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Arsenic contamination in groundwater in Zimapan, Hidalgo, Mexico

F. Prieto García[a,*], O.A. Acevedo Sandoval[b], F. Pérez Moreno[c], J. Prieto Méndez[b], R.A. Canales Flores[d]

[a]*Academic Area of Chemistry, Autonomous University of Hidalgo State, Road Pachuca—Tulancingo km. 4.5, P.O. Box 42076, Mineral de la Reforma, Hidalgo, Mexico, email: prietog@uaeh.edu.mx*
[b]*Institute of Agricultural Sciences, Autonomous University of Hidalgo State, Road Pachuca—Tulancingo km. 4.5, P.O. Box 42076, Mineral de la Reforma, Hidalgo, Mexico, emails: acevedo@uaeh.edu.mx (O.A. Acevedo Sandoval), jud_292003@yahoo.com.mx (J. Prieto Méndez)*
[c]*Academic Area of Materials and Earth Sciences, Autonomous University of Hidalgo State, Road Pachuca—Tulancingo km. 4.5, P.O. Box 42076, Mineral de la Reforma, Hidalgo, Mexico, email: perezmoreno@yahoo.com.mx*
[d]*Environment, Autonomous University of Hidalgo State, Road Pachuca—Tulancingo km. 4.5, P.O. Box 42076, Mineral de la Reforma, Hidalgo, Mexico, email: racf1306@yahoo.com.mx*

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ABSTRACT

Arsenic contamination in groundwater has been detected in several studies in the region of Zimapan, Mexico. Arsenic contamination sources have been reported as natural and anthropogenic. In this study, the variation in concentrations of pollutants in mineralized zones, sedimentary rocks and anthropogenic activities was determined. Arsenic and lead concentrations of 5.0–248.0 and 0.10–132.0 μg L^{-1} were observed in groundwater, respectively. Water mineralization was medium to medium high. Conductivity values between 198.0 and 970.0 μS cm^{-1} were determined. Some contaminated wells showed conductivity values as much as 1,700 μS cm^{-1}. Open wells near mining waste showed conductivity values up to 1,485.0 μS cm^{-1}. Groundwater showed neutral pH and alkalinity tendency. Arsenic concentrations were low in the volcanic aquifer, and high in the aquifer with carbonate rocks. The average total arsenic concentration was 47.8 μg L^{-1}. Arsenic concentrations of 20–100 μg L^{-1} were observed in 93% of the sampled points; concentrations greater than 50 μg L^{-1} were found in 20% of the total. Forty-seven percent of the evaluated points correspond to samples associated with carbonate rocks.

Keywords: Arsenic; Antimony; Groundwater quality in Zimapan; Hidalgo

1. Introduction

Knowledge and evaluation of groundwater has been studied in Zimapan, Hidalgo, Mexico. As, Pb, Sb, and other contaminants have been found. In some cases, the concentrations exceed national and international standards for drinking water. In this sense, 10 μg L^{-1} is the permissible limit of arsenic in drinking water in Mexico [1–7].

Arsenic bioconcentration depends on the rate of absorption and elimination. Arsenic is known as a carcinogen to which humans are exposed everywhere in the world. The genotoxic activity of this element

*Corresponding author.

in humans has been demonstrated in several investigations [2–7].

The presence of arsenic in groundwater in the region of Zimapan has been reported by several authors [4,5,7–16]. In 1998, the well El Muhi was closed by state health authorities due to high concentrations of As, Pb, and Sb were detected [13,14]. The presence of these three elements in the aquifers of Zimapan is attributed to natural sources (such as proximity to mineralized zones and mineralization processes of carbonate rocks) and anthropogenic sources (such as smelters and mine tailings).

Toxicity and collateral damage caused by the consumption of water with high levels of As is known from the decade of the 1960s. Arsenic can be found in various chemical forms. As(V) is the most common form in surface water and is the predominant form in groundwater. It is known that these chemical forms As(III) is more toxic than As(V). This regard, toxicology studies have established a correlation between chronic exposure to this element and the development of cancer in humans [3,6,17–19].

