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Artículo Científico

Temporal evolution of nitrate in Meoqui-Delicias aquifer in Chihuahua, Mexico

Evolución temporal de nitrato en el acuífero Meoqui-Delicias en Chihuahua, México

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Abstract

The continued input of nitrate (NO_3) into groundwater is a global problem, mainly associated to excess fertilizer and improper disposal of human and livestock waste. Nitrate accumulation in oxic aquifers of semiarid areas makes these zones especially susceptible to pollution. Nitrate in Meoqui-Delicias aquifer, located in an important irrigation district in Chihuahua, Mexico, was quantified in 2021 in 63 drinking water wells. Samples collected were analyzed in laboratory and results were compared to 2003 and 2006 data available for those wells. Nitrate values varied from 0.7 to 23.2 mg/L and 22 % of the wells contained NO_3 above drinking water guidelines (10 mg NO_3-N/L). A low to moderate nitrate pollution index (NPI) and a slight NO_3-N variation with time was observed for most wells. Values of NO_3-N/Cl < 1.0 support an anthropogenic origin of nitrate. No association was found between NO_3-N and well depth. The most susceptible areas to nitrate contamination were identified as those areas with high NO_3-N and increasing concentration with time. The lack of a pattern of contamination suggested leakage of manure leachate at a few points as the most likely contamination source. The consistently high NO_3-N content (>10 mg/L) in three deep wells constitutes a serious concern.

Keywords: **Nitrate pollution, irrigation, semiarid, water quality, manuere leachate, Chihuahua**

Resumen

La aportación continua de nitrato (NO_3) en aguas subterráneas es un problema mundial, asociado principalmente al exceso de fertilizantes y a la disposición inadecuada de desechos humanos y ganaderos. En 2021 se cuantificó el nitrato en 63 pozos de agua potable del acuífero Meoqui-Delicias, ubicado en un importante distrito de riego en Chihuahua, México. Las muestras colectadas fueron analizadas en el laboratorio y los resultados se compararon con datos de 2003 y 2006 de los mismos pozos. Los nitratos variaron de 0.7 a 23.2 mg/L; el 22 % de los pozos sobrepasaron la norma (10 mg de NO_3-N/L). Se observó un índice de contaminación por nitrato entre bajo y moderado y una ligera variación del NO_3-N con el tiempo para la mayoría de los pozos. Valores de NO_3-N/Cl < 1.0 sugieren origen antropogénico del nitrato. No se encontró relación entre NO_3-N y la profundidad del pozo. Las áreas de alto contenido de NO_3-N y concentración creciente con el tiempo son las más susceptibles a la contaminación. La falta de un patrón de contaminación sugiere que los residuos ganaderos constituyen la fuente probable de contaminación en algunos puntos. El constante y alto contenido de NO_3-N (>10 mg/L) en tres pozos profundos constituye una preocupación.

Palabras clave: **Contaminación por nitratos, irrigación, semiárida, calidad del agua, residuos ganaderos, Chihuahua.**

1. Introduction

The presence of groundwater nitrate (NO_3) has been recognized as a global problem because of its gradual increase in aquifers around the world (Galloway *et al.*, 2008), its capacity to degrade drinking water quality, and its ability to cause eutrophication to surface water bodies and to the ocean (Diaz and Rosenberg, 2008; Hamlin *et al*., 2022). The main source of NO_3 in groundwater has been traced back to an excess of N-fertilizer applied to crops, either in the form of mineral N-fertilizer or as manure (Galloway *et al*., 2008; Vitousek *et al*., 2009; Bijay and Craswell, 2021). Fertilizers are often applied to crops in excessive amounts or out of the appropriate season (Gomes *et al*., 2023).

Nitrate (NO_3) is a highly stable and soluble ion under the oxic and alkaline conditions that prevail in aquifers in arid and semiarid areas (Re *et al*., 2021). Under these conditions, NO_3 barely adsorbs or precipitates, and thus accumulates in aquifers (Hansen *et al*., 2017; Xiao *et al*., 2022) where it may remain in dissolved form for a long time (Gutiérrez *et al*., 2018; Mateo-Sagasta and Albers, 2018). In arid and semiarid regions that have intensive agriculture and livestock farming a high content of NO_3 is thus expected.

Since water quality of open aquifers under intense agricultural and livestock land use varies with time, time series statistical methods have been a useful tool to determine and map its evolution and to confirm statistically significant trends (Ducci *et al*., 2020; Hamlin *et al*., 2022). These methods require a minimum of 8 to 10 measurements (e.g., years), an asset that many semiarid areas may not have. Therefore, and in absence of a better alternative, information about the spatial NO_3 concentrations and a rough estimation of their variation with time can be used to strategize water management in agricultural and livestock areas lacking proper monitoring (Li *et al*., 2021), at least until more data are gathered.

Groundwater is the major source of drinking water in arid and semiarid areas. The gradual increase of NO_3 in the groundwater is an adverse finding since chronic ingestion of water containing high levels of NO_3 is harmful to humans, especially to infants under 6 months, who may develop methemoglobinemia or cyanosis (Panneerselvam *et al*., 2020; Hamlin *et al*., 2022). The WHO limit for drinking water is 10 mg NO_3-N/L; however, threshold values lower than 10 mg NO_3-N/L have been proposed since human and environmental health complications starting at lower concentrations after prolonged exposure, including 4.5 mg NO_3-N/L (Panneerselvam *et al*., 2020) and 2 mg NO_3-N/L (Hamlin *et al*., 2022). Environmental impacts of NO_3 include a major role in stimulating phytoplankton growth, which may result in eutrophication and the development of hypoxic zones in oceans (Diaz and Rosenberg, 2008). To curb these negative effects on the environment, a threshold concentration of 3 mg NO_3-N/L has been proposed (USEPA, 2007).

