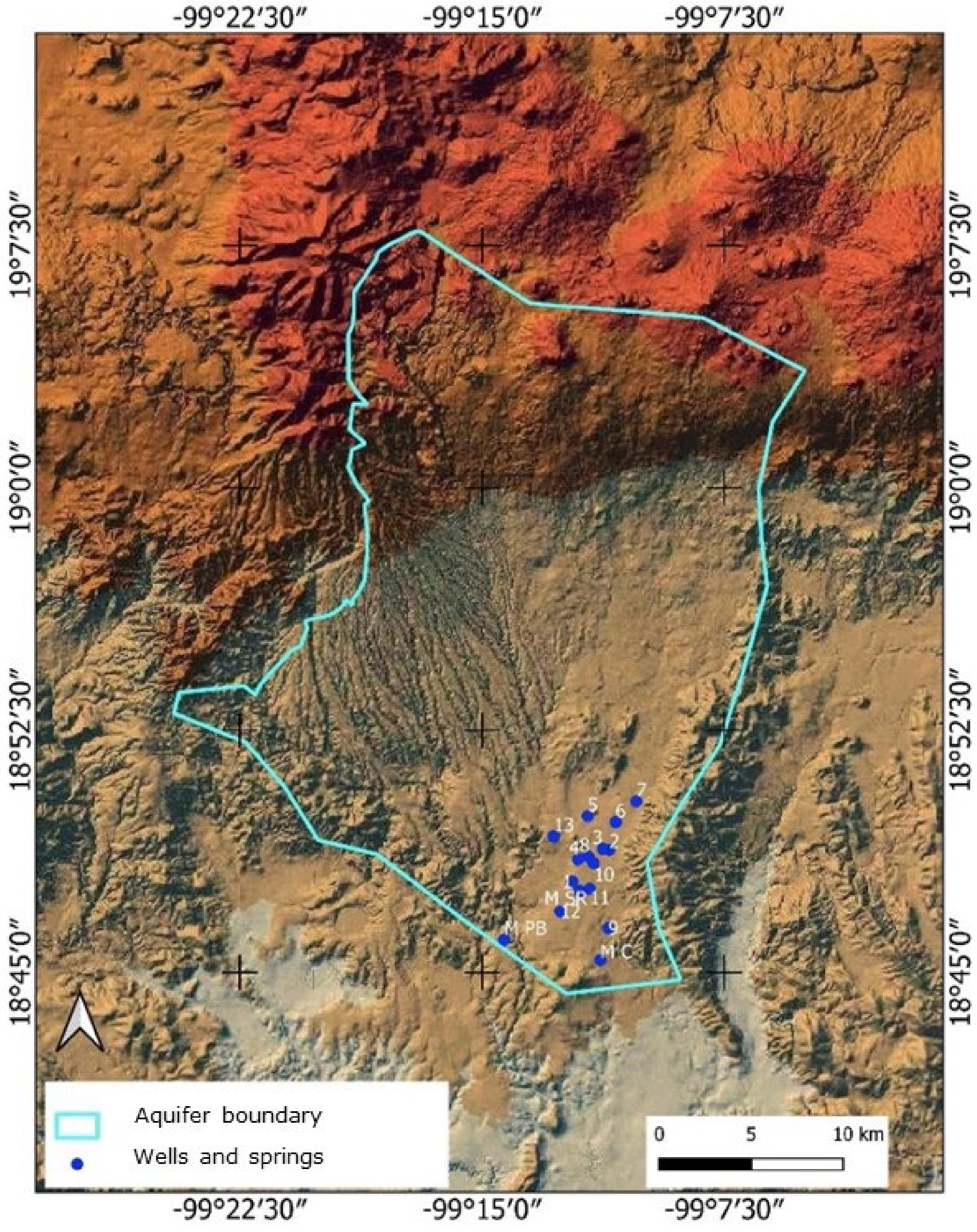

Figure 5. Location of the groundwater developments.

Samples were taken at the discharge train of the wells. Prior to measuring field parameters and collecting samples, the entire volume of water in the well annulus was renewed by pumped extraction, in order to sample and measure representative water from the aquifer.

Samples were filtered using cellulose membranes with 0.45 μm pore size and collected in high-density polyethylene bottles with conical and polyseal insert caps.

Samples for anions and stable isotopes were preserved at 4 °C. Samples for metals and cations were acidified to pH < 2 with ultrahigh purity nitric acid.

In order to reduce as much as possible the modification of the prevailing physicochemical properties of the water in the aquifer, isolation cells were used for the measurement of field parameters. These devices are connected with hoses to valves located in the discharge train of the well. This eliminates the contact of the water with the atmosphere, avoiding modifications due to the release of gases occluded in the water, such as carbon dioxide and/or hydrogen sulfide.

Field parameters

The parameters measured in the field were: hydrogen potential (pH), electrical conductivity (EC), dissolved oxygen (DO), redox potential (Eh), water temperature and ambient temperature (T). The methodologies used are:

1. The measurement of hydrogen ion activity in groundwater was carried out by means of a potentiometer. The calibration of the equipment was performed using the three-point calibration technique, from buffer solutions of known pH. In this technique, the pH to be recorded must be located between these extreme values. Solutions of 4.0, 7.0 and 10.0 pH units were used in the calibration of the equipment.
2. Electrical conductivity was measured by means of a portable conductivity meter with temperature compensation. Calibration of the equipment was performed using a standard potassium chloride (KCl) solution of 1 412 µS/cm at 25 °C.
3. Measurement of dissolved oxygen in groundwater was carried out using a temperature-compensated, coated galvanic membrane sensor electrode. The dissolved oxygen electrode was calibrated according to the atmospheric conditions at the site, i.e. in equilibrium with the air at site elevation.
4. The redox potential of groundwater was measured with a combined type electrode, consisting of a platinum filament and a silver-silver chloride reference electrode with 3 mol/l potassium chloride reference electrolyte. The sensitive element where the electron transfer takes place is the platinum filament, so it was kept clean and polished to avoid erroneous measurements. A standard Zobell's solution, Eh of 428 mV (a mixture of potassium ferrocyanide and ferricyanide) was used to verify the operation of the electrode and adjust the field measurement.
5. Ambient and water temperature were measured with electrodes previously calibrated in the laboratory and verified in the field.

Parameters measured in the laboratory

The measurement of dissolved chemical constituents in the groundwater samples was carried out by ion chromatography for anions and by two instrumental techniques for cations, depending on their concentrations: plasma-induction coupled with optical emission spectroscopy (ICP/OES) and plasma-induction coupled with mass spectrometry (ICP/MS). The ICP/MS technique is approximately 1,000 times more sensitive than the ICP/OES technique. The analyses were performed by Activation Laboratories Ltd of Canada (ACTLABS). For analytical quality control of NO3 (as N), the laboratory used a certified reference standard of 2.87 mg/l. In the control tests applied to the measurements, the analytical error was less than 5 %.

Results

Rainfall

The arithmetic average and precipitation-weighted average values of the $\delta^{18}O$ (‰) and $\delta^{2}H$ (‰) are shown in Table 1. Precipitation-averaged values (δ_P) were calculated as $\delta_P = \Sigma P_i * \delta_i / P_T$, where P_i is the amount of rainfall, δ_i is the isotopic composition of the sample (δ2H or δ18O), and PT is the amount of total precipitation.

Table 1. Values of the arithmetic average and precipitation-weighted average values of the $\delta^{18}O$ (‰) and $\delta^{2}H$ (‰).

Station	**Latitude N**	**Longitude O**	**Altitude (masl)**	**$\delta^{18}O$ (‰)**	**$\delta^{2}H$ (‰)**	**$\delta^{18}O_P$ (‰)**	**$\delta^{2}H_P$ (‰)**
Tres Cumbres	19.061267	-99.218317	3290	-10.84	-70.46	-12.18	-81.22
Huitzilac	19.032850	-99.268800	2600	-9.43	-60.99	-10.71	-71.38
Tres Marías	19.051483	-99.246283	2813	-9.31	-59.69	-10.4	-68.91
Cuernavaca	18.942833	-99.215139	1638	-8.6	-57.83	-9.99	-68.12
Temixco	18.857528	-99.223917	1264	-7.45	-49.85	-8.95	-61.11
Tezoyuca	18.800383	-99.196733	1194	-7.43	-50.41	-8.74	-60.05

Figure 6 presents the precipitation-weighted average of the $\delta^{18}O$. The most enriched values in heavy isotopes correspond to the site with lower altitude and warm temperature, while the most depleted values correspond to the area with higher altitude and cold temperature.

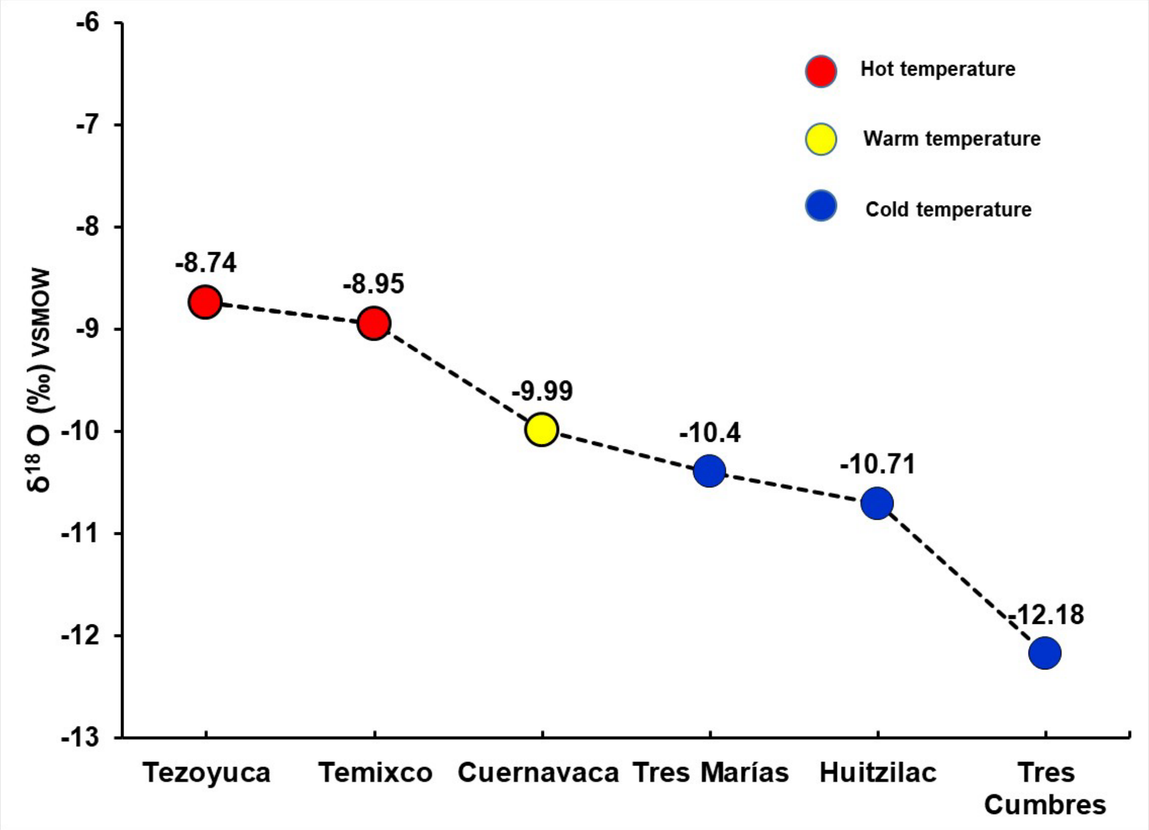

Figure 6. Average values of the $\delta^{18}O$ in the collectors.

With the isotopic data of the samples, plots of the ratio $\delta^{18}O$ ‰ (Vs. VSMOW) versus $\delta^{2}H$ ‰ (Vs. VSMOW) were elaborated. Figure 7 shows the data for the collectors located in the Chichinautzin mountain range, and Figure 8 shows the data for the valley. Using a least-squares linear regression of the isotope data, the Meteoric Water Lines were determined which are: $\delta^{2}H = 8.15\ \delta^{18}O + 17.86$ (Tres Cumbres); $\delta^{2}H = 8.15\ \delta^{18}O + 16.24$ (Tres Marías); $\delta^{2}H = 8\ \delta^{18}O + 14.58$ (Huitzilac); $\delta^{2}H = 7.7\ \delta^{18}O + 8.14$ (Cuernavaca); $\delta^{2}H = 7.53\ \delta^{18}O + 6.28$ (Temixco); $\delta^{2}H = 7.42\ \delta^{18}O + 4.7$ (Tezoyuca).