Arsenic toxicity in humans can be acute and chronic. The acute toxicity results from the consumption (in a short time) of water with a high content of As, and chronic toxicity is a consequence of consumption (for a long time) of water with small amounts of As. In addition, chronic exposure to As causes liver, kidney, and bladder cancer. On the other hand, some symptoms of acute and chronic poisoning As are: abdominal pain, vomiting, diarrhea, muscle pain, rash, and keratosis and hyperkeratosis of the hands and feet. Other symptoms are renal and cardiovascular dysfunction, encephalopathy, and headaches [20–22].

In recent years, industrialized countries have shown a tendency to reduce the maximum permissible levels of arsenic in drinking water due to the carcinogenic risk for its consumption [22]. Until 1996, the maximum allowable level of arsenic in drinking water in Mexico was 0.05 mg L^{-1}; however, Mexico modified this value to 0.045 mg L^{-1} in 2000, then to 0.025 mg L^{-1} in 2005, and finally to 10 μg L^{-1} in 2010 [22].

According to the literature, arsenic can be absorbed through the skin, inhalation, and consumption of contaminated water [2,3,5,6]. During several investigations some cases of chronic arsenic toxicology have been observed in Zimapan. Most cases were the result of consumption of arsenic-contaminated water for prolonged periods.

The region of Zimapan is on the edge of the Sierra Madre Oriental and Trans-Mexican Volcanic Belt. Therefore, the relief of this area is related to the type of rock, and endogenous and exogenous processes that have occurred in the region [23]. This began in the Jurassic Period of the Mesozoic Era with the formation called Las Trancas (sedimentary and volcanic composition consisting of calcareous shale, limonite, limestone, pyrite, and arsenopyrite). Then, in the Cretaceous Period two sedimentary formations were

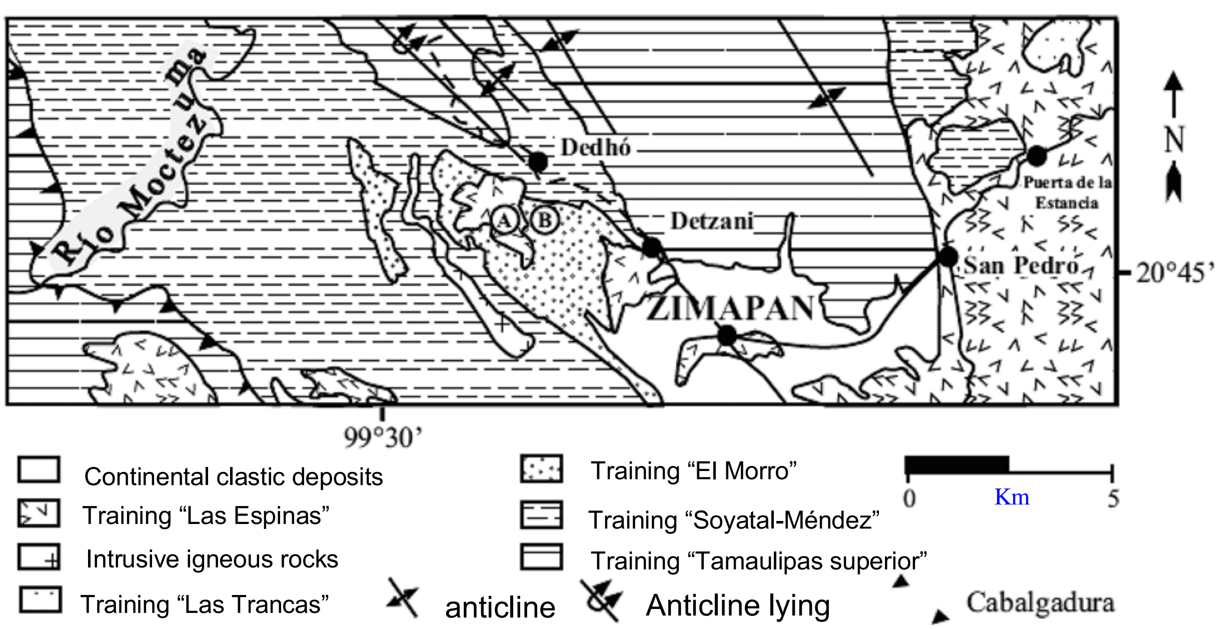

Fig. 1. Geographic location and local geology. Presence of arsenic in northwest Zimapán and the hills El Morro (location B) and Las Espinas (location A) [23–25].