The N-cycle is complex, which makes the origin of NO_3 difficult to pinpoint because of the many possible natural or anthropogenic sources. Natural sources include atmospheric deposition and dissolution of N-containing minerals. For most aquifers, these sources combined amount to a minor contribution of the NO_3 compared with its anthropogenic sources. As a result, NO_3 is considered an anthropogenic contaminant associated with an excess of N-fertilizer applied in agriculture (either mineral fertilizer or manure) and domestic wastewater effluents (raw or treated wastewater and septic tanks) (Mateo-Sagasta and Albers, 2018; Gutiérrez *et al*., 2022). A notable exception includes arid regions where natural NO_3 deposits form in the subsurface of arroyo floodplains (Walvoord et al., 2003; Linhoff, 2022). These natural deposits have a ratio NO_3-N/Cl > 1.5 and $\delta^{15}N$ (NO_3) < 8 ‰. (Linhoff, 2022). These deposits have been observed in several desert areas around the world, including parts of the Chihuahuan Desert (Walvoord *et al*., 2003).

Besides contributing to aquifer contamination and eutrophication of surface waters, NO_3 from agricultural non-point sources represents a loss of fertilizer resource and loss of profit to the farmer. Therefore, management techniques that would minimize these losses are constantly sought (Rudolph *et al*., 2015; Hansen *et al*., 2017; Sapkota *et al*., 2020). Another N compound generated in irrigated agricultural fields is nitrous oxide (N_2O), a potent greenhouse gas that is cumbersome to measure and thus scarcely reported (Sapkota *et al*. 2020). However, the amount of N that becomes N_2O is generally a small fraction of the amount of NO_3 formed (Millar *et al*., 2018).

If the content of NO_3 and other N-compounds in surface waters and soil is high, plants will absorb the amount they need, a small part will become gases, and most of the remaining N-compounds will infiltrate through the soil and into the underlying aquifer. The infiltration rate is a complex process that depends on many factors, including the soil type, amount of nitrogen in soils, recharge of the aquifer, and depth to water table. Nitrate leakage into groundwater is faster and easier through soils of high permeability (Gomes *et al.,* 2023). Because of their high solubility, NO_3 is easily carried by infiltration water and becomes a common contaminant in oxic aquifers underlying agricultural and livestock areas (Gutiérrez *et al.,* 2018). An economically sustainable N-management is needed to prevent NO_3 leakage to groundwater (Rudolph *et al*., 2015; Gutiérrez *et al*., 2018; Li *et al*., 2021; Hamlin *et al.,* 2022). Mechanisms that would naturally remove nitrate from the soil, surface water reservoirs, and aquifers, include denitrification and ammonification, both occurring under reduced conditions (Tesoriero *et al*., 2021; Gutiérrez *et al.,* 2022). Among the most promising methods for reducing loss of nitrogen in irrigated arid areas are the synchronization of fertilization to coincide with rapid plant growth, slowly released fertilizer, and nitrification inhibition. For example, sensor technology can match the application of fertilizer to the nutritional needs of the plant during the different growth stages, and inhibitors of biological nitrification are commercially available (Norton and Ouyang, 2019; Drazic *et al*., 2020). However, these novel methods may take years to become an across-the-board procedure used by farmers.

The Meoqui-Delicias aquifer in northern Mexico underlies an irrigated agricultural area within the Chihuahuan Desert. Like other semiarid agricultural areas (Mukherjee and Singh, 2021; Re *et al.*, 2021; Liu *et al*., 2022), high levels of nitrate (> 10 mg/L NO_3-N) in wells within this aquifer have been reported (Espino *et al*., 2007; Barrera, 2008; Rascón, 2011). Despite this, groundwater quality studies of this region are scarce, and a temporal analysis has not been reported for this region, nor the distribution of N-species (NO_3, N_2O, NH_4, N_{org}).

Although an upward trend in NO_3 concentration is expected in this region due to its semiarid climate, oxidizing conditions in the aquifer, and ongoing intense agriculture, this assumption needs to be confirmed. The objectives of this study were to: (1) update the spatial distribution of NO_3 concentrations for 2021 throughout the aquifer using data from 63 wells; (2) compare the 2021 NO_3 values with those reported for these same wells in 2003 and 2006 to determine the variation with time for each well and their possible source, and (3) plot the spatial distribution of NO_3 variation to identify the most affected and at-risk areas.

2. Materials and methods

2.1 Description of study area

The Meoqui-Delicias aquifer underlies an irrigated agricultural region known as Distrito de Riego 005 in northern Mexico (Fig. 1). The climate is semiarid, with an average annual precipitation of 284 mm. The Meoqui-Delicias aquifer occupies a surface area of 4,830 km^2, has a maximum thickness of about 500 m and an average thickness of 300 m. The aquifer is composed of alluvial fill, a material of medium to high permeability, which is mainly recharged at alluvial fans present at the base of the hills rising on its western and eastern parts. This is primarily an open aquifer, but under clay lenses it operates as a confined aquifer (Villalobos-Gutiérrez, 2021).

Recharge in the aquifer occurs naturally and induced. In the first case, rainwater is collected directly in the valley area and through underground horizontal flows coming from the surrounding foothill areas. The induced recharge comes from the infiltration of surplus irrigation water, which contains both groundwater and surface water (CONAGUA, 2020). This component in the groundwater balance represents an important contribution to the vertical flow that allows for the introduction of solutes to the unsaturated and saturated zones of the aquifer.