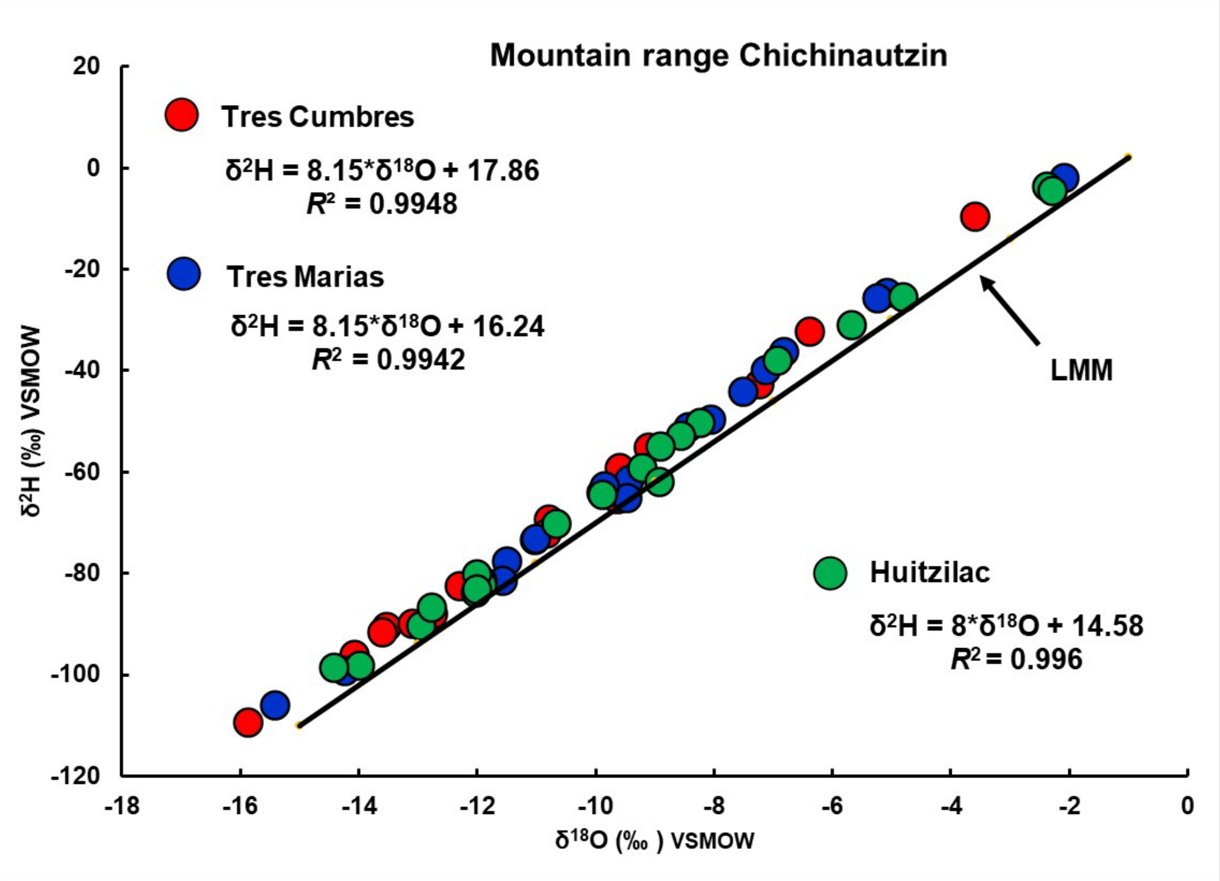

Figure 7. Meteoric water lines of the collectors in the Chichinautzin mountain range (Tres Cumbres, Tres Marías and Huitzilac).

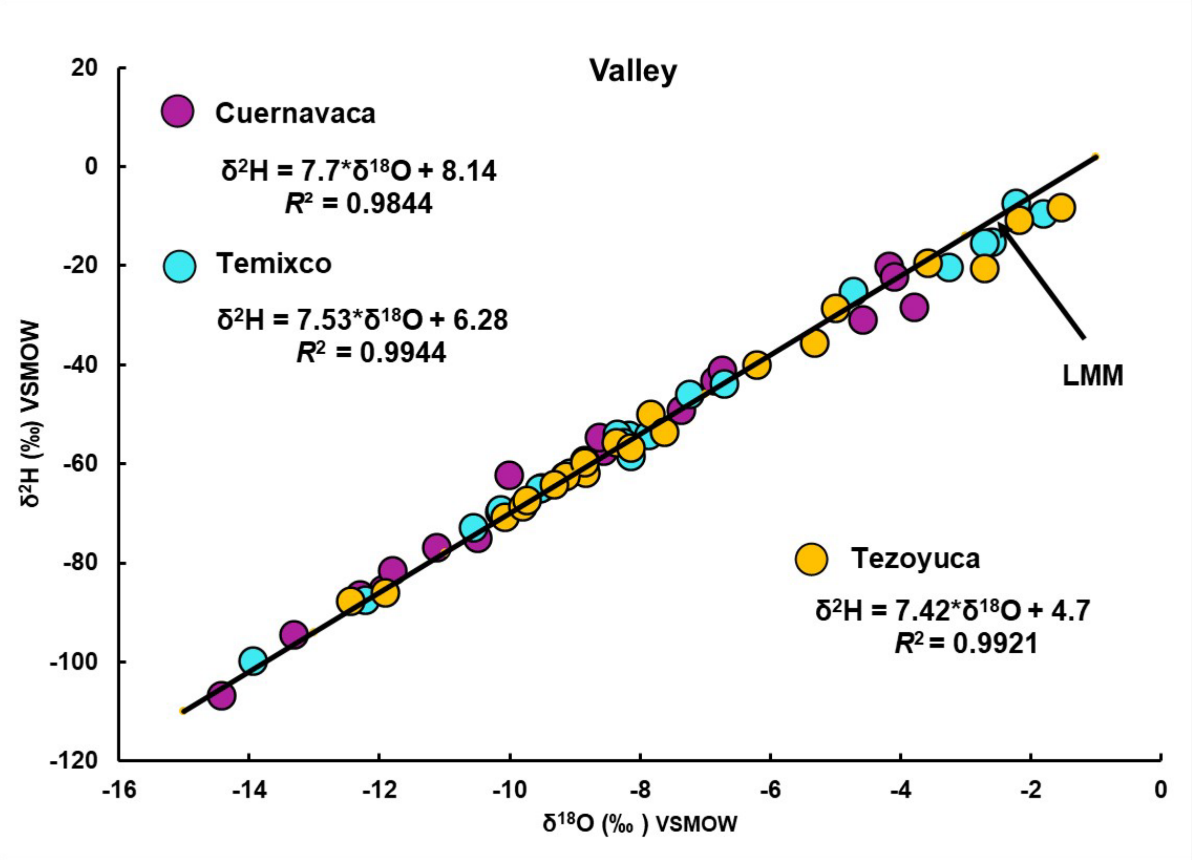

Figure 8. Meteoric water lines of the collectors in the valley (Cuernavaca, Temixco and Tezoyuca).

Taking into account the data from all collectors (Figure 9), the equation of the Meteoric Water Line of the Cuernavaca Aquifer zone is: $\delta^2H = 7.8\ \delta^{18}O + 8.82$, $R^2 = 0.9841$, $n = 118$.

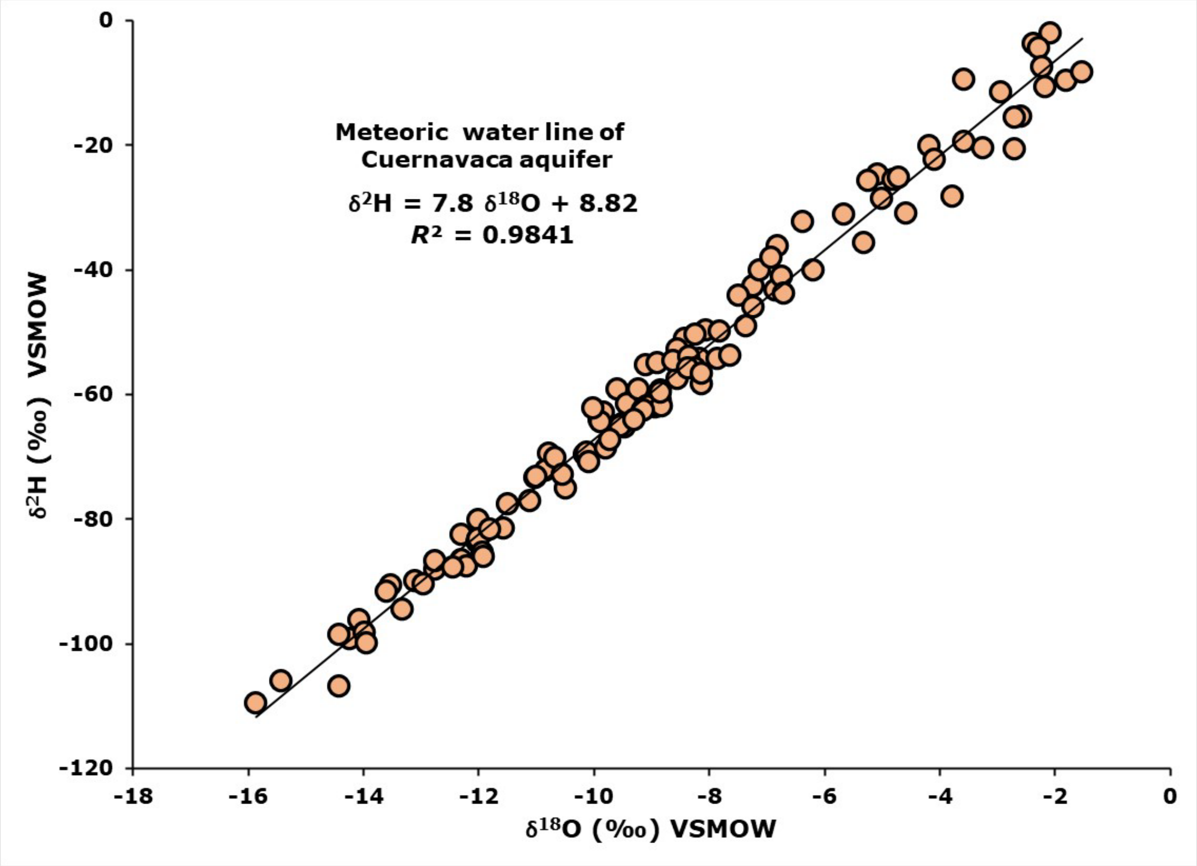

Figure 9. Meteoric Water Line of the Cuernavaca Aquifer.

When evaporation takes place in environments with relative humidity less than 100 %, the fractionation of ^{18}O exceeds that of ^{2}H, producing vapor with high deuterium content (excess deuterium) relative to ^{18}O (Martinelli, Victoria, Sternberg, Ribeiro, & Moreira, 1996; Gat, 2000). This effect, defined as $d= \delta^{2}H-8*\delta^{18}O$, produces a deuterium excess (d) of 10‰ at the Global Meteoric Water Line (GMWL) at relative humidity of 85 % (Merlivat & Jouzel, 1979; Clark & Fritz, 1997).

The Meteoric Water Lines of the mountain range collectors (>2600 masl) have intercept values (+17.86 ‰, +16.24 ‰, +14.58 ‰) greater than the GMWL value (+10 ‰). The deuterium excess (d) values of these collectors are also greater than those of the GMWL (Figure 10).

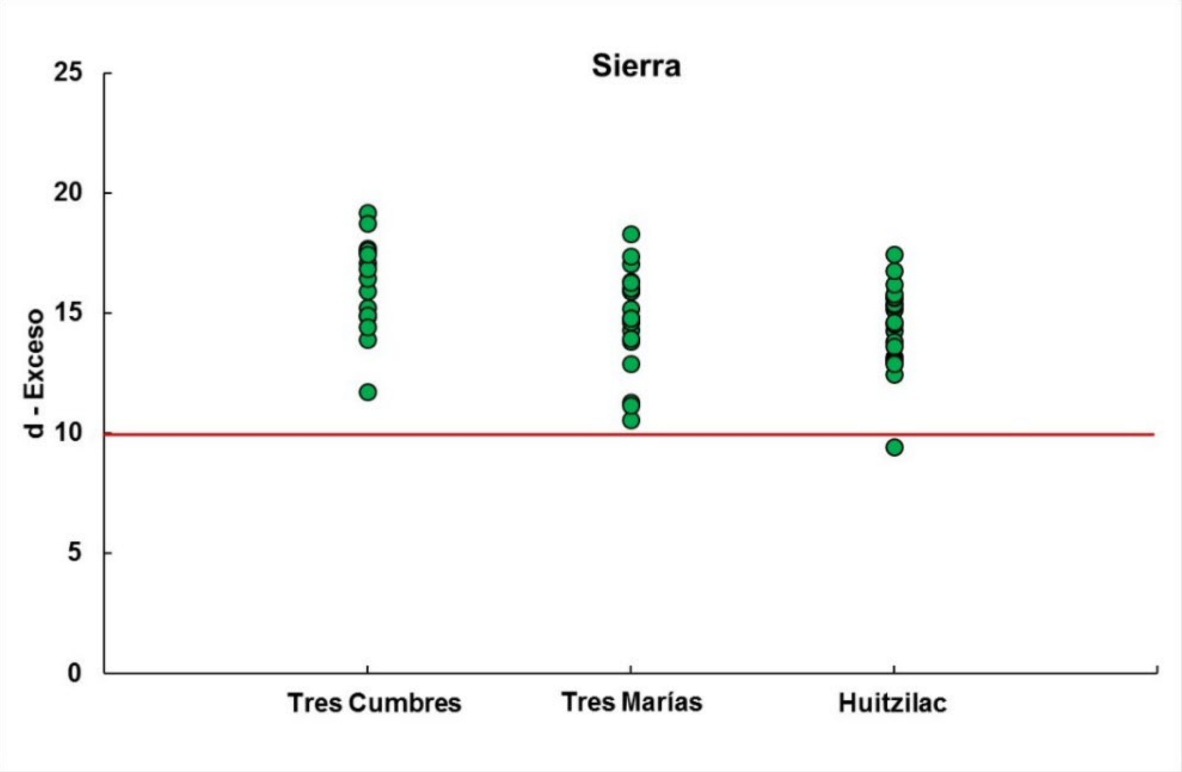

Figure 10. Deuterium excess values (d) of rainfall from collectors located in the Chichinautzin mountain range.

Vapor generated by evapotranspiration from forested areas has deuterium excess values greater than 10 (Galewsky *et al*., 2016). When this vapor condenses, it produces rain with deuterium excess values greater than 10. Therefore, the high values of deuterium excess and rainfall intercept of Tres Cumbres, Tres Marías, and Huitzilac indicate that evapotranspiration is an important source of moisture in the Chichinautzin mountain range. Gastmans *et al*. (2017) found a similar effect in Brazil. The Meteoric Water Lines of the Belo Horizonte, Rio de Janeiro, Campo Grande, Carolina, Betania and Rio Claro stations presented intercept values greater than +10 ‰. They attributed this effect to the recirculation of vapor generated in the Amazon rainforest. Esquivel-Hernández *et al*. (2019) also reports this effect in a tropical ecosystem in Ecuador.

In arid atmospheres, when rainfall is light or just beginning, raindrops on their descent from the clouds to the ground undergo evaporation (Peng, Mayer, Harris, & Roy, 2007). This is known as secondary evaporation or evaporation under the cloud. Fractionation produces enrichment of the $\delta^{18}O$ and $\delta^{2}H$, and decreases the deuterium excess (Froehlich, Kralik, Rank, & Scheifinger, 2008; Stewart, 1975; Dansgaard, 1964). When precipitation is intense or long duration, the environment becomes saturated and secondary evaporation stops.