Era	Period	Series	Training	Thickness (m)	Lithology	Lithological description
Cenozoic	Quaternary	Pleistocene Holocene	Aluvion	12	Qal	Gravel, sand, silts, clays composition sedimentary
			Agglomerated Zimapán	15		angular clasts and sub-angular to cemented limestones by caliche
	Tertiary	Miocene	“Las Espinas”	375	Te	Lavas, tuffs and agglomerates andesitic-basaltic
		Oligocene	Agglomerated “El Morro”	400 Unconformably	Tm	Alluvial fans, semiangulares fragments of limestone and volcanic
Mesozoic	Cretaceous	top	“Soyatal”	1000	Ks	Lean alternating limestones The lithology varies laterally In some places more calcarea and other more pelitic
		lower	“Tamaulipas” “El Doctor” “Tanimul”	to 1200 and to 2000	Kit, Kid, Kita	These formations are correlated Distinguished by facies and geographical location. Tamaulipas: limestone slope and basin El Doctor and Tanimul: platform limestones
	Jurasic	top	“Las Trancas”	to 3000		Phyllites calcareous shales siltstones, micritic limestones with pyrite, sandstone and chert

Fig. 2. Stratigraphic column of Zimapán region (Simons and Mapes, 1956, as amended by Martin, 2000, and Yta and Moreno, 1997. Taken from [23]).

observed. This formations are El Doctor or El Abra (lower), and El Soyatal (top) (Figs. 1–3) [23,26].

2. Materials and methods

Zimapan is located between geographic coordinate 20˚40´ and 20˚50´ N, and 99˚20´ and 99˚25´ W, in the western of the state of Hidalgo, Mexico (Fig. 4) [23,28]. This region is located 1,813 m above sea level in a micro-watershed, and has an area of 905.8 km^2 within the watersheds of the Panuco River and the Moctezuma River.

In Zimapan, 30 water sampling points were selected. These points have been monitored since 2000

Fig. 3. View from Detzani northwest of the hill “El Morro”.

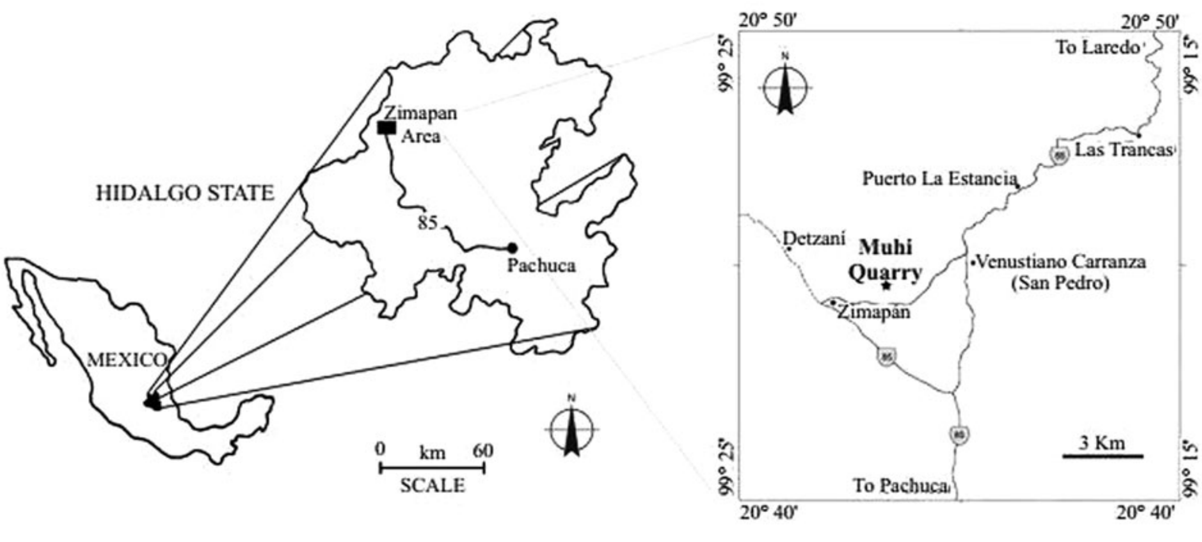

Fig. 4. Location of Zimapán City, Hidalgo, Mexico [23,27].

until 2010. Measurements of anions, cations, and trace elements (As, B, Cu, Fe, Mn, Pb, and Zn, among others) were performed. All analyses were conducted by volumetric and spectrophotometric methods as reported in several papers [4,5,7,12,14,17,22,23]. The collection of samples and the methods used for physical and chemical characterization were those reported in previous studies [15,26]. This was done in two different series.