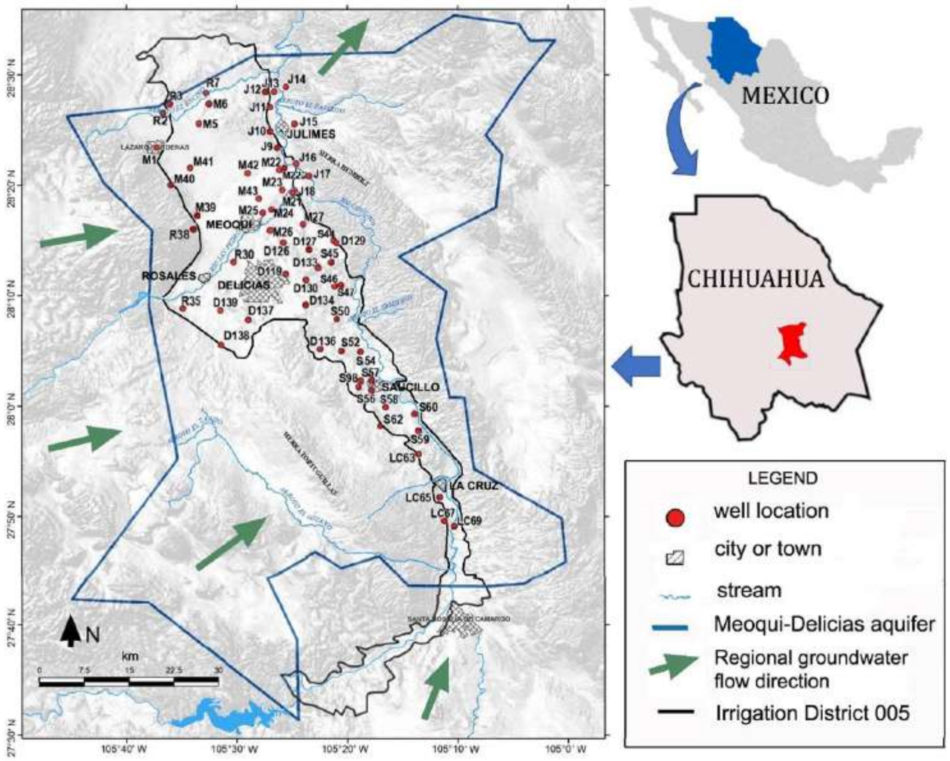

Figure 1. Location of the Meoqui-Delicias aquifer (blue line), Irrigation District 005 (black line) and sampled wells (red dots).

Figura 1. Localización del acuífero Meoqui-Delicias (línea azul), Distrito de Riego 005 (línea negra) y pozos muestreados (puntos rojo).

The irrigated area grows summer and winter crops. Summer crops include mainly alfalfa, pecan nut and jalapeño pepper, and to a lesser extent onions, sorghum, and cotton. Winter crops consist of mainly forage grasses. Pecan nut has gained relevance in the region, for which its production has increased significantly in recent years. This crop requires basic fertilizers or macroelements, such as nitrogen (N), phosphorus (P) and potassium (K) to improve yield levels and product quality. Pecan producing orchards are applied 80 to 100 kg of nitrogen per hectare; a large quantity considering that nitrogen applications by gravity irrigation incur in losses between 30 and 45 % of the fertilizer (INIFAP, 2013). Both pecan nut and jalapeño peppers receive large amounts of nitrogen fertilizers, which are applied through irrigation. Some common choices of fertilizer include organic fertilizers as urea and manure, but also a wide variety of mineral fertilizers such as potassium nitrate, potassium phosphonitrate, ammonium nitrate and ammonium sulfate (INIFAP, 2015).

Alfalfa is a crop that, through the symbiotic relationship with atmospheric nitrogen-fixing bacteria of the genus *Rhizobium,* produces its own nitrogen compounds. However, significant amounts of organic fertilizers are applied in the area to improve soil quality and to retain moisture, as well as ammonium phosphate to improve productivity (Lara and Jurado, 2014).

This area is also an important dairy producer nation-wide, with more than 100 farms housing over 72,000 dairy cows (Rivas *et al*., 2008, 2018). The dairy farms are scattered within the irrigation district DR-005, mainly in the municipalities of Delicias, Rosales, Meoqui and Saucillo, in the central part of the aquifer. Among these, there are large dairy cattle farms in which advanced technology is applied for the handling of livestock and dairy. However, small dairy farming operations are common as well (Barrera, 2008). The wastes generated by dairy farms are used to irrigate nearby fields (Rivas-Lucero *et al*., 2018), which allows us to assume that large amounts of solid and liquid waste with high nitrogen content are spread over the fields, and that a part of it may leach into the groundwater (Rivas-Lucero *et al.*, 2018).

In addition, domestic sewage was mostly discharged into streams with limited to no treatment. In this regard, a previous study by Espino *et al*. (2007) found that 34 % of 134 wells in this area contained high NO_3 concentration. A NO_3-N source apportionment (manure, inorganic fertilizer, sewage) study was conducted by Espino *et al*. (2011) using N-15 isotope analyses of 39 groundwater samples corresponding to a small area of the study region. The results showed that 15 wells (38 % of total) surpassed the drinking water limit of 10 mg/L NO_3-N. Isotope analyses of 19 samples, including urban and rural wells, found that mineral fertilizer was the source for NO_3 in 11 % of the wells, sewage, and manure in 52 % of the wells, and a combination of fertilizer and sewage and manure in 37 % of the wells. In rural areas, the source was a mixture of fertilizer residues, whereas in urban areas, the source for 90 % of the analyzed wells was organic wastes.

Based on the results obtained by Barrera (2008) and Rascón (2011), groundwater is primarily of Na-Ca-SO_4-HCO_3 type. A Piper diagram (Fig. 2) shows the water composition of representative wells that are classified according to their location within the aquifer. Only 25 out of the 63 wells are included in Fig. 2 to allow a better visualization.

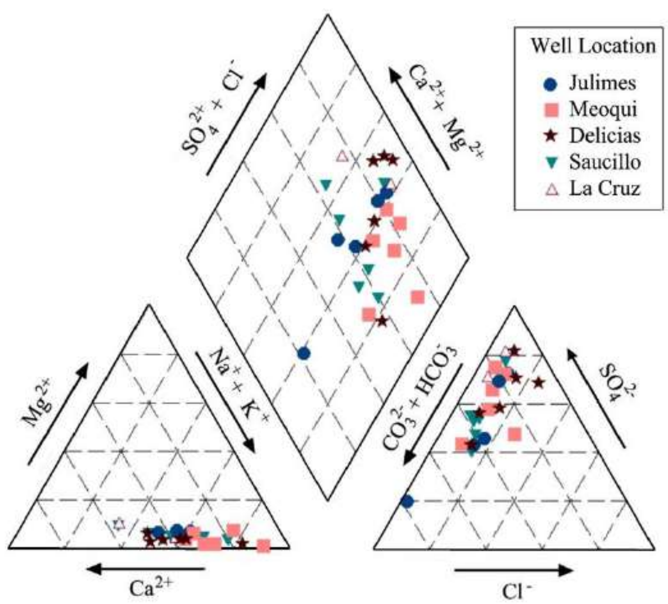

Figure 2. Piper diagram showing major ion composition of groundwater according to regions within the Meoqui-Delicias aquifer.
Figura 2. Diagrama de Piper que muestra la composición de iones mayores del agua subterránea por regiones en el acuífero Meoqui-Delicias.