In the case of the Cuernavaca valley, with two collectors located in very warm climate (Temixco and Tezoyuca), the lowest deuterium excess values correspond with the smallest precipitations, indicating secondary evaporation of light rain (Figure 11). With respect to the most abundant rainfall, their deuterium excess values are similar to the GMWL value (+10 ‰), indicating that these samples did not evaporate (Figure 11). Therefore, the Meteoritic Lines of the Cuernavaca valley collectors, have intercept values (+8.14 ‰, +6.28 ‰, +4.7 ‰) lower than the GMWL value (+10 ‰), due to secondary evaporation.

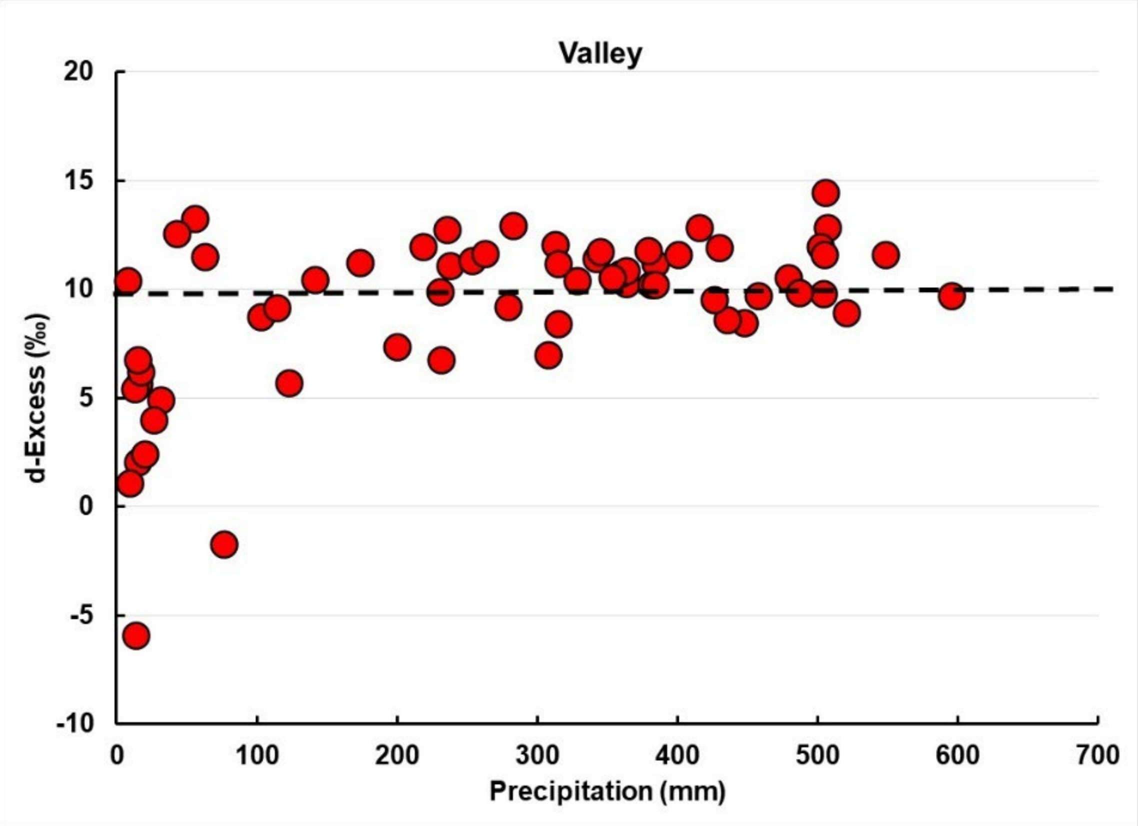

Figure 11. Values of deuterium excess (d) versus precipitation, of the collectors located in the valley of the Cuernavaca Aquifer.

The altitudinal isotopic gradient is represented by the equation (Figure 12): $\delta^{18}O$ = -0.0014 (Z)-7.1933 con R^2 = 0.8792 (Z in meters). Hence, the vertical gradient is -0.14 ‰/100m. In Figure 12, the $\delta^{18}O$ values correspond to precipitation-weighted average. This vertical gradient is similar to that found by Scholl Gingerich and Tribble (2002) (-0.12 ‰/100m) for rainfall from Maui, Hawaii; and Otte *et al*. (2017) (-0.11 ‰/100m) for rainfall from Mt. Kilimanjaro, Tanzania. The altitude effect explains 87 % of the variation in isotopic composition (R^2 = 0.87).

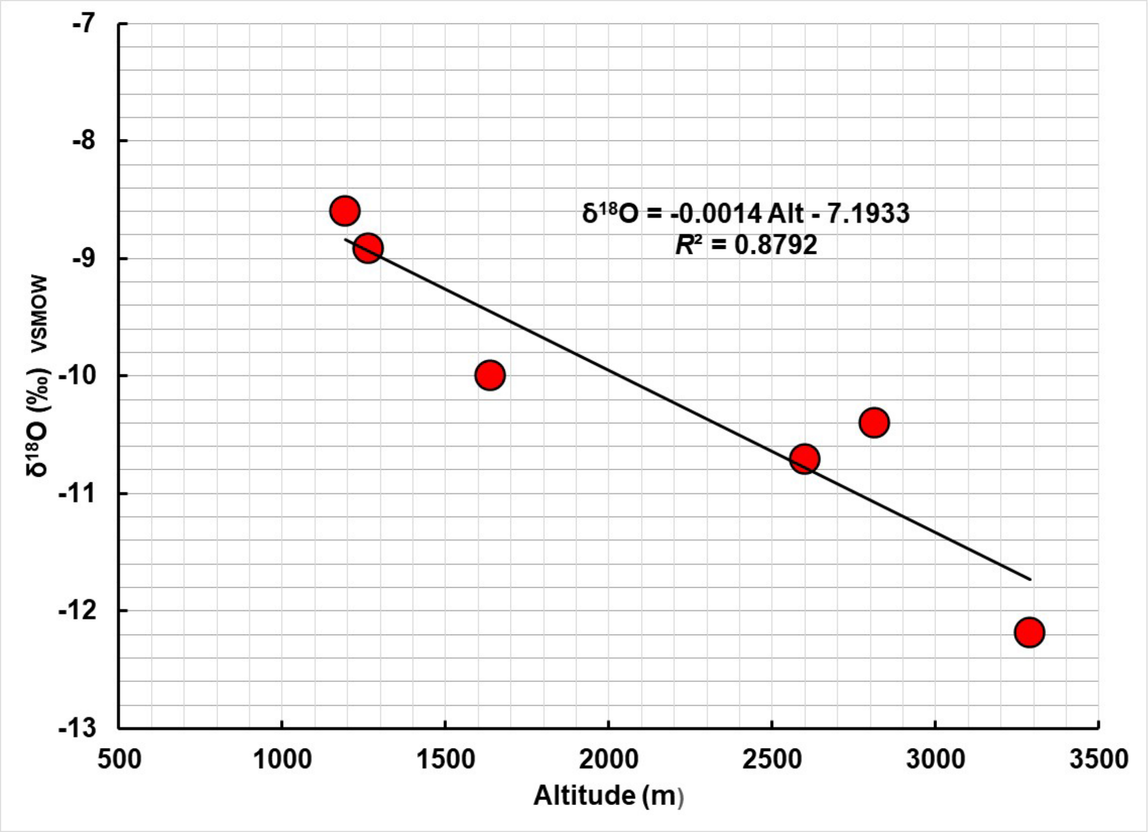

Figure 12. Altitudinal isotopic gradient of the $\delta^{18}O$.

Groundwater

Tables 2a, 2b, 2c and 2d shows the values of the physical-chemical parameters measured in the field and laboratory, of the samples from the south of the Cuernavaca aquifer.

Table 2a. Physical-chemical parameters of the samples from the south of the Cuernavaca aquifer.

ID	Date	Location			Temperature (°C)	Temperature of the sample (°C)	pH	EC (µS/cm)	DO (mg/l)
		X	Y	Altitude					
Well 1	30/01/2014	18°47.813	99°12.198	1186	21.2	19.4	6.85	641	0.30
Well 2	30/01/2014	18°48.792	99°11.058	1217	28.5	20.0	6.86	268	5.60
Well 3	31/01/2014	18°48.829	99°11.266	1220	23.3	22.7	7.07	620	7.30
Well 4	31/01/2014	18°48.500	99°12.030	1202	23.8	19.4	7.45	506	0.60
Well 5	04/02/2014	18°49.838	99°11.727	1219	23.7	24.0	6.75	902	6.50
Well 6	04/02/2014	18°49.650	99°10.865	1241	24.1	22.8	6.88	674	4.30
Well 7	05/02/2014	18°50.296	99°10.222	1241	25.5	22.7	6.81	831	3.90
Well 8	05/02/2014	18°48.588	99°11.713	1203	31.1	19.8	7.40	281	0.80
Well 9	06/02/2014	18°46.368	99°11.089	1138	23.9	20.6	6.80	279	5.20
Well 10	06/02/2014	18°48.376	99°11.538	1280	22.9	21.1	7.13	697	1.50
Well 11	06/02/2014	18°47.580	99°11.667	1182	26.7	19.5	7.29	777	0.90
Spring San Ramón	08/02/2014	18°47.519	99°11.983	1189	27.0	20.5	6.80	1207	0.60
Well 12	08/02/2014	18°46.893	99°12.576	1151	29.7	22.6	6.94	1270	4.30
Spring Chihuahuita	18/02/2014	18°45.385	99°11.338	1096	30.7	20.4	6.69	392	4.80
Well 13	19/02/2014	18°49.216	99°12.782	1232	27.6	22.4	6.97	542	3.50
Spring Palo Bolero	18/09/2014	18°46.015	099°14.312	1482	31.2	21.7	6.60	2240	1.08

Table 2b. Physical-chemical parameters of the samples from the south of the Cuernavaca aquifer (continued).

ID	Date	NO3 (mg/l)	PO4 (mg/l)	SO_4 (mg/l)	Na (mg/l)	Li (mg/l)	Mg (mg/l)	Al (mg/l)	Si (mg/l)	K (mg/l)	Ca (mg/l)	Ti (µg/l)	V (µg/l)	Mn (mg/l)
Well 1	30/01/2014	5.79	0.1530	145	13	0.028	27	0.020	25	2.2	100	< 2	< 2	0.0048
Well 2	30/01/2014	16.71	0.0306	26	17	0.005	14	0.010	26	3.0	30	< 1	14.8	0.0022
Well 3	31/01/2014	17.11	0.1530	82	23	0.032	22	0.010	29	4.4	86	< 1	11.7	0.0020
Well 4	31/01/2014	9.95	0.1530	77	14	0.010	19	0.020	25	2.3	64	< 2	4.2	0.0034
Well 5	04/02/2014	42.08	0.1530	97	54	0.010	25	0.020	31	6.8	109	< 2	4.6	0.0050
Well 6	04/02/2014	28.02	0.1530	69	24	0.012	33	0.010	35	5.3	72	2.9	13.3	0.0005
Well 7	05/02/2014	26.87	0.1530	102	29	0.013	38	0.010	34	5.3	96	2.8	12.6	0.0005
Well 8	05/02/2014	11.54	0.1530	67	15	0.016	19	0.449	28	2.7	57	24.1	12.6	0.0275
Well 9	06/02/2014	16.84	0.5508	30	16	0.010	14	0.043	27	2.8	33	< 2	10.8	0.0034
Well 10	06/02/2014	14.19	3.0600	52	18	0.010	24	0.020	28	3.5	79	< 2	3.6	0.0030
Well 11	06/02/2014	6.19	0.1530	240	13	0.023	32	0.001	25	2.4	127	1.8	5.5	0.0055
Spring San Ramón	08/02/2014	0.11	0.1530	429	16	0.054	49	0.020	24	2.8	230	< 2	< 2	0.0054
Well 12	08/02/2014	9.50	0.1530	451	20	0.059	56	0.010	29	3.7	225	2.4	1.6	0.0005
Spring Chihuahuita	18/02/2014	19.14	0.0306	33	14	0.005	13	0.112	23	2.5	32	2.6	15.7	0.0005
Well 13	19/02/2014	27.32	0.1530	45	22	0.031	36	0.010	37	2.6	34	2.7	14.1	0.0005
Spring Palo Bolero	18/09/2014	0.0001	0.0001	1100	20	0.060	86	0.020	24	3.2	437	2.8	< 2	0.0010

Table 2c. Physical-chemical parameters of the samples from the south of the Cuernavaca aquifer (continued).