Principal component analysis (PCA) with the use of SPSS version 17.0 [29] to the results from 30 water samples were performed. As random variables were considered: Ca^{2+}, HCO_3^-, SO_4^{2-}, K^+, Mg^{2+}, Na^+, Cl^-, NO_3^-, SiO_2, Mn^{2+}, Fe^{3+}, Sb^{3+}, As(V) in form of

Table 1
Significant associations of the factor loadings above 0.6

Sampling carried out in 2000–2010	
Component	Variables
1	SO_4^{2-}, Ca^{2+}, Mg^{2+}
2	$HAsO_4^{2-}$, Fe^{3+}, Sb^{3+}, Pb^{2+}
3	$HAsO_4^{2-}$, HCO_3^-, Cl^-, NO_3^-
4	Mn^{2+}, Na^+, K^+

Table 2
Variation coefficient and average of the water characterization analysis of the Zimapán wells

Analysis	NOM [22]	Average
Al (μg L^{-1})	<200	22.00 (6.10)
As (μg L^{-1})	<10	186.3 (2.20)
Ba (μg L^{-1})	<700	48 (3.10)
Ca^{2+} (mg L^{-1})	NN	28.32 (8.62)
Calcium hardness (mg L^{-1})	NN	106.6 (2.80)
Cd (μg L^{-1})	<5.0	2.085 (0.76)
Cl^- (mg L^{-1})	<250	1.11 (7.0)
Color UPC	<20	<5
Conducivity (mS cm^{-1})	NN	0.49 (5.90)
Cr (μg L^{-1})	<50	<0.10
Cu (μg L^{-1})	<2,000	11.0 (0.02)
Fe (μg L^{-1})	<300	180.0 (2.40)
HCO_3^- (mg L^{-1})	<300	242.0 (8.59)
Hg (μg L^{-1})	<1	<0.5
K^+ (mg L^{-1})	NN	2.24 (2.02)
Mg^{2+} (mg L^{-1})	NN	8.81 (8.04)
Mn^{2+} (μg L^{-1})	<150	230.0 (2.0)
Na^+ (mg L^{-1})	<200	5.70 (6.35)
NO_2^- (mg L^{-1})	<1	0.002 (10.2)
NO_3^- (mg L^{-1})	<10	0.04 (4.30)
Odor	Nice	Nice
Pb (μg L^{-1})	<10	7.2 (4.00)
pH (U)	6.5–8.5	7.30 (0.40)
Residual Cl_2	0.2–1.5	0.02 (0.30)
SO_4^{2-} (mg L^{-1})	<400	48.5 (2.12)
Temperature (°C)	NN	25.6 (2.5)
Total dissolved solids (mg L^{-1})	<1,000	244.3 (3.10)
Total Solids (mg L^{-1})	NN	307.3 (7.10)
Turbidity (UTN)	<5	0.5 (1.30)
Zn (μg L^{-1})	<5,000	8.00 (6.20)

$HAsO_4^{2-}$, and Pb^{2+}. Table 1 shows the most significant associations indicated to the point where the factor loadings of a random variable are superior to those of the original variables. The treatment performed was varimax orthogonal. The variables with factor loadings below 0.6 are not represented.

3. Results and discussion

Table 2 shows average values of all analyses performed in wells studied. The results of the water characterization are shown in Fig. 5. Based on a piper diagram [30], Zimapan water is classified as water rich in calcium, magnesium, and bicarbonate. The points of different colors in the piper diagram show the results in the two sampled series.

Water samples from the aquifer system of Zimapan showed conductivity levels between 198 and 974 μS cm^{-1}. On the other hand, water of contaminated wells showed high conductivities values of 1,485–1,700 μS cm^{-1}.

Groundwater showed neutral pH tending to alkalinity and high oxidizing character. In addition, the collected water of the volcanic aquifer showed very low levels of arsenic while the aquifer with carbonate rocks showed very high arsenic content [2,7,23,31].