2.2 Sampling and analysis

Groundwater from 63 wells were sampled from December 2020 to January 2021. The wells were selected to match those previously studied by Espino *et al*. (2007), which had been originally selected as to provide a good coverage of the area. Temperature, pH, and electrical conductivity were determined *in situ* using a portable multi-parameter probe HANNA HI 9828 equipment. The TDS values were obtained by direct conversion in this equipment based on the linear relationship between both parameters. Samples were collected in 250 ml clean plastic bottles and kept cool during their transport to the laboratory. Once in the laboratory, NO_3-N concentrations were measured with a Hach DR/890 colorimeter using the cadmium reduction method. In this method, NO_3 is reduced to nitrite (NO_2) and measured together as NO_3+NO_2 (Villalobos-Gutiérrez, 2021). Due to the oxidizing conditions in this and adjacent aquifers (Mahlknecht *et al*., 2008; Reyes-Gómez *et al*., 2015; CONAGUA, 2022), the amount of NO_2 is about two orders of magnitude smaller than NO_3, for which (NO_3+NO_2) can be approximated to NO_3. Chloride concentrations were determined by tritation using the argentometic method (Secretaría de Economía, 2001). Blanks of three-distillated water were used for equipment calibration and at least one out of every 10 samples were run in duplicate.

2.3 Data processing

The locations of the wells were plotted using ArcMap and nitrate isoconcentration lines were constructed at 2 mg/L NO_3-N interval for the 2021 data to show the spatial distribution of NO_3-N concentrations. To observe the evolution of this parameter over time, results obtained in the sampling campaign carried out in January 2021 (2021 data) were compared with values reported by the Comisión Nacional del Agua (CONAGUA) in 2003 and with samples collected in 2006 reported by Barrera (2008) and Rascón (2011) for the same wells.

For temporal variations, the total number of samples was reduced from 63 to 60 after removing wells missing a measurement. Each well was classified into one of five possible groups based on the slope of the line obtained in the regression analysis applied to the variation in nitrate concentration with respect to time: increase, minor increase, no change, minor decrease, and decrease. The small number of available measurements (3) precluded a formal calculation of a trend, which requires a minimum of eight measurements (Ducci *et al*., 2020). Therefore, a simplification was devised to provide an estimation rather than a calculation of a concentration trend based on the regression coefficient of the best-fitting line of NO_3-N versus time graphs. The slope of its regression line was used as an indicator value of the trend in the variation of the concentration.

2.4 Nitrate Pollution Index

The Nitrate Pollution Index (NPI) for 2021 data was calculated according to the formula below (Obeidat *et al.,* 2012):

$$\text{NPI} = \frac{C - HAV}{HAV} \tag{1}$$

where C is the analytical concentration of nitrate in the sample and HAV is the threshold value of anthropogenic source (human affected value) taken as 4.51 mg/L NO_3-N (20 mg/L NO_3).

3. Results and discussion

The water quality results of the 2021 sampling campaign are listed in Table 1. Wells are labeled with a letter corresponding to the nearest town (D for Delicias, J for Julimes, LC for La Cruz, M for Meoqui, R for Rosales and S for Saucillo) followed by a number.

Nitrate concentrations varied between 0.7 and 23.2 mg/L NO_3-N and had an average of 7.5 mg/L NO_3-N. Most of the wells (62 %) had a modest nitrate content, 22 % of the wells surpassed the WHO guideline of 10 mg/L NO_3-N, and 16 % had low values (< 3 mg/L NO_3-N). These results agree with the obtained NPI results, which show that the sampled wells cover all levels of pollution and overall has a moderate NO_3 pollution, with 33.3 % of the wells having moderate to very significant NO_3-N concentrations. The NPI results are listed in Table 2. Another important observation from data in Table 1 is the NO_3-N /Cl ratio < 1 in all cases, which according to Linhoff (2022) relates nitrate with an anthropogenic origin derived from human and animal manure.

Table 1. Water quality data of wells in the 2021 sampling campaign
Tabla 1. Datos de calidad del agua de los pozos en la campaña de muestreo 2021

Well	T °C	pH	TDS mg/L	NO_3-N mg/L	Cl mg/L	NO_3-N/Cl	Well	T °C	pH	TDS mg/L	NO_3-N mg/L	Cl mg/L	NO_3-N/Cl
D119	18.2	7.1	1128	11.5	50	0.34	M23	18.7	8.0	690	17.0	52	0.27
D126	21.3	7.3	775	11.8	40	0.27	M24	21.9	7.4	539	11.4	36	0.15
D127	23.5	7.4	1333	8.0	221	0.05	M25	22.0	7.3	604	10.4	27	0.32
D129	26.1	6.4	784	23.2	66	0.23	M26	27.2	7.5	355	11.2	n.d.	n.d.
D130	18.2	7.3	2059	10.8	363	0.03	M27	27.7	7.1	675	15.8	n.d.	n.d.
D133	24.0	6.7	1307	20.1	87	0.13	M39	10.8	7.9	383	4.6	8	0.89
D134	24.7	7.5	862	9.8	n.d.	n.d.	M40	21.8	7.0	398	4.9	19	0.21
D136	25.7	7.2	863	7.0	n.d.	n.d.	M41	17.5	7.4	434	7.0	26	0.22
D137	20.0	7.3	519	5.9	8	0.93	M42	22.3	7.5	618	9.0	49	0.10
D138	21.4	6.0	1057	15.0	83	0.08	M43	24.3	7.4	727	5.1	46	0.12
D139	25.0	7.7	520	4.4	33	0.53	R2	22.4	7.4	587	5.4	59	0.20
J9	18.3	8.3	1056	0.7	149	0.03	R3	17.5	7.5	429	4.2	31	0.38