ID	Date	Fe (mg/l)	Co (µg/l)	Cu (mg/l)	As (mg/l)	Rb (µg/l)	Sr (µg/l)	Mo (µg/l)	Cs (µg/l)	Ba (µg/l)	Tl (µg/l)	Pb (mg/l)	U (µg/l)	Tritium (UT)	Error % ionic balance
Well 1	30/01/2014	0.1	< 0.1	0.002	0.05	5.7	584	10.0	1.34	35.6	0.800	0.00010	0.86	0.83	2.14
Well 2	30/01/2014	0.05	0.070	0.002	0.0002	7.6	188	1.0	0.24	33.4	0.020	0.00200	0.76	0.79	2.87
Well 3	31/01/2014	0.05	< 0.05	0.001	0.01	4.0	979	1.0	8.28	102.0	0.020	0.00005	1.91	2.17	10.67
Well 4	31/01/2014	0.1	< 0.1	0.002	0.04	5.5	502	6.0	0.60	85.0	0.280	0.00010	1.25	0.76	6.86
Well 5	04/02/2014	0.1	0.540	0.006	0.01	12.1	321	2.0	0.48	90.2	0.140	0.00200	2.31	2.16	2.32
Well 6	04/02/2014	0.05	0.070	0.001	0.00	8.8	515	1.0	0.08	161.0	0.040	0.00005	2.76	1.35	2.23
Well 7	05/02/2014	0.05	0.140	0.001	0.00	8.3	736	1.0	0.04	264.0	0.040	0.00005	4.76	2.11	0.03
Well 8	05/02/2014	0.05	< 0.05	0.002	0.05	6.9	614	5.0	0.47	83.6	0.050	0.00200	1.43	0.12	0.08
Well 9	06/02/2014	0.1	< 0.1	0.002	0.0003	7.1	224	< 2	0.20	53.4	0.060	0.01000	0.95	0.75	4.67
Well 10	06/02/2014	0.1	< 0.1	0.002	0.03	7.6	950	2.0	0.58	136.0	0.100	0.00010	2.16	1.53	1.03
Well 11	06/02/2014	0.005	0.023	0.001	0.07	6.9	1430	10.0	1.25	29.2	0.440	0.00240	0.56	0.86	3.62
Spring San Ramón	08/02/2014	0.1	< 0.1	0.002	0.03	8.1	1550	2.0	2.46	21.2	0.160	0.00010	0.24	0.12	0.97
Well 12	08/02/2014	0.05	0.050	0.001	0.03	11.8	1960	2.0	2.02	30.9	0.080	0.00005	0.25	0.86	1.11
Spring Chihuahuita	18/02/2014	0.05	0.070	0.001	0.00	6.8	239	1.0	0.21	46.7	< 0.01	0.00005	0.91	0.12	8.16
Well 13	19/02/2014	0.05	0.060	0.003	0.00	5.5	333	< 1	0.14	39.0	< 0.01	0.00200	3.05	1.47	0.77
Spring Palo Bolero	18/09/2014	0.46	< 0.1	0.002	0.00	10.3	3180	< 2	3.04	14.4	< 0.02	0.00040	0.02		0.55

Table 2d. Physical-chemical parameters of the samples from the south of the Cuernavaca aquifer (end).

ID	Date	Redox (mV)	Alkalinity Total (mg/l)	HCO$_3$ (mg/l)	SDT (mg/l)	Hardness Total (mg/l)	δ^{18}O (‰)	δ^{2}H (‰)	Deuterium excess (d)	F (mg/l)	Cl (mg/l)	NO$_2$ (mg/l)	Br (mg/l)
Well 1	30/01/2014	152	212	259	462	361	-11.0	-77.4	10.9	0.59	8	0.082	0.05
Well 2	30/01/2014	477	121	148	193	131	-10.7	-74.4	11.2	0.23	9	0.016	0.13
Well 3	31/01/2014	349	168	205	446	305	-10.3	-73.1	9.6	0.41	27	0.082	1.12
Well 4	31/01/2014	453	211	257	364	240	-10.8	-76.7	10.1	0.56	16	0.082	0.45
Well 5	04/02/2014	354	256	312	649	375	-10.0	-71.7	7.9	0.60	86	0.082	3.71
Well 6	04/02/2014	370	232	283	485	315	-9.8	-70.6	8.2	0.23	39	0.082	1.34
Well 7	05/02/2014	429	277	338	598	396	-10.1	-71.9	8.8	0.21	58	0.082	2.28
Well 8	05/02/2014	434	160	195	202	222	-10.8	-75.7	10.7	0.88	20	0.082	0.79
Well 9	06/02/2014	278	144	176	201	138	-10.6	-74.4	10.4	0.26	12	0.016	0.17
Well 10	06/02/2014	402	240	293	502	294	-10.3	-73.8	8.3	0.05	36	0.033	1.21
Well 11	06/02/2014	255	184	224	559	447	-11.0	-77.8	10.6	0.74	8	0.082	0.05
Spring San Ramón	08/02/2014	36	328	400	869	775	-11.0	-77.0	11.3	0.95	16	0.082	0.05
Well 12	08/02/2014	320	323	394	914	791	-10.9	-76.6	10.4	0.62	21	0.082	0.05
Spring Chihuahuita	18/02/2014	425	144	176	282	133	-10.6	-74.2	10.7	0.31	13	0.016	0.19
Well 13	19/02/2014	389	208	254	390	233	-10.4	-73.4	9.4	0.28	17	0.082	0.05
Spring Palo Bolero	18/09/2014	120	307	375	1613	1448	-11.0	-76.7	11.5	0.76	19	0.0001	150

Water temperature ranges from 19.4 to 24 ºC; dissolved oxygen from 0.3 to 7.3 mg/l; pH from 6.6 to 7.45, and TDS from 193 to 1 613 mg/l. Nitrate concentrations range from 0 to 42 mg/l, below the permissible limit of 44.3 mg/l. The main anion is HCO_3^-, with the following order of abundance HCO_3^-» SO_4^{2-}» CL^-»NO_3^- (Figure 13).

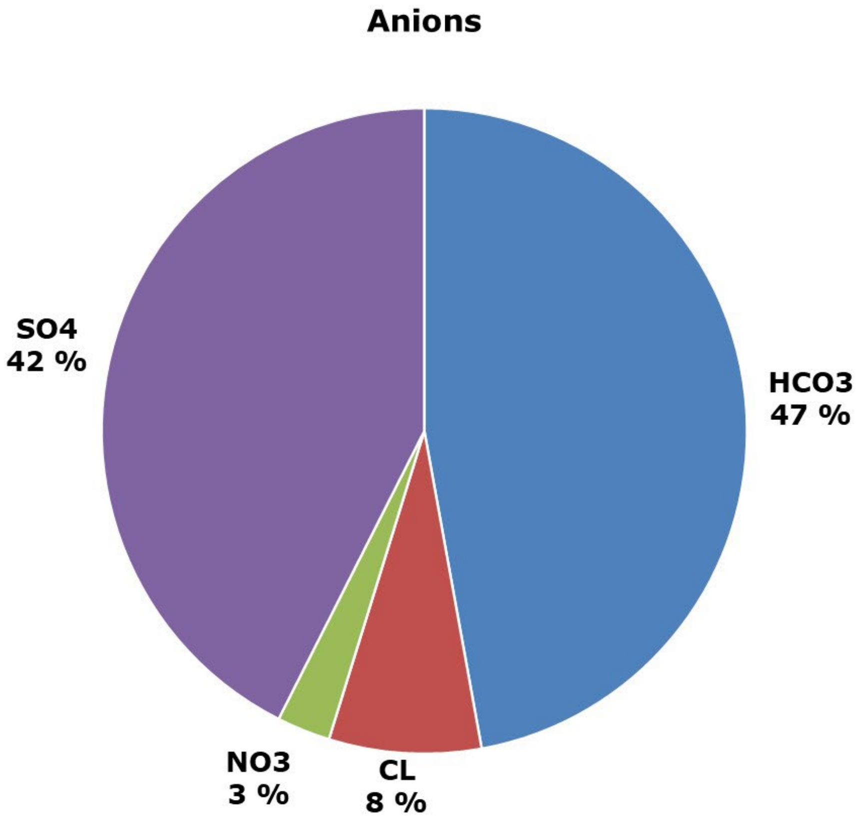

Figure 13. Anion composition of groundwater (% in meq/l).

The main cation is Ca^{2+}, with the following order of abundance Ca^{2+} » Mg^{2+} » Na^{+} » K^{+} (Figure 14).

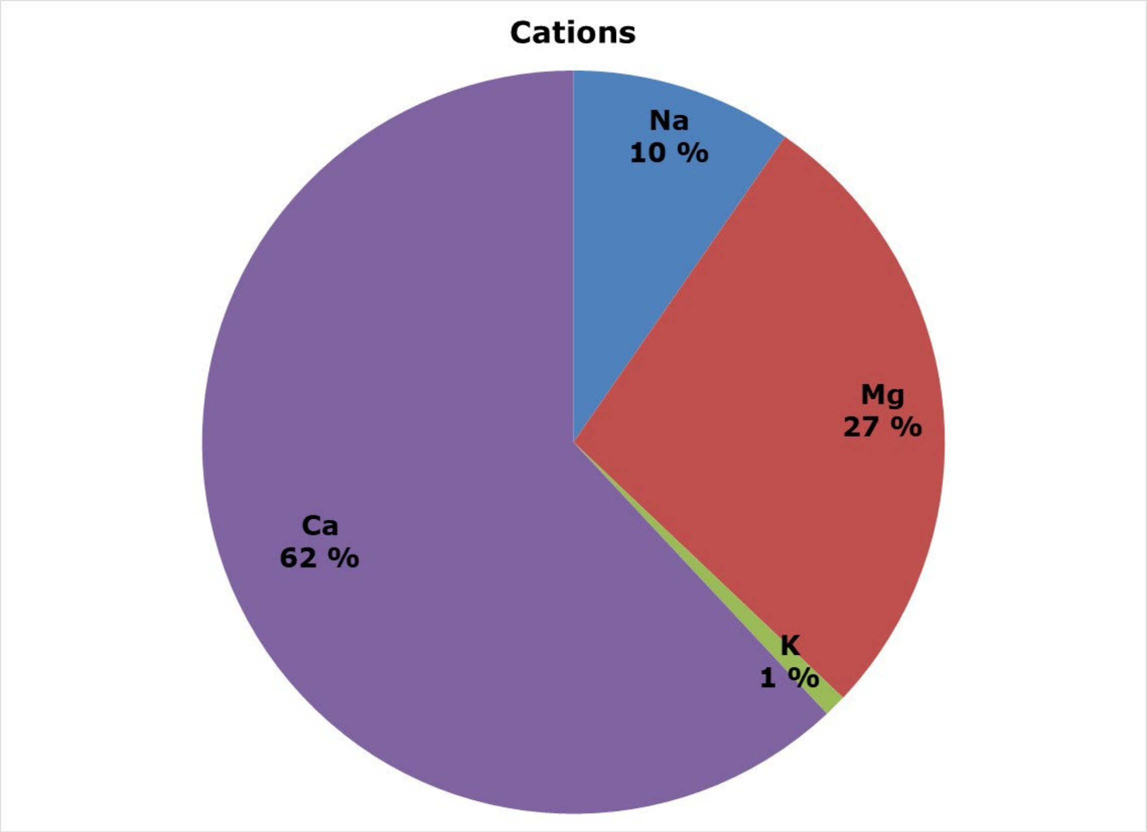

Figure 14. Cation composition of groundwater (% en meq/l).

Flow through the Chichinautzin formation produces Ca-Mg-HCO3 type waters (Figure 15) (Chihuahuita spring, well 9 and well 2). Flow through the Cuernavaca and Balsas formations produces Ca-Mg-HCO3-SO4 type waters (well 1, well 4 and well 8). Flow through the Morelos and Cuautla formations produces Ca-Mg-SO4-HCO3 type waters (Palo Bolero spring, San Ramon spring, well 11 and well 12). A group of wells (3, 5, 6, 7, 10 and 13) have higher concentrations of chloride than the rest of the wells.

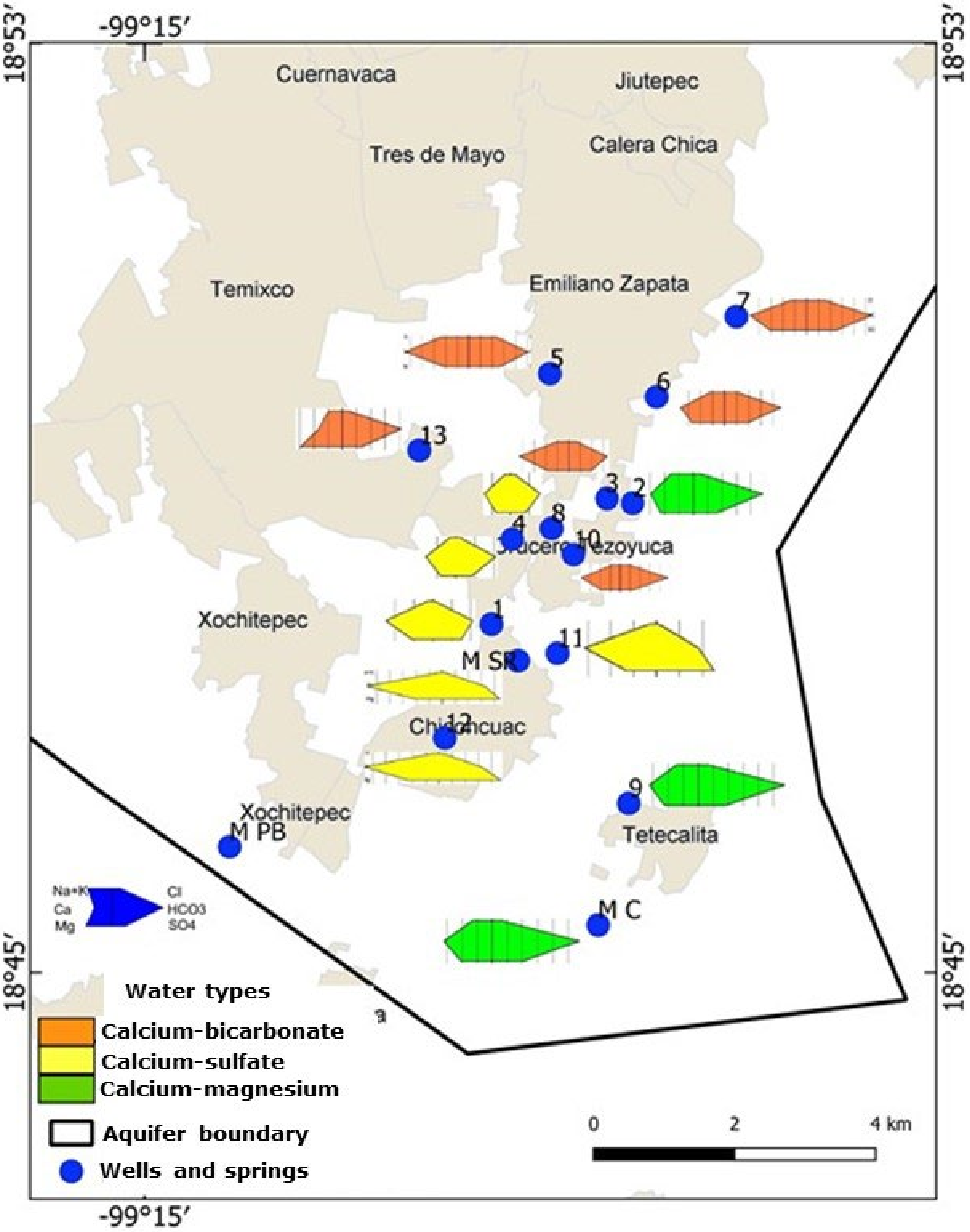

Figure 15. Stiff diagrams of groundwater samples.