Fig. 6 shows the total arsenic concentrations. In Zimapan, the average total arsenic concentration for the 30 sampled points is 47.8 μg L^{-1} which is 4.8 times the normative value [22]. Arsenic concentrations of 20–100 μg L^{-1} were observed in 93% of the sampled points; concentrations greater than 50 μg L^{-1} were found in 20% of the total. Fig. 6 also shows the values of standard deviation of the average results of the sampled points.

Fig. 7 was obtained as results of the PCA performed with SPSS version 17.0 [29]. The component 1 was characterized by ion correlations that contribute to water hardness and the component 2 by correlations between some trace elements, especially contaminants as $HAsO_4^{2-}$ and Pb^{2+}. This last component interrelates As, Pb, and Fe, and indicates the presence or contact with sulfides or sulfoarsenides. Therefore, the component 2 indicates water circulation for carbonate rocks and particularly by the formation called Tamaulipas which is enriched with sulfides or sulfoarsenides [13,26].

The binary diagram for the components 1 and 3 is shown in Fig. 7. In this diagram, three groupings of water samples are differentiated. The first group is located in the sector to negative values at the bottom left of the diagram. This group consists of 47% of the evaluated points and corresponds to samples associated with carbonate rocks. Added to this, samples with negative values in the component 3 were associated with volcanic rocks.

In contrast, the second group of samples can be considered more dispersed. This group presented positive values in the component 3 with developments in mineralization from highest to lowest (the component 3 is dependent on the concentrations of $HAsO_4^{2-}$,