J10	22.9	7.1	1587	1.0	273	0.03	R7	24.3	8.1	490	2.0	46	0.05
J11	23.1	7.5	1031	1.4	118	0.02	R30	15.9	8.2	418	1.4	19	0.51
J12	22.3	7.1	1429	2.6	108	0.04	R35	23.1	6.9	951	7.9	78	0.04
J13	22.2	7.1	1429	2.7	101	0.06	R38	12.8	7.6	330	2.5	11	0.22
J14	19.0	7.3	1221	4.4	85	0.07	S44	19.9	7.6	583	5.2	42	0.26
J15	23.5	7.0	1193	2.4	106	0.09	S45	21.1	7.8	575	7.8	23	0.30
J16	19.8	7.4	890	2.2	52	0.03	S46	21.2	7.0	817	17.8	n.d.	n.d.
J17	13.7	7.5	720	1.8	78	0.04	S47	22.4	7.0	735	7.3	n.d.	n.d.
J18	25.5	7.2	840	5.2	66	0.05	S50	23.0	7.5	581	13.4	n.d.	n.d.
LC63	18.6	7.3	762	6.6	101	0.08	S52	23.0	7.6	755	19.8	n.d.	n.d.
LC65	21.9	7.8	914	3.5	n.d.	n.d.	S54	21.0	7.3	786	7.2	n.d.	n.d.
LC67	21.6	7.2	739	5.2	33	0.28	S56	24.6	6.7	834	9.0	n.d.	n.d.
LC69	25.1	7.4	884	6.2	50	0.18	S562	27.0	7.2	843	10.9	n.d.	n.d.
M1	20.9	7.3	401	4.6	31	0.17	S57	24.9	7.6	456	4.5	n.d.	n.d.
M5	24.5	8.3	469	3.6	32	0.59	S58	15.4	7.2	1020	7.2	47	0.29
M6	18.7	7.9	953	11.9	113	0.11	S59	22.4	7.6	444	5.9	30	0.44
M19	14.1	8.0	701	8.9	n.d.	n.d.	S60	22.4	7.3	454	7.3	27	0.27
M21	22.1	7.2	739	4.2	52	0.27	S62	23.4	7.4	793	6.6	40	0.16
M22	25.2	7.5	842	3.4	36	0.15	S98	22.2	7.2	668	9.6	n.d.	n.d.
M22-2	13.5	7.2	1000	2.5	27	0.33							

n.d. = not detected

Table 2. Nitrate pollution index (NPI) classification of groundwater (Obeidat *et al.*, 2012) for 2021 samples
Tabla 2. Clasificación del agua subterránea de acuerdo con el Índice de Contaminación por Nitrato (NPI) (Obeidat *et al.*, 2012) para las muestras 2021

NPI value	Degree of pollution	No. wells	% wells
< 0	Clean	17	27.0
0 - 1	Light	25	39.7
1 - 2	Moderate	13	20.6
2 - 3	Significant	5	7.9
> 3	Very significant	3	4.8

The nitrate concentration values reported for these wells in 2003 and 2006 were added to the 2021 data in orden to compare the changes of NO_3-N concentrations with time. The data are listed in Table 3 and their spatial distribution in Figure 3, with shaded areas corresponding to high NO_3-N concentrations. After visually comparing the three maps (Fig. 3), one can see that the high NO_3-N concentrations areas shift with time but there are some areas with persistent high concentrations.

Table 3 Nitrate values (in mg/L NO_3-N) of studied wells in 2003, 2006 and 2021
Tabla 3 Valores de nitrato (en mg NO_3-N/L) de los pozos estudiados en 2003, 2006 y 2021

Well	Depth m	NO_3-N 2003	NO_3-N 2006	NO_3-N 2021	Well	Depth m	NO_3-N 2003	NO_3-N 2006	NO_3-N 2021
D119	-	17.2	12.5	11.5	M23	-	13.9	9.1	17.0
D126	-	10.7	9.4	11.8	M24	36	5.4	7.4	11.4
D127	-	10.6	5.9	8.0	M25	-	8.8	10.1	10.4
D129	-	14.9	12.1	23.2	M26	150	10.1	9.5	11.2
D130	320	12.1	12.5	10.8	M27	150	11.8	15.5	15.8
D133	-	11.4	14.8	20.1	M39	-	6.7	5.4	4.6
D134	-	5.3	8.3	9.8	M40	150	4.0	4.8	4.9
D136	200	10.2	9.6	7.0	M41	-	5.8	6.4	7.0
D137	-	7.1	10.0	5.9	M42	-	4.8	3.4	9.0
D138	-	6.8	8.9	15.0	M43	-	5.3	6.3	5.1
D139	70	17.7	20.0	4.4	R2	152	11.9	10.7	5.4
J9	-	4.2	3.9	0.7	R3	-	11.9	6.3	4.2
J10	-	8.5	2.5	1.0	R7	-	2.5	4.6	2.0
J11	-	1.9	1.9	1.4	R30	-	9.6	1.5	1.4
J12	-	4.2	5.1	2.6	R35	-	3.2	3.2	7.9
J13	-	5.8	4.2	2.7	R38	-	2.5	1.4	2.5
J14	-	6.2	4.9	4.4	S44	-	10.8	4.5	5.2
J15	79	9.5	4.0	2.4	S45	-	7.0	12.3	7.8
J16	15	1.3	2.2	2.2	S46	-	34.3	25.3	17.8
J17	-	3.3	4.0	1.8	S47	90	11.8	12.6	7.3
J18	-	3.1	1.9	5.2	S50	-	8.9	8.5	13.4
LC63	-	8.2	9.9	6.6	S52	-	7.0	5.9	19.8
LC67	-	9.2	4.2	5.2	S54	60	6.5	11.3	7.2
LC69	-	9.0	6.4	6.2	S56	185	--	8.1	9.0
M1	-	5.1	6.6	4.6	S57	180	13.0	5.6	4.5
M5	-	19.0	13.4	3.6	S58	250	13.6	13.6	7.2
M6	181	12.3	14.8	11.9	S59	-	13.0	12.0	5.9
M19	60	8.3	5.4	8.9	S60	-	7.4	6.9	7.3
M21	-	7.8	6.4	4.2	S62	-	6.5	7.0	6.6
M22	-	12.5	3.6	3.4	S98	137	4.5	4.5	9.6