The concentration of nitrate (NO_3^-) for 2014 showed levels lower than 5 mg/l in the eastern portion of the aquifer, where rocks of the Chichinautzin Formation predominate, composed mainly of fractured basalts (Figure 16). In contrast, the western portion showed the highest nitrate levels, with concentrations greater than 30 mg/l, where the Temixco-Acatlipa urban zone is located (Figure 16). Most of the urban zone of Cuernavaca, Jiutepec, and Emiliano Zapata, showed concentrations ranging from 10 to 20 mg/l (Figure 16).

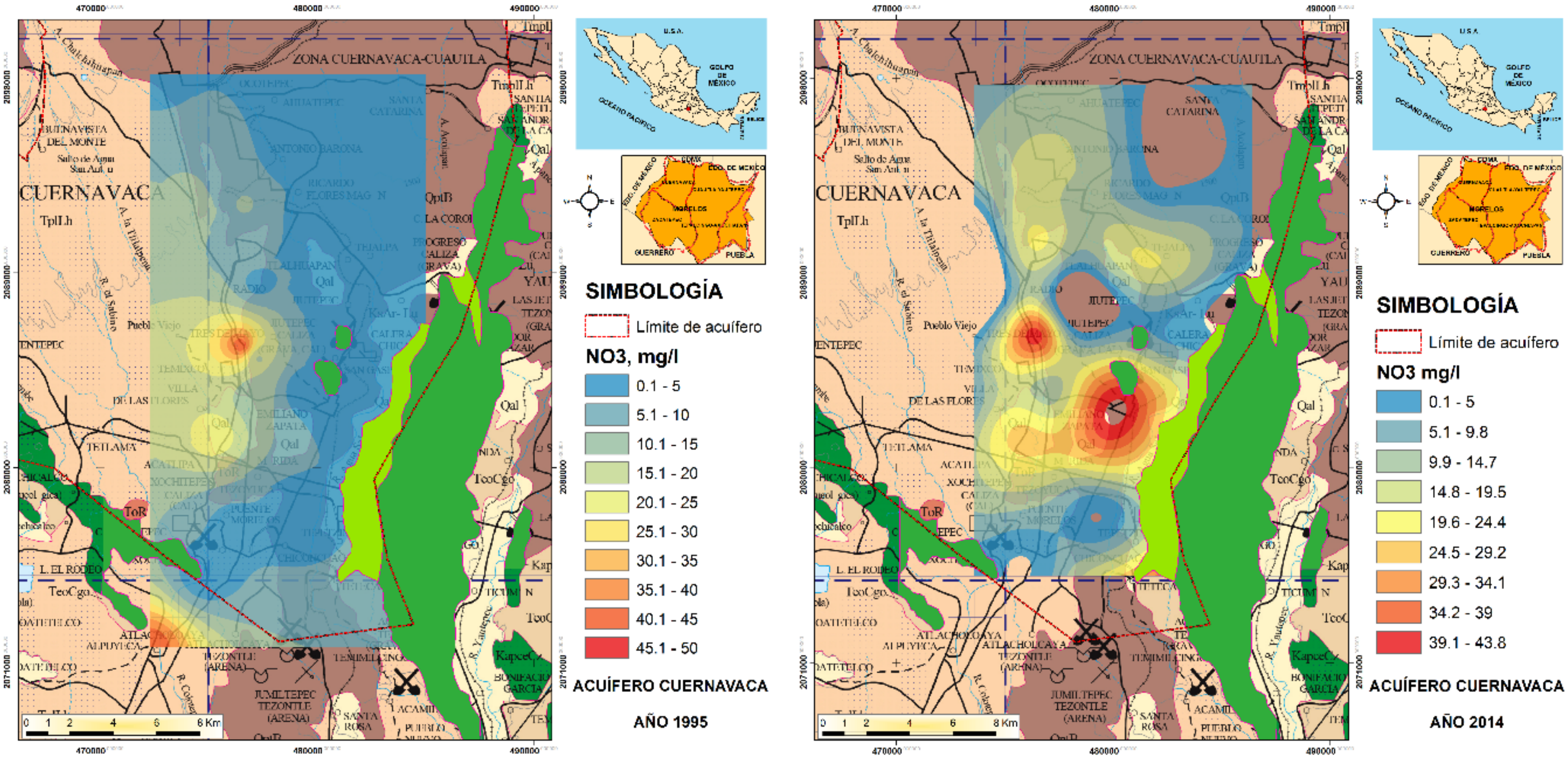

Figure 16. Nitrate concentrations in the years 1995 and 2014.

It is important to note the higher concentrations of uranium in wells with high concentrations of nitrate (Figure 17). Ingestion of drinking water with high uranium concentrations can cause damage to the liver (Brugge & Buchner, 2011) and bones due to alpha radiation (Kurttio *et al*., 2005). The World Health Organization (WHO) established a maximum permissible limit of 15 µg/l for uranium (WHO, 2008).

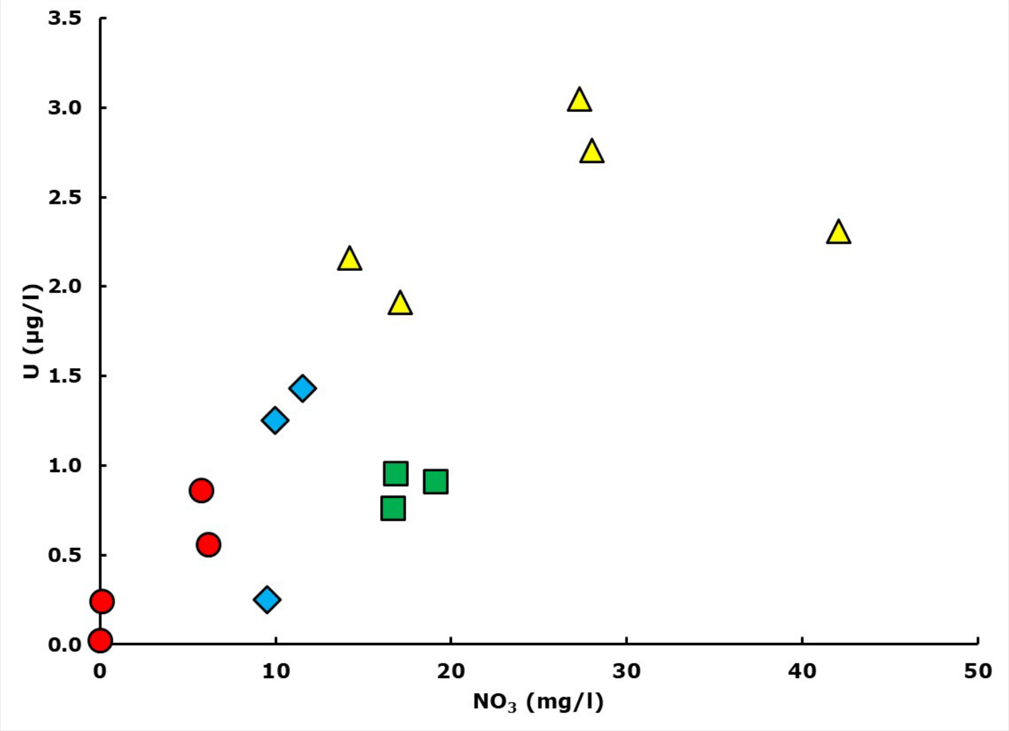

Figure 17. Uranium *versus* nitrate.

The oxidized states of uranium U(VI) are soluble in groundwater, while the reduced U(IV) species are immobile (Singh *et al*., 2014; Nolan & Weber, 2015; Paradis *et al*., 2016). Reduced U(IV) uranium species are very susceptible to being oxidized, when groundwater contains oxidizing species such as nitrate (NO_3^-, y NO_2^-). Nolan and Weber (2015) found in two major aquifers in the United States, High Plains and Central Valley, a significant correlation between nitrate and uranium. They conclude that nitrate, the primary contaminant in these aquifers, should be considered as a factor inducing secondary uranium contamination.

Stable isotopes

The stable isotope data of oxygen and hydrogen in a scatter plot $\delta^{18}O$ vs. δ^2H allow us to identify and differentiate the types of water of each well (Figure 18). The graph identifies four groups of waters characterized by the altitude at which the effective recharge of groundwater takes place. The first group corresponds to wells that capture water of depleted isotopic composition, with $\delta^{18}O$ values between -11.05‰ and -11.02‰, and δ^2H between -77.75‰ and -76.71‰ (wells 1 and 11, and San Ramón and Palo Bolero springs), due to the fact that their recharge zone is located at higher elevations than those corresponding to the rest of the wells.

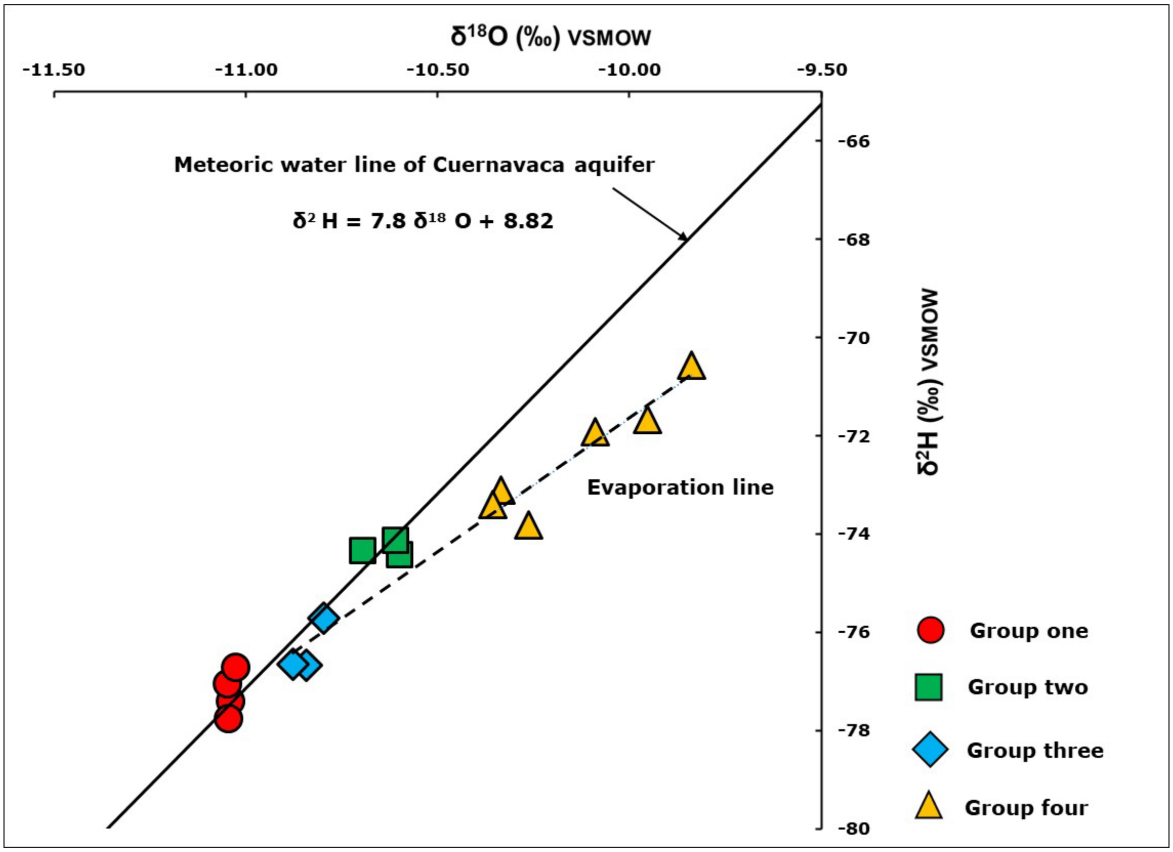

Figure 18. Isotopic composition ($\delta^{18}O$, $\delta^{2}H$) of water from wells.

The second group is characterized by $\delta^{18}O$ values between -10.69 and -10.60 ‰, and $\delta^{2}H$ between -75.6 and -74.1‰, which corresponds to the recharge zones with the lower elevation (wells 2, 9 and Chihuahuita spring).

A third group, formed by wells 4, 8 and 12, which capture water with $\delta^{18}O$ and $\delta^{2}H$ values intermediate between group one and two.

The fourth group corresponds to waters that evolved isotopically from the previous groups, because they contain water components enriched in heavy isotopes due to intense evaporation. The isotopic values are located around a line with a slope lower than the Cuernavaca Aquifer line (LMAC). This group is formed by wells 3, 5, 6, 7, 10 and 13. In the area where these wells are located there are extensive areas of irrigated crops, a golf course and several wastewater canals. The isotopic compositions vary for δ^{18}O between -10.35 and -9.84‰, and for δ^{2}H between -73.4, and -70.6‰. This fourth group was classified as wells with evaporated water recharge.

Figure 19 shows for the fourth group a proportional increase in δ^{18}O and Cl^{-}, evidencing recharge with water subjected to intense evaporation before infiltrating into the soil.