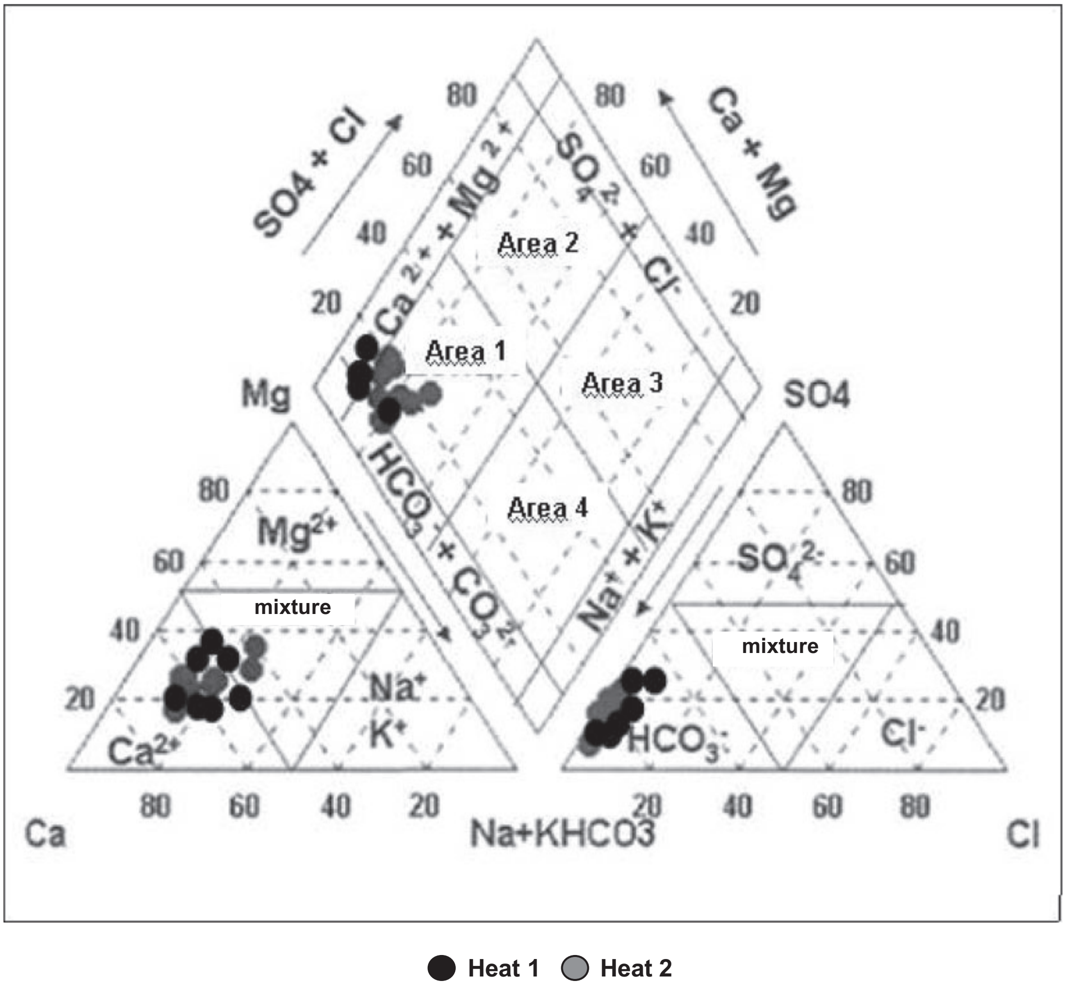

Fig. 5. Piper diagram showing the chemical species (anions and cations) found in the study area.

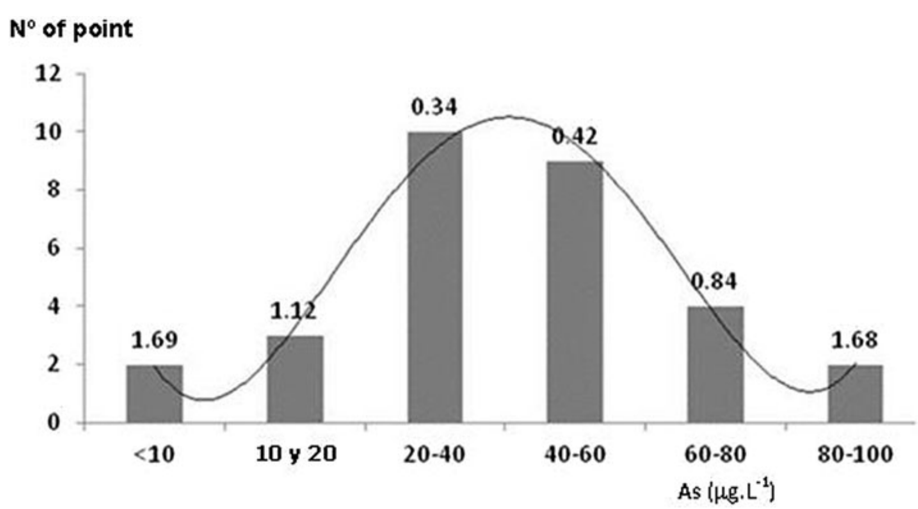

Fig. 6. Histogram of distribution of values of total As concentrations in the sampled points. Shows the standard deviation values.