Except for two locations where the pH values were 6.0 and 6.4 (Table 1), the pH values were within the drinking water limit of 6.5-8.5 set by the World Health Organization, favoring the alkaline condition. The pH values below 7.0 likely result from the infiltration of leachate from dairy farm wastes, which have an acidic pH (Rivas-Lucero *et al.*, 2018). In contrast, TDS values seemed to associate with irrigation that extends to about 30 km radius, in localities water enriched with salts after evaporation or from being in contact with salt-rich soils. TDS values exceeded the WHO norm (1,000 mg/L) in twelve wells spread over two main areas, one at the center of the aquifer (Meoqui and Delicias municipalities) and the other in its discharge area northeast corner of the aquifer, where the Rio Conchos receives irrigation return flows (Julimes municipality). In contrast, the lowest TDS values, around 400 mg/L, were observed in wells located near the recharge areas in the western part of the aquifer.

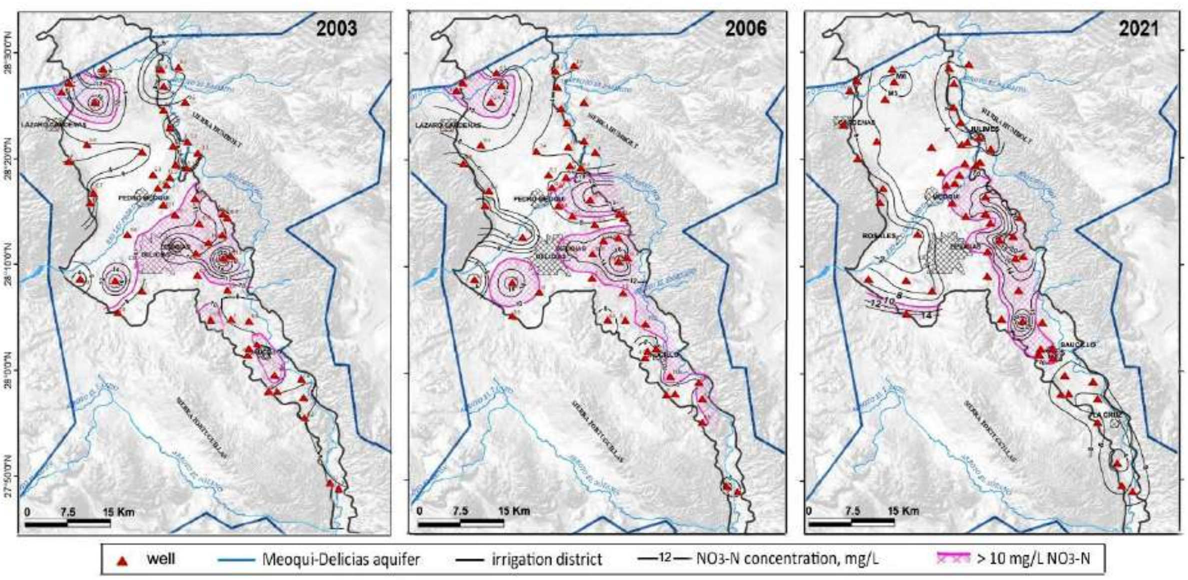

Figure 3. Evolution of NO_3-N concentrations in 2003 to 2021. Zones >10 mg/L NO_3-N shaded in pink.
Figura 3. Evolución de las concentraciones de NO_3-N en 2003 a 2021. Las zonas con NO_3-N >10 mg/L están sombreadas en rosa.

The moderate NO_3-N values for most samples and a ratio of NO_3-N/Cl < 1 in all samples (Table 1) support an anthropogenic origin of nitrate, and thus a negligible presence of natural subsoil NO_3-N deposits (Linhoff, 2022). The spatial distribution of 2021 NO_3-N concentrations in Fig. 3 shows the highest concentrations in the eastern part of Delicias, spreading over an area that belong to the municipalities of Delicias, Meoqui and Saucillo. Important areas of cultivation and large dairy farms are also located there. In addition, sources of pollution also include inadequately treated domestic waste from rural communities, such as septic tanks and other waste treatment systems.

At this point in time (2021 data), having one third of the wells in the categories of moderately to highly polluted, it seems crucial to find out if this is a relatively stable behavior or if the aquifer is borderline to worsening. Thus, NO_3-N data reported for 2003 and 2006 were added (Table 3) and plotted in maps displayed next to each other for comparison purposes (Fig. 3). To ease the comparison, areas where NO_3-N >10 mg/L were shaded in pink. Comparison among these maps shows that the high NO_3-N concentrations vary with time. One shaded area (NO_3-N >10 mg/L) located in the center of the aquifer persists through the years, extending south towards Saucillo in

2021, and encompassing wells from Delicias, and Saucillo municipalities. Interestingly, two areas of high NO_3-N concentrations in the north and western part of the aquifer decreased in concentration in 2021.

A quantitative approach was then necessary to denote their variation with time. A first glance to the data in Table 3 indicates little variation (1 to 10 mg NO_3-N/L) in NO_3-N concentrations for most wells between 2003 and 2021. The linear regression analysis of NO_3-N concentrations for the years 2003, 2006 and 2021 shown in Table 3 was used to determine the trend in variation over time for each well, based on the slope of their regression line (R). A visual inspection of the range of R and the slope indicated that some wells had an imperceptible change with time while others had a well-delineated change (either increase or decrease). Once those two were separated, adding an intermediate range was necessary to include all data. The selected five categories and the distribution of wells into each of these categories are listed in Table 4.