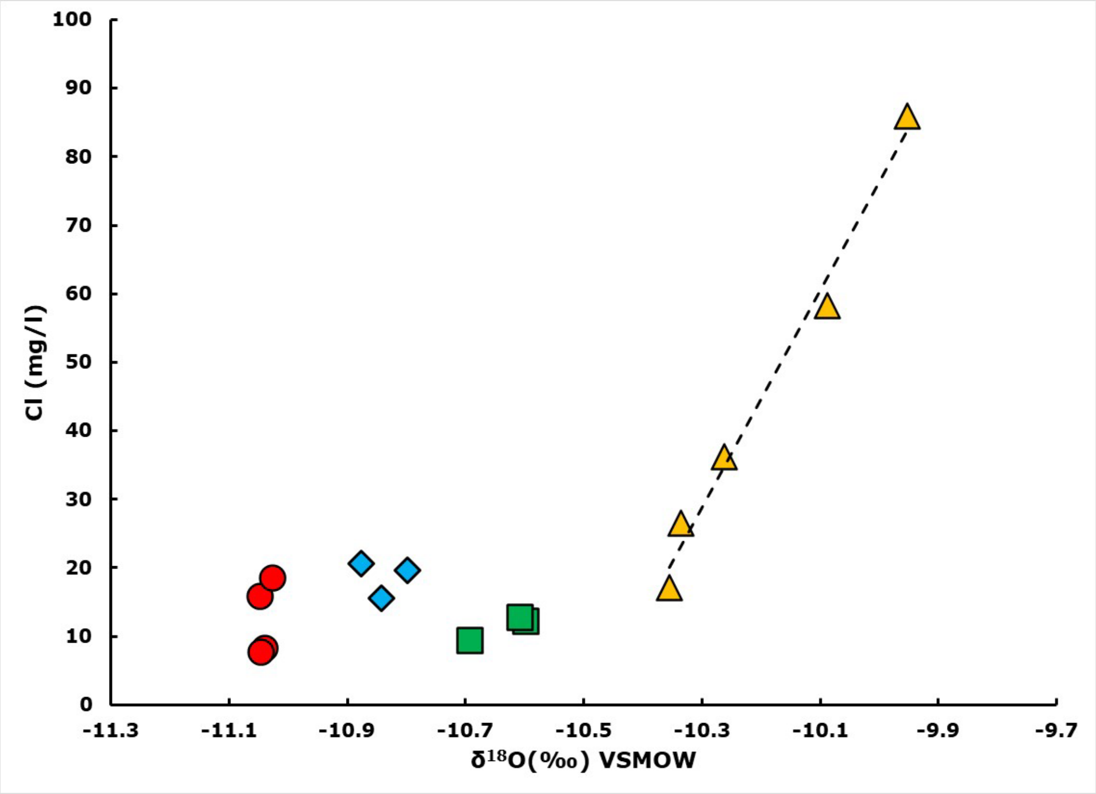

Figure 19. Values of δ^{18}O *versus* Cl.

Evaporation decreases the deuterium excess, while recycled moisture increases it (Froehlich *et al*., 2008). Evaporation of a surface water body produces deuterium excess values lower than 10 (Victoria, Martinelli, Mortatti, & Richey, 1991). Therefore, Figure 20 indicates that water samples with higher NO_3 content contain evaporated water.

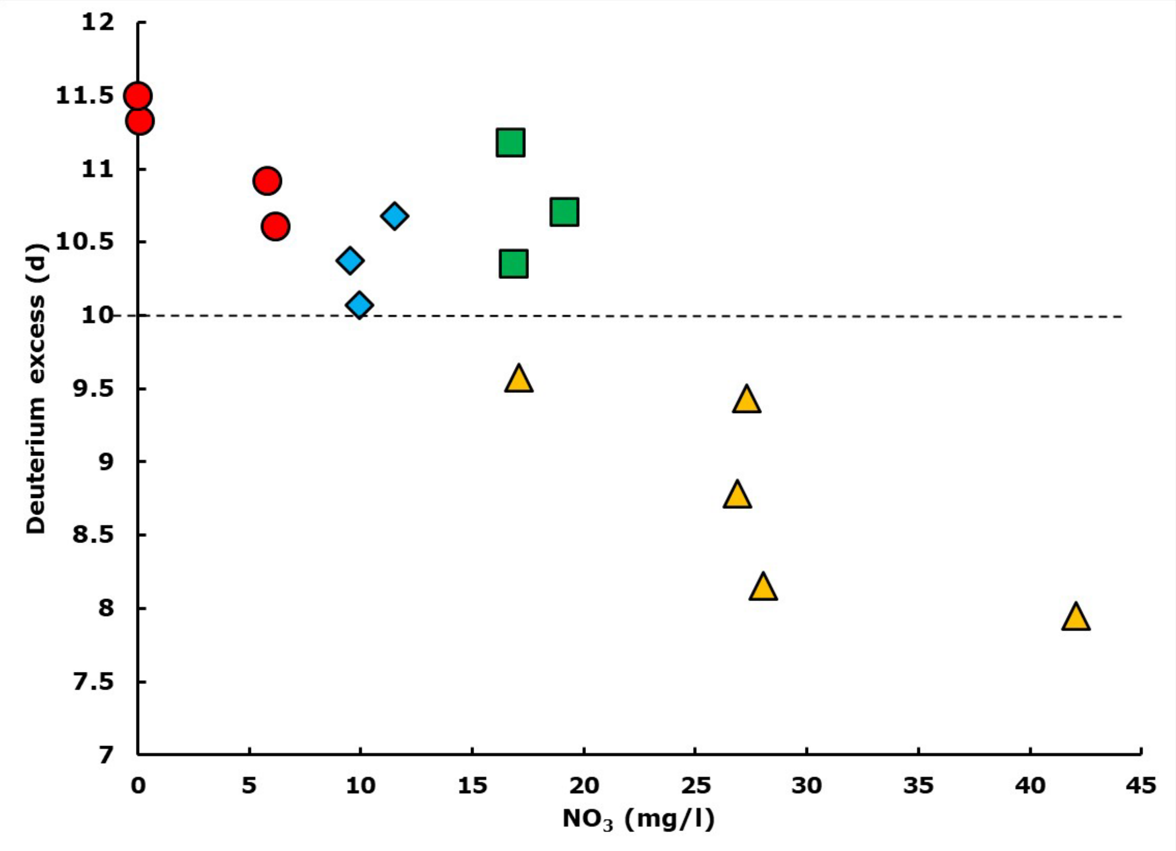

Figure 20. Values of NO_3 *versus* deuterium excess (d).

Tritium

The group of wells with evaporated water contain between 1.5 and 2.5 tritium units (TU), indicating very short residence times, i.e., they receive modern water recharge (Figure 21). On the other hand, wells with non-evaporated water have tritium contents of less than 1 TU, indicating longer residence times. In the case of the San Ramón spring, the absence of tritium indicates water with residence times of more than thirty years after infiltrating the aquifer.

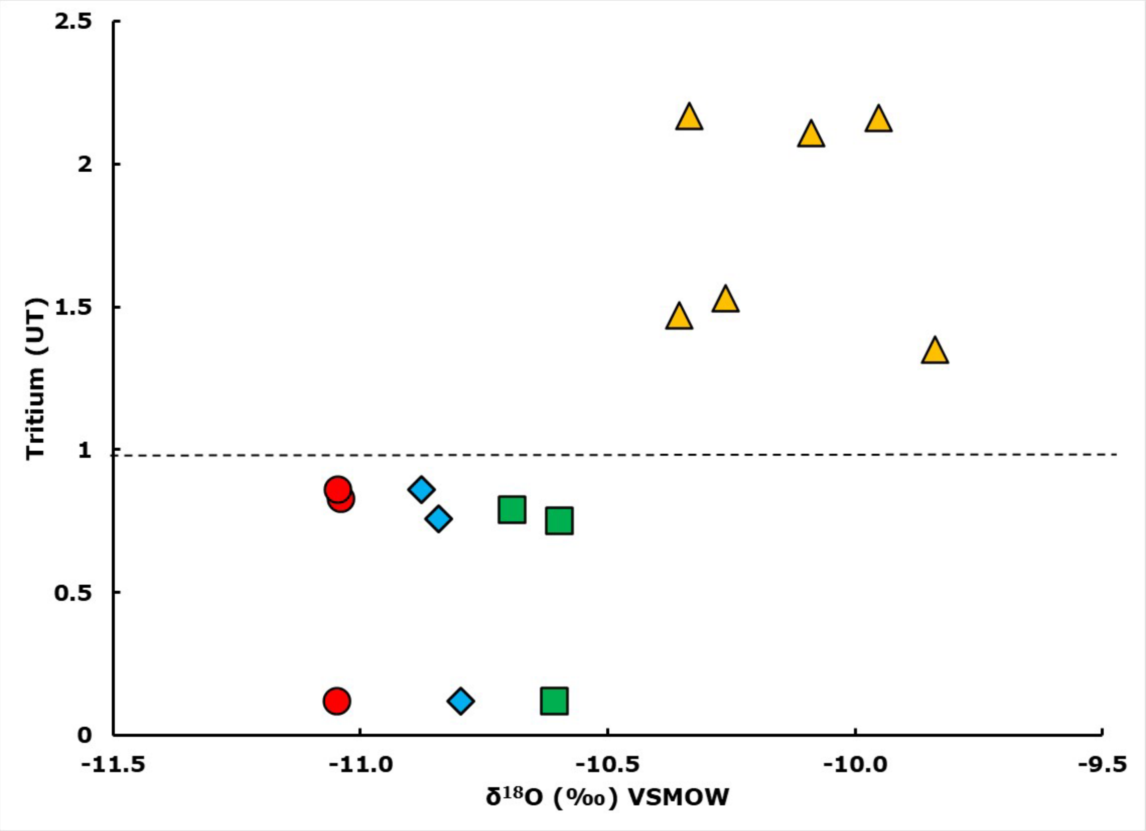

Figure 21. Values of δ^{18}O *versus* tritium.

The water samples with the highest NO_3 values contain between 1.5 and 2.5 tritium units. This indicates very short subsurface residence times (Figure 22).

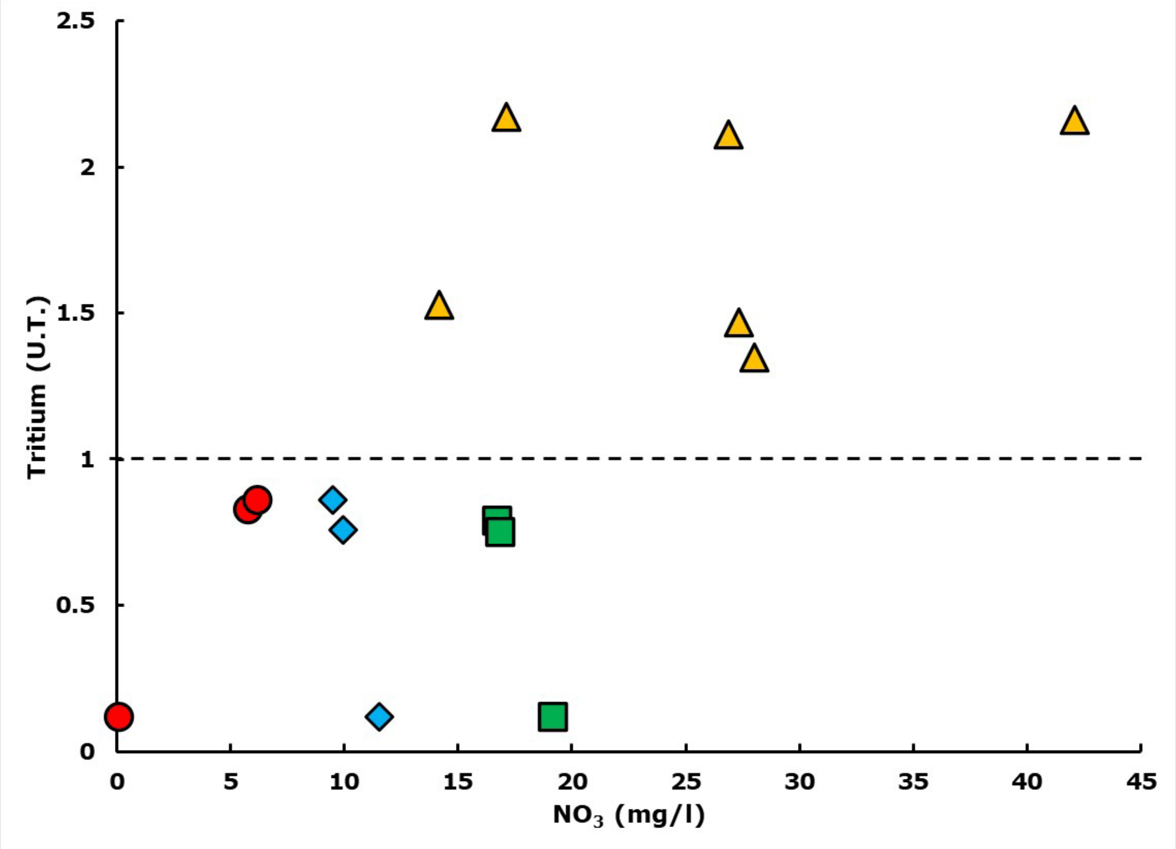

Figure 22. Values of NO_3 *versus* tritium.

Ionic relations

The dissolution of gypsum produces a Ca^{2+}/ SO_4^{2-} equal to 1:

$$CaSO_4 \cdot 2H_2O \leftrightarrow Ca^{2+} + {SO_4}^{2-} + 2H_2O$$

Samples from the Palo Bolero spring, San Ramón spring, and wells 11 and 12 are in the 1:1 equiline, indicating that gypsum dissolution is an important process in the chemical composition of these samples (Figure 23).