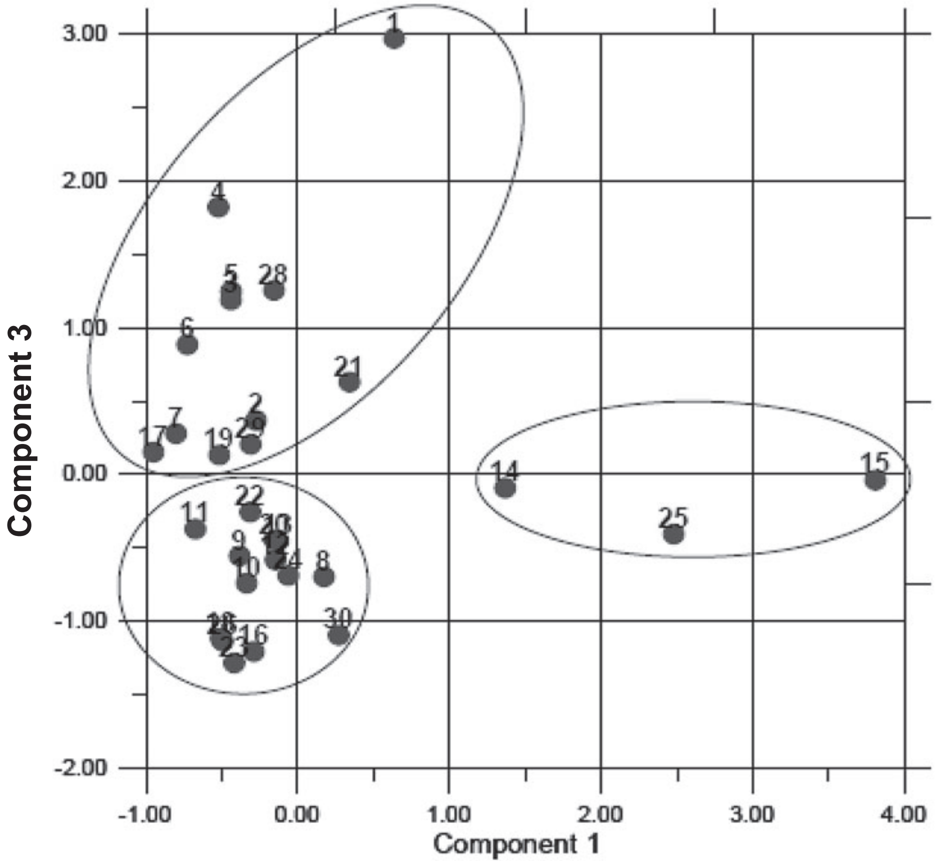

Fig. 7. PCA representation of the samples analyzed under the criterion of projection of the factor loadings of the components 1 (input element concentrations providing hardness: SO_4^{2-}, Ca^{2+} and Mg^{2+}) and 3 (input element concentrations that provide salinity: HCO_3^-, $Cl^-$and NO_3^-).

HCO_3^-, Cl^-, and NO_3^-), and correspond to volcanic lithologies. On the other hand, samples with higher values correspond to samples contaminated by anthropogenic activities. In this sense, samples with Cl^- and NO_3^- contents indicate contamination by sewage and leaching of agricultural land, respectively [4,7,23,31].

The third group includes the results of the points 14, 15, and 25 in which the component 1 is high, and corresponds to highly mineralized waters due to the presence of HCO_3^-. In addition, these samples showed high As contents and low contents of Fe and Pb. This may be related to leachate from mine tailings that have been accumulated for several years. Furthermore, the high contents of SO_4^{2-} may indicate the oxidation of sulfides and release large amounts of As. In this sense, the literature indicates that high contents of SO_4^{2-} are due to sulfide oxidation processes (water samples near mine tailings) and the contact of water with gypsum (Barron area, in area B in Fig. 1).

Arsenic results of the samples of the aquifer system of Zimapan presented concentrations of medium (≈0.010–0.025 mg L^{-1}) to high (>0.025 mg L^{-1}) and markedly heterogeneous distribution as shown in Fig. 8. As shown, the trends for arsenic are increased in the wells marked with the numbers 2, 8, 9, 10, 11, 13, 14, and 18. Some of the wells are contaminated by mining activities (anthropogenic source) or by mineralization processes and marine sedimentary rocks (natural sources). Considering the value of 0.025 mg L^{-1}, arsenic concentrations of these wells are outside the permissible limits established by the standard. Therefore, the well 8 (El Muhi) was definitively closed since 1998 [32].

Some of elevated arsenic values correspond to water samples of wells exploited and unexploited (used only for monitoring). The relative increases in heavy metals indicated that mechanisms that facilitate their incorporation into the groundwater flow occur. The presence of As can be correlated with mining activities, areas of mineralization, foundry slag, smelters, and mine tailings [2,8–12,14,23,25,32–36]. The largest source is geomorphological origin. This is supported by Ongley et al. study [25] which reveals that high concentrations of As in groundwater are