To help distinguish which wells increased their NO_3 content, they were separated by code letter (municipality). Once this was done, D-wells (Delicias) were more abundant in the increase category and relatively absent in the decrease category, a behavior that was closely followed by the M (Meoqui) wells. In total, an increase behavior was observed in 25 % of the wells. On the other hand, 45 % of the wells showed a decrease or minimum decrease, among which wells from Julimes and some of Saucillo stood out. Regarding the increased number of wells whose nitrate concentration was lower in 2021, it is important to point out that these samples were collected during the winter, which is a time of low agricultural activity, unlike the samples of previous years collected in summer. On the other hand, the climatic factor does not affect milk production activity, which justifies the observed increase in nitrates in wells located adjacent to the largest dairy farms.

The overall results show that many of the D-, M- and S- wells (located in the Delicias, Meoqui, and Saucillo municipalities, respectively) contain high concentration of NO_3-N and identifies this area is a vulnerable part of the aquifer. The nine most affected wells were M23 (Loreto), M24 (Puentes), M27 (Nuevo Loreto), D129 (La Merced), D133 (Virginias), D138 (Nicolas Bravo), S46 (Santa Rosa), S50 (Altamirano) and S52 (Madero). Although not yet affected in the same proportion, other municipalities that have the same alluvial fill (e.g., Julimes, La Cruz, and Rosales) may become affected in the future if the amount of N-fertilizers, manure and other animal and human wastes were to increase.

Besides the small variation in NO_3-N concentrations overall (Table 3), one can note that there are more wells in the decrease category than in the increase category (Table 4). Among the possible reasons that could explain the decrease observed in 2021 with respect to 2003 and 2006 are: 1) the proximity of recovered wells to the natural recharge zones of the aquifer after a wet rainy season in late 2020; 2) more effective application of nitrogen fertilizers by farmers in 2020; 3) decrease in nitrate concentration from the unsaturated zone due to denitrification; and 4) a better containment of animal wastes in farms near the recovered wells. However, more information would be necessary to validate each of the above points.

Table 4. Slope of regression line and number of data falling into each of the five assigned categories
Tabla 4. Pendiente de línea de regresión y número de datos que caen dentro de cada una de las cinco categorías asignadas

Category	Slope	Number of Wells	Wells
increase	> 0.290	10	D129, D133, D138, M23, M24, M42, R35, S50, S52, S98
minor increase	0.090 to 0.290	5	D126, D134, J18, M19, M27
no change	-0.089 to 0.089	18	D127, D130, J11, J14, J16, M1, M6, M25, M26, M40, M41, M43, R7, R38, S54, S56, S60, S62
minor decrease	-0.090 to -0.290	14	D119, D136, D137, J9, J12, J13, J17, LC63, LC67, LC69, M21, M39, S44, S45
decrease	< -0.290	13	D139, J10, J15, M5, M22, R2, R3, R30, S46, S47, S57, S58, S59

The spatial distribution of the variation categories is shown in Fig. 4 for a visualization of points in the study area that experienced the most variation (red dots for increase, blue dots for decrease) and for those wells where no change was observed (light green). The color-coded wells in Fig. 4 scatter throughout the area instead of grouping in a zone where a certain tendency (increase or decrease) prevails. However, we can observe that the wells with tendency to increase (red dots for high increase and orange dots for low increase) scatter within the central part of the aquifer. These wells plot the central part of the aquifer in the same region where, as we can observe in Fig. 3, high NO_3-N concentrations are found, coinciding with the location of large dairy farms (Fig. 4). On the other hand, the areas with no change (green dots) or decrease in nitrate concentration (blue dots) correspond to recharge zones at north and south of the aquifer, identified by their higher altitude, i.e., the mountain ranges that limit the aquifer.