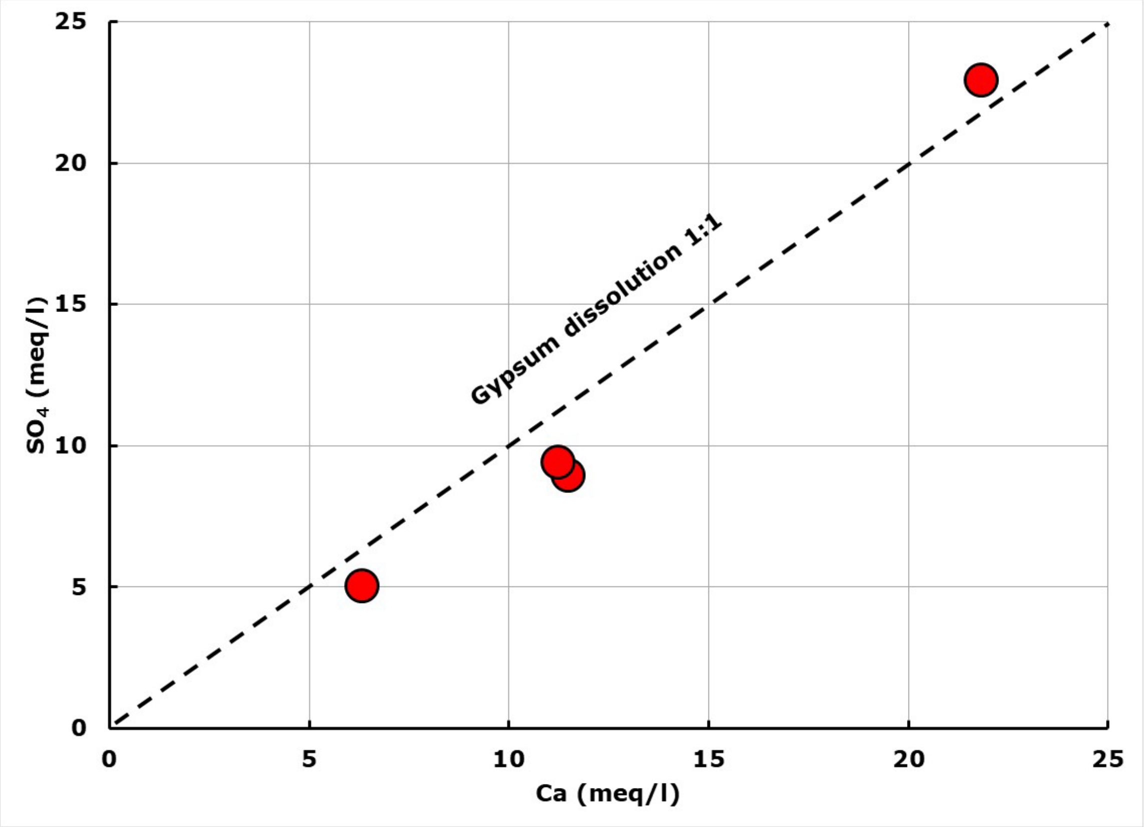

Figure 23. Values of Ca *versus* SO_4.

The geochemical processes that control the chemical composition of water can be elucidated using the Gibbs diagram (Gibbs, 1970). In Figure 24, total dissolved solids (TDS) are plotted as a function of the cation ratio ($Na^+/(Na^+ + Ca^{2+})$). Four groups can be distinguished in Figure 24. In the flow through the Morelos and Cuautla formations (red circles), the dominant process is the weathering of geologic material. In the flow through the Chichinautzin formation (green squares), the dominant process is rainfall. In the flow of the Cuernavaca formation (blue diamonds), its ionic composition is determined by weathering of the geologic material, but also by the effect of pluvial precipitation. In the fourth group (yellow triangles), the predominant process is evaporation.

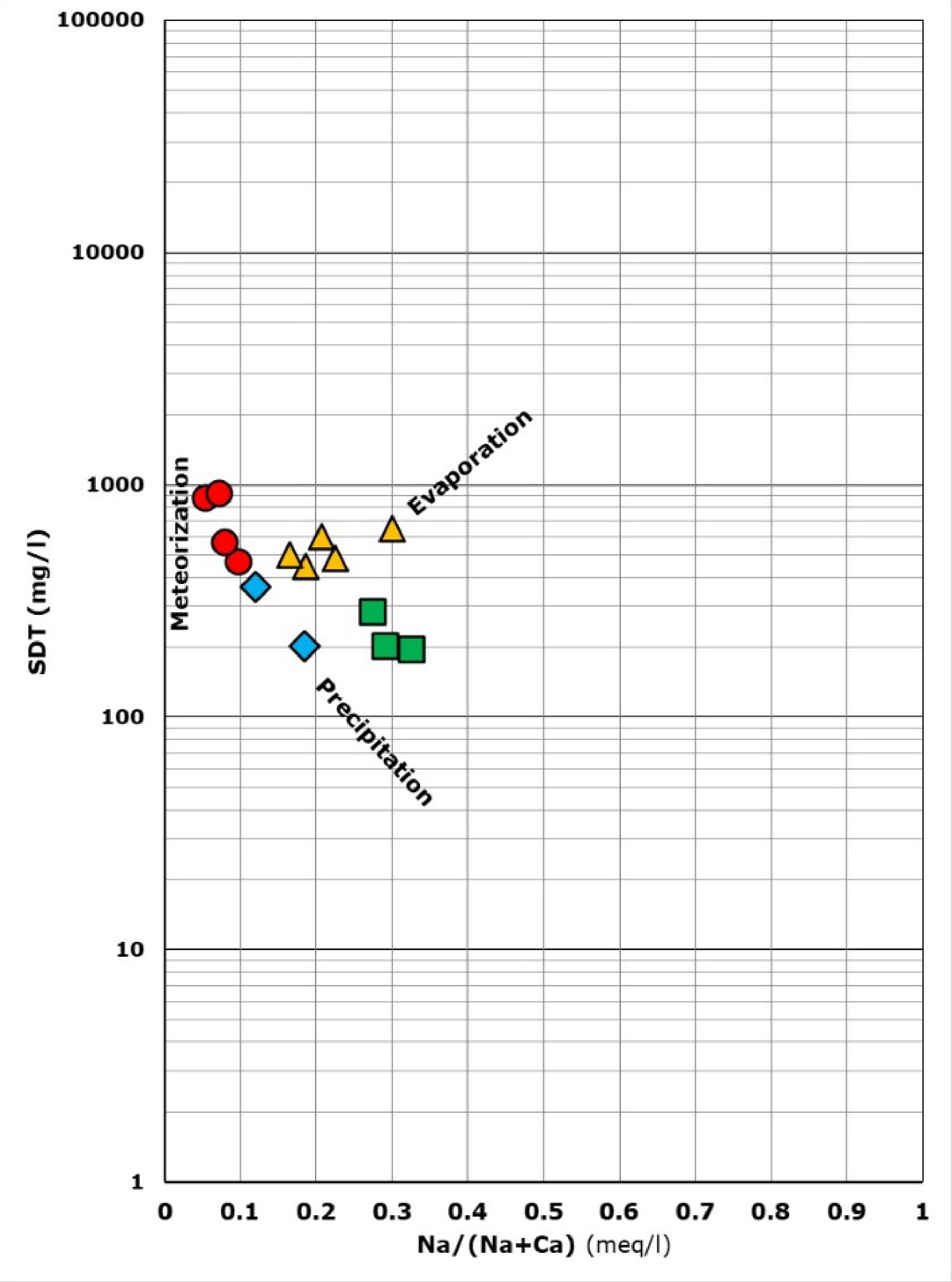

Figure 24. Gibbs diagram: Cation ratio.

Linear regression between nitrate and chloride content yielded a value of R^2 = 0.531 for the evaporated samples (Figure 25). Correlation values greater than r^2=0.35 indicate that the groundwater contains domestic or municipal wastewater (Ritter & Chirnside, 1984; Pacheco, Marín, Cabrera, Steinich, & Escolero, 2001).

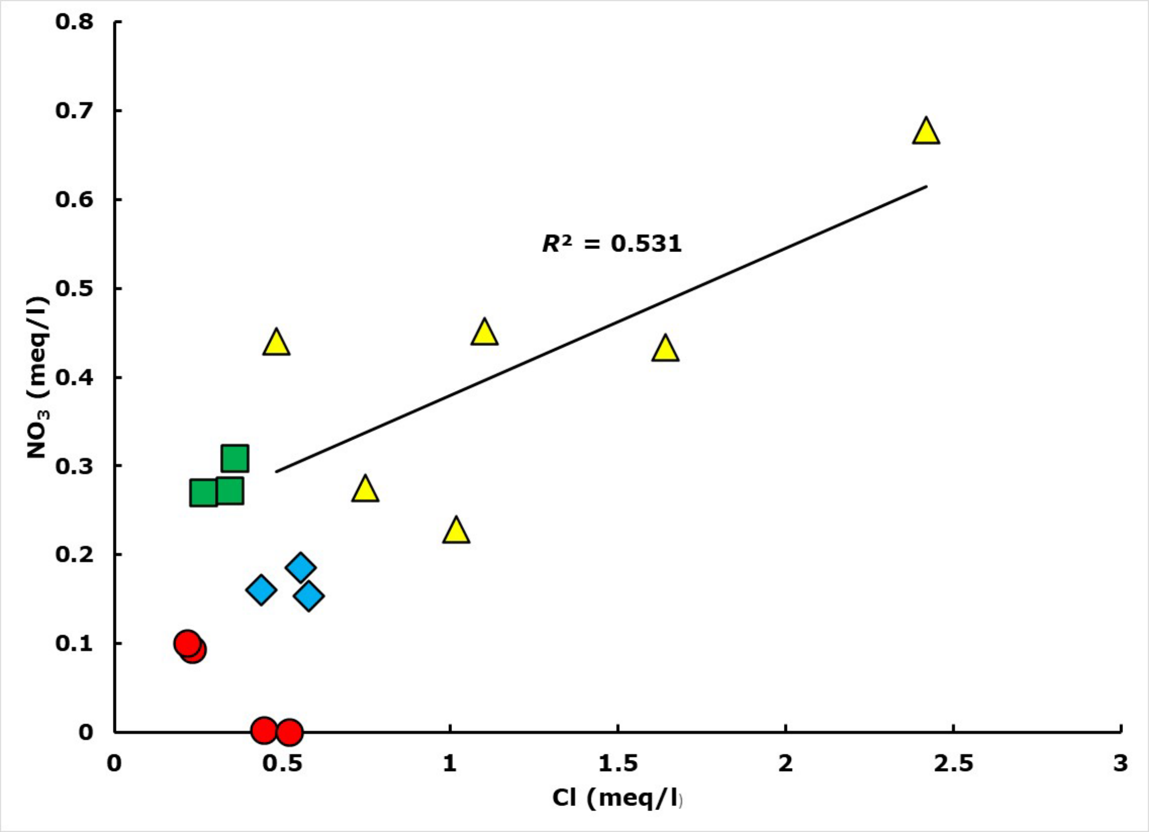

Figure 25. Values of Cl *versus* NO_3.

The graph (molar ratio) of chloride versus nitrate normalized by chloride (NO3/Cl), allows to elucidate the origin of the nitrate (Figure 26).

Samples fall in the blue box if the origin is agricultural, in the purple box if the nitrate is from manure, etc. The graph indicates that the nitrate in the evaporated samples comes from wastewater (green box) (Liu, Li, Lang, & Xiao, 2006; Guo, Yan, Wang, Xu, & Yang, 2020; Torres-Martínez *et al*., 2021).

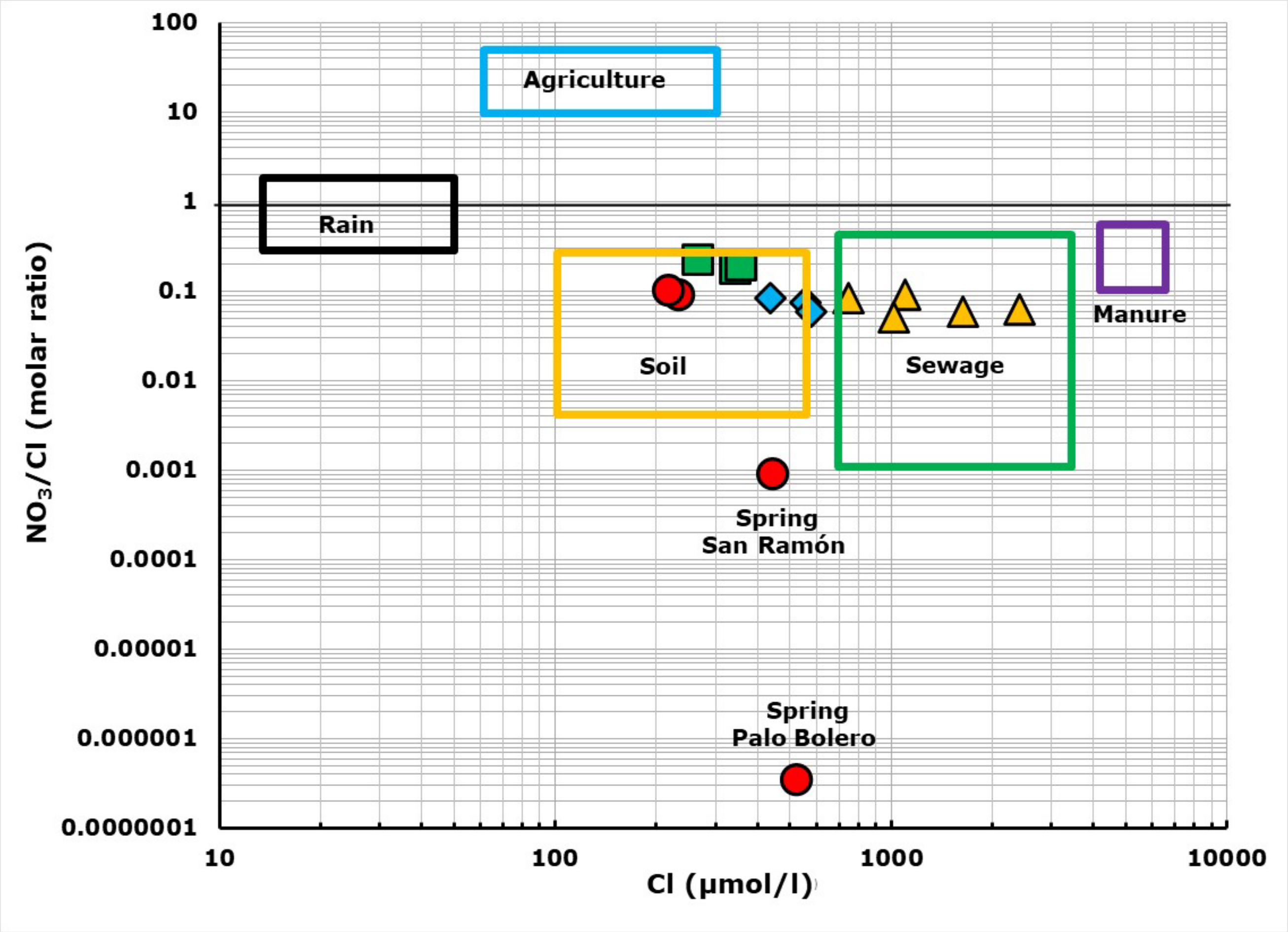

Figure 26. Chloride (molar concentration) *versus* NO_3/Cl (molar ratio).