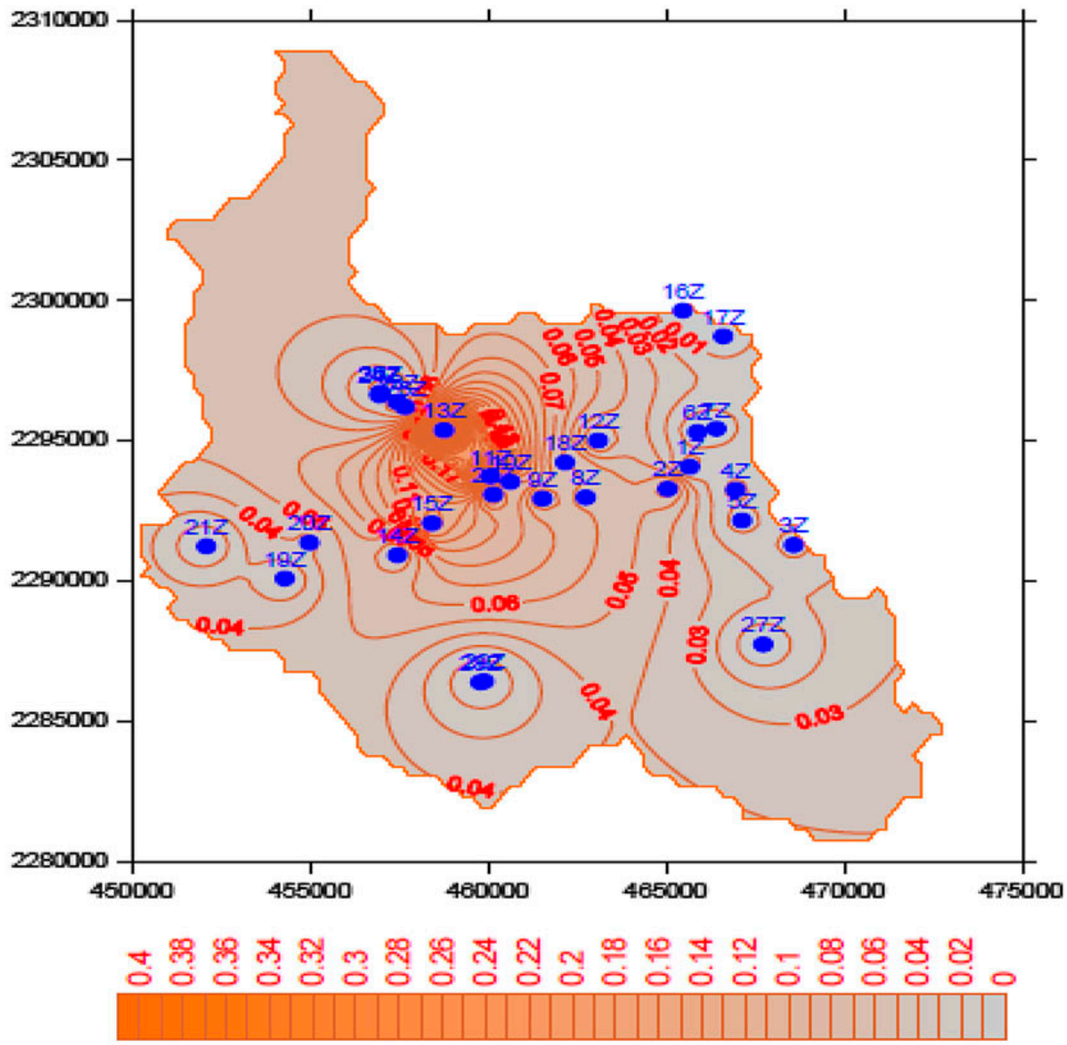

Fig. 8. Map of spatial distribution of arsenic (ppm) of the analyzed water.

related to the composition of the natural solution of the aquifer matrix and the proximity to mine tailings; however, anthropogenic effects are not reported. Arsenic concentrations of 0.1 mg L^{-1} were found in 22 samples, which is equivalent to 44% of total samples; 19 of these had concentrations greater than 0.05 mg L^{-1}. In wells located east of Zimapan high arsenic concentrations were not observed (see Fig. 8). Drinking water of the municipal water network in Zimapan showed an average concentration of arsenic of 0.385 mg L^{-1} which is 40 times the normative value.

4. Conclusion

In general, groundwater aquifer system in Zimapan showed conductivity levels between 198 and 974 μS cm^{-1}. By contrast, the water from contaminated wells showed high conductivity levels between 1,485 and 1,700 μS cm^{-1}. These values allow classifying the water of this region as water rich in calcium, magnesium, and bicarbonate.

Water samples from the study area have a neutral pH and alkalinity tendency. Arsenic concentrations were low in the volcanic aquifer, and high in the aquifer with carbonate rocks. The average total arsenic concentration in water samples was 47.8 μg L^{-1} which is 4.8 times the normative value. Ninety-three percentage of the sampled points showed arsenic concentrations ranging from 20–100 μg L^{-1}, 20% of the total had concentrations greater than 50 μg L^{-1}, and 47% of the evaluated points correspond to samples associated with carbonate rocks. This may be associated with leachate from mine tailings that have been accumulated for several years. Furthermore, the high contents of

SO_4^{2-} in groundwater may indicate the oxidation of sulfides and release large amounts of arsenic.

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