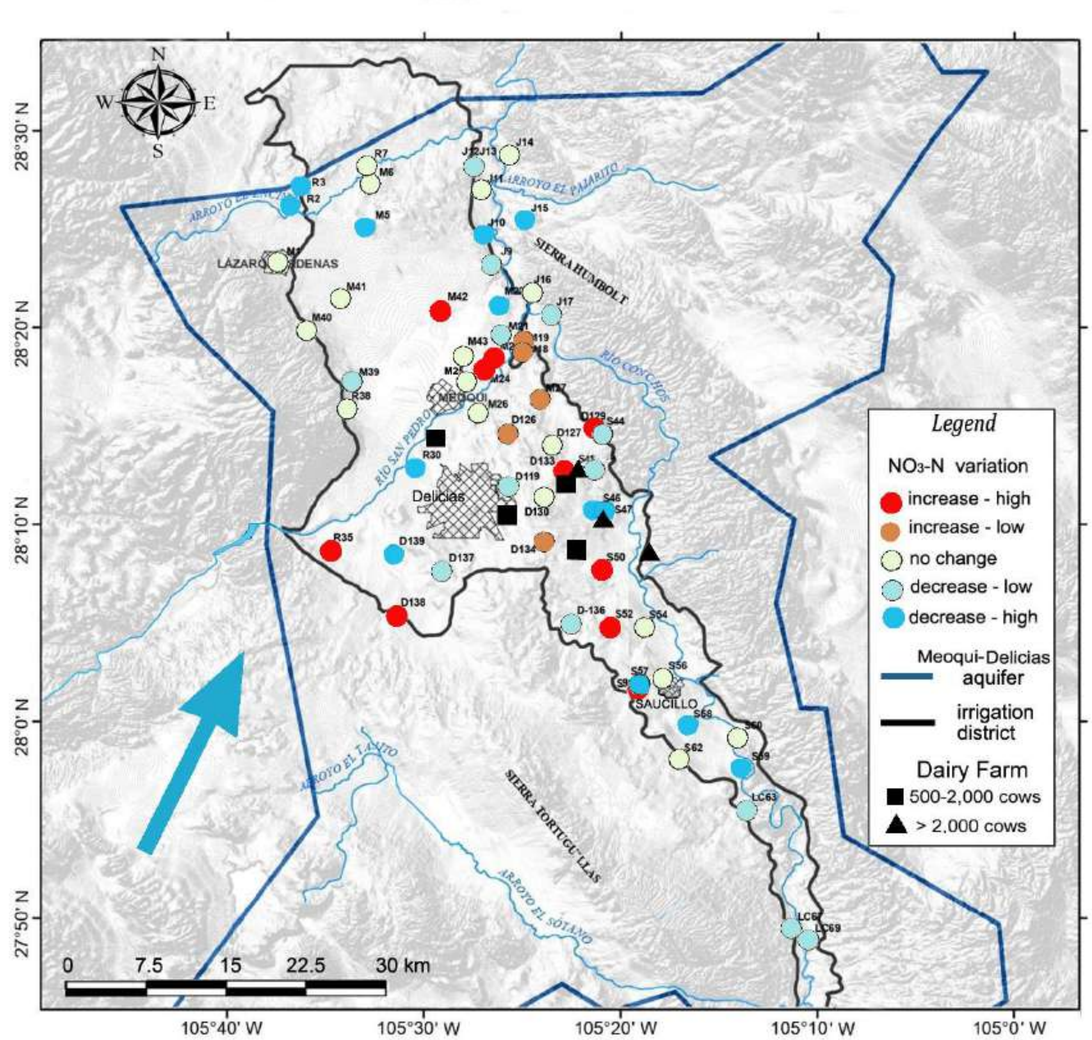

Figure 4. Spatial distribution of the variation of NO_3-N with time according to the five variation intervals: high increase (red), minor increase (orange), no change (light green), minor decrease (light blue), high decrease (dark blue). Regional groundwater flow (blue arrow) and location of largest dairy farms shown for reference purposes.
Figura 4. Distribución espacial de la variación de NO_3-N con el tiempo de acuerdo con los cinco intervalos de variación: incremento alto (rojo), incremento bajo (naranja), sin cambio (verde claro), decremento menor (azul claro), decremento alto (azul oscuro). Flujo regional del agua subterránea (flecha azul) y localización de grandes establos mostrados con propósito de referencia.

A further attempt to explain the spatial distribution of NO_3-N concentrations and their variation with time was to consider the well depth. Well depth information was found only for 18 of the 63 wells and was added to Table 3. The depths vary between 15 and 320 m. One should recall from Section 2 that the average depth of the aquifer is 300 m but can be up to 500 m in places. Especially troubling was to find concentrations above 10 mg/L NO_3-N in 50 % of the deep wells (150-320 m deep). This finding points to nitrate contamination in parts of the aquifer generally considered to be free of anthropogenic contamination. Again, the disposal of animal waste was considered a likely source of contamination because of the high content of nitrogen in animal waste spills and the high NO_3-N concentration locations roughly coinciding with the areas where large dairy farms operate. It is possible that oxidation of dairy farm waste transforms the nitrogen compounds to nitrate (a soluble compound), which is readily transported by water into deeper parts of the aquifer.

Although NO_3-N contamination is not yet severe in the study area, actions to prevent contamination leakage to groundwater are advisable, especially in the areas identified as vulnerable. Recommended actions include improving the waste management of manure and the application of only the needed amount N-fertilizer to reduce N-losses (Rivas-Lucero *et al*., 2008; Millar *et al*., 2018). Also, the successful implementation of any best management practices requires attention to social aspects and a clear and sensible communication between stakeholders, e.g., farmers, city officials, and water managers (McCullogh and Matson, 2016), as farmers often resist the adoption of new procedures until they are convinced of their effectiveness. This step alone may take several years (McCullogh and Matson, 2016).

Among the preventive methods known to reduce leakage of nitrate to the aquifer, the planting of cover crops stands out. Planting a cover crop has been implemented in the Meoqui-Delicias region for decades, although not by all farmers and, in the past few years, water-efficient methods such as drip-irrigation have started to become a new norm. Other preventive methods that are promising but have not been implemented in the region include no-tilling and the use of automatic sensors for fertilizer and water application (Gutiérrez et al., 2021). The use of sensors has been reported as an effective and sustainable method but can be costly (Norton and Ouyang, 2019; Drazic *et al*., 2020). Examples of corrective methods to reduce NO_3 concentrations include the chemical treatment of contaminated water by adsorption to a variety of materials or the use of bio-barrier substrates for nitrate removal by denitrification (Özkaraova *et al*., 2022).

4. Conclusions

Groundwater NO_3 concentrations varied widely from 0.7 to 23.2 mg/L NO_3-N throughout the study area. The distribution of nitrate content and the ratio NO_3-N/Cl<1 suggests an anthropogenic origin of this contaminant related to human and animal wastes, which should be verified in future research. The spatial distribution of NO_3-N concentrations, according to the reviewed literature and information on the area, indicates a possible association with waste from dairy farms, leaking of domestic wastewater, and/or excess of applied N-fertilizer. Specific studies are proposed to corroborate this assumption. According to NPI, 2/3 of the wells are slightly- to non-polluted, and 1/3 are moderately to very polluted. The distribution of the most affected wells was relatively scattered, which suggests the discharge of large amounts of waste at a few specific points. After comparing their variation in concentration from 2003, 2006 and 2021, a small increase in concentration was observed in 17% of the wells, a decrease in 22% of the wells and minor to no change in 61% of the wells. The central part of the aquifer had the highest NO_3-N concentrations and increasing trends; however, wells with less nitrate were also present. Despite relying on only three years of data, the approach followed here successfully identified the affected wells, although the trends need to be confirmed using more years' data. Some deep wells surpassing the 10 mg/L NO_3-N concentration guideline was a concerning find. The implementation of preventive measures to curb pollution, such as an efficient application of N-fertilizer, water-efficient irrigation, and better practices in disposing of dairy farm wastes, are needed to move towards sustainably managing this aquifer.

Author contributions

Conceptualization: M.S.E.V. and M.G.; methodology: M.S.E.V., H.S.H. and A.P.M.; software: N.V.G., H.S.H. and A.P.M.; validation: M.G., A.P.M. and H.S.H..; formal analysis: N.V.G. and M.G.; investigation: M.S.E.V. and N.V.G.; resources: M.S.E.V. and M.G..; data curation: N.V.G..; writing-original draft preparation, M.S.E.V. and N.V.G.; wrinting-review and editing, M.G.; visualization: M.G..; supervision: M.S.E.V..; Project administration: M.G..; funding acquisition: M.S.E.V. All authors have read and agreed to the published version of the manuscript.

Conflict of Interest

Authors have no conflict of interests to declare that are relevant to the content of this article.

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