The chloride and sodium values of the evaporated water samples (Figure 27) are similar to the values of septic tank effluent in the United States (Panno *et al*., 2002).

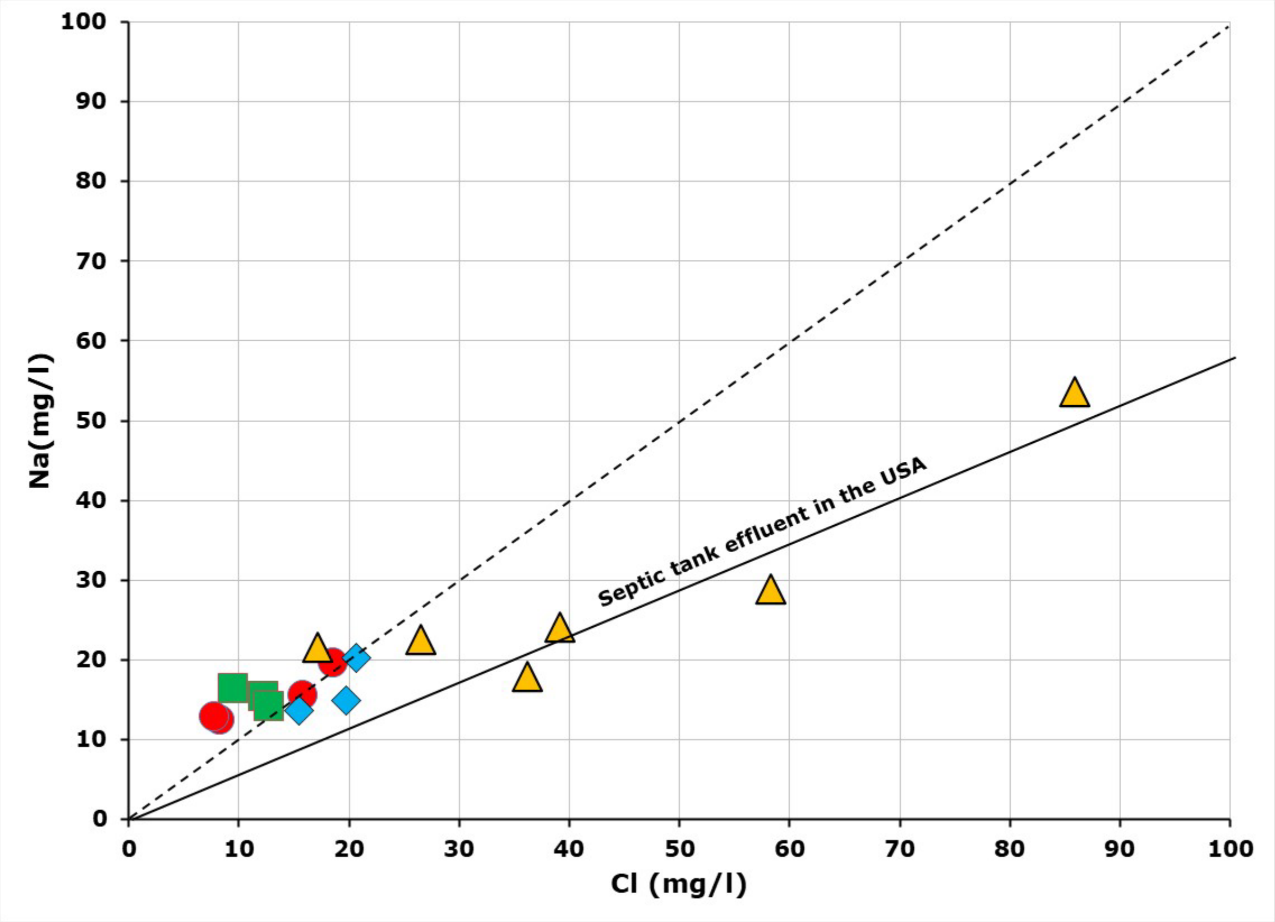

Figure 27. Chloride *versus* sodium.

Samples that exceed the permissible limit for arsenic (NOM 127-SSA1-127 modified in 2000) (SSA, 1994) are those with low dissolved oxygen concentrations (Figure 28) and tritium values lower than 1 TU

(Figure 29), indicating that wells with high arsenic values are recharged by deep flows.

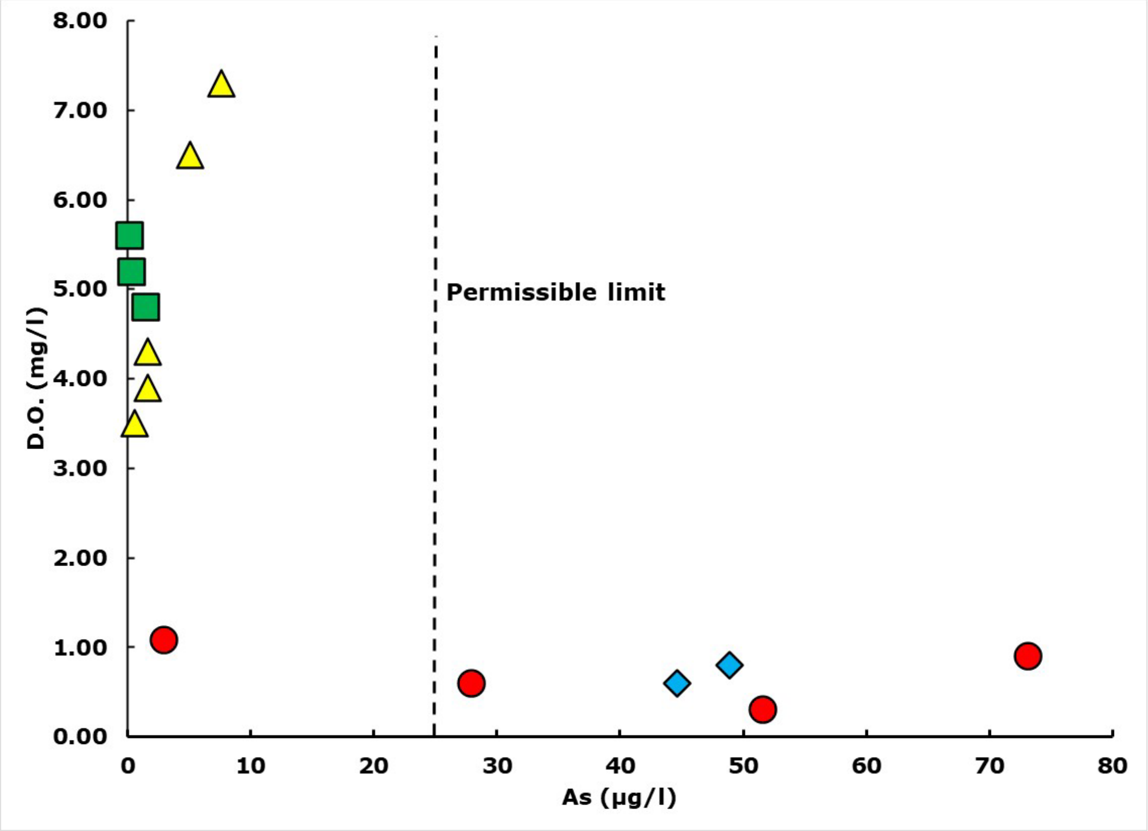

Figure 28. Values of Arsenic *versus* dissolved oxygen.

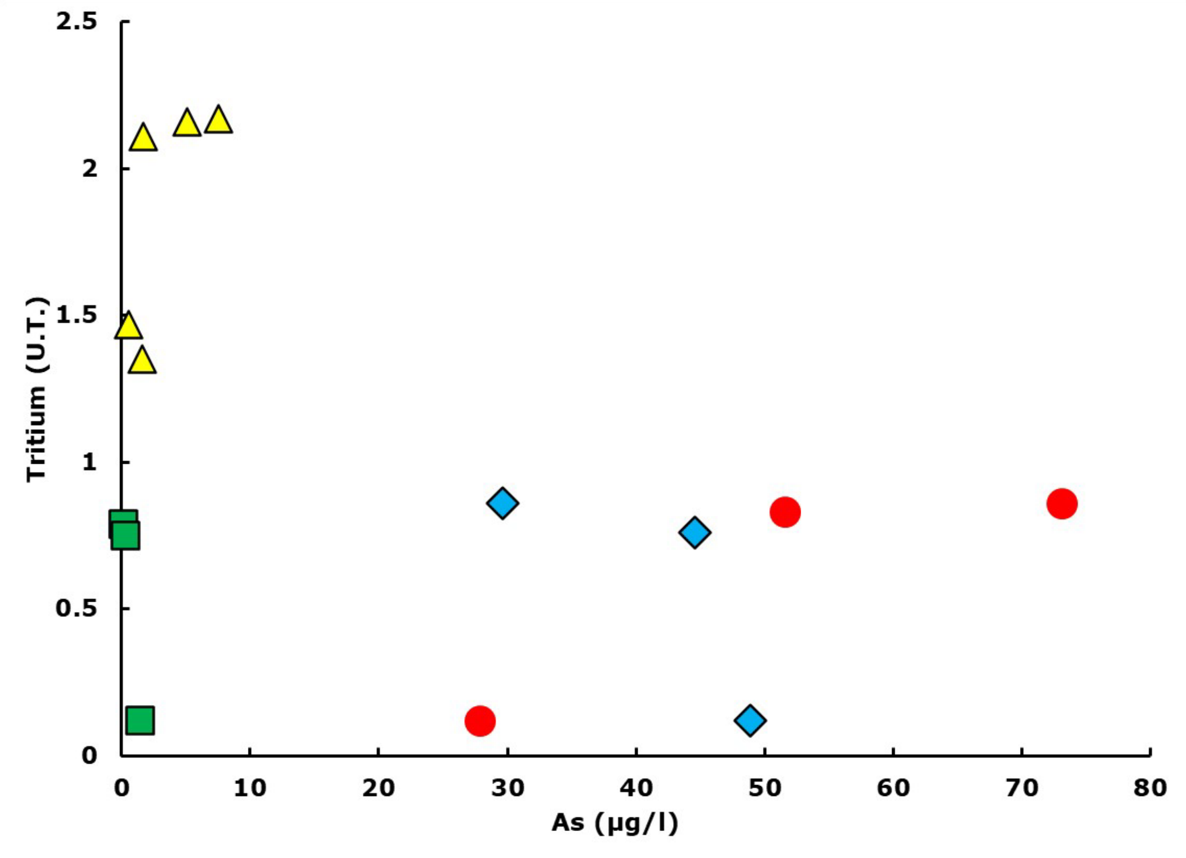

Figure 29. Arsenic *versus* tritio.

Torres-Martínez *et al*. (2021) determined in the Comarca Lagunera, an agricultural region in northern Mexico, the sources of nitrate to groundwater: Manure (47 %), sewage (43 %), fertilizers (5 %), soil organic nitrogen (3 %), and atmospheric deposition (2 %). Their results indicate that even though the Comarca Lagunera is a very important agricultural area, synthetic fertilizers are not the main source of nitrate. Ren, Zhang, Wang and Wang (2021) conducted a study to determine the sources of nitrate in surface and groundwater in the Ye River basin of northern China. It is a predominantly agricultural area, whose populations do not have sanitary sewers. For their study, they used stable isotopes of nitrogen and water, as well as a Bayesian mixing model. Their results indicate that the main contribution comes from wastewater and manure. They suggest building infrastructure for wastewater treatment. Pastén-Zapata, Ledesma-Ruiz, Harter, Ramírez and Mahlknecht (2013) conducted a study in a citrus-growing area in northeastern Mexico to determine the sources of nitrate in shallow groundwater. Based on geochemical evidence, they determined that the sources of nitrate in rural areas are the use of manure as fertilizer and wastewater, while in urban areas it is wastewater.

Our results indicate that the nitrate present in the south of the Cuernavaca aquifer comes mainly from wastewater, which is consistent with sites with similar problems, such as those mentioned above.

Nitrate in surface water and groundwater originates from multiple sources, and it is difficult to accurately distinguish these sources using hydrogeochemical methods alone. Nitrate isotopes have been shown to be a very useful tool for identifying nitrate sources (Torres-Martínez *et al*., 2021; Ren *et al*., 2021). In a new study being conducted in the Cuernavaca aquifer, hydrogeochemical methods as well as stable isotopes of water and nitrate are being used.

Conclusions

This study presents the use of environmental water isotopes and geochemistry to determine the concentrations and origin of nitrate that degrade water quality in the southern Cuernavaca aquifer.

The results show that the zones of the aquifer recharged with intensely evaporated water have the highest nitrate concentrations. The results also indicate that the nitrate comes from domestic or municipal wastewater, probably generated by untreated sewage discharges. None of the samples exceed the permissible limit of 44.3 mg/l of nitrate. However, the evolution of the last thirty years shows a significant increase in the area, which is why it is important to implement public policies that promote the construction and rehabilitation of infrastructure to collect wastewater and send it to existing treatment plants (La Gachupina and Emiliano Zapata WWTPs).

The data obtained also revealed a correlation between nitrate and uranium. Even though all samples have uranium concentrations below the permissible limit, it is recommended to continue monitoring this parameter, since an increase in nitrate concentrations could be accompanied by an increase in uranium concentrations. Because uranium decay increases the radioactivity of groundwater, it is recommended to measure radioactivity to establish the current values and monitor its evolution over time.

With respect to arsenic, the groundwater samples that exceed the permissible limit for arsenic have low dissolved oxygen and tritium concentrations, indicating that they come from the deep strata of the aquifer. If groundwater extraction in the area increases in the future, it could induce the extraction of deeper and deeper flows, with a consequent increase in arsenic.

The data obtained also allowed the determination of the Meteoric Water Line of the Cuernavaca Aquifer. This line will be very useful for future geohydrological and environmental studies.

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