Science of the Total Environment 857 (2023) 159347

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Hydrochemical controls on arsenic contamination and its health risks in the Comarca Lagunera region (Mexico): Implications of the scientific evidence for public health policy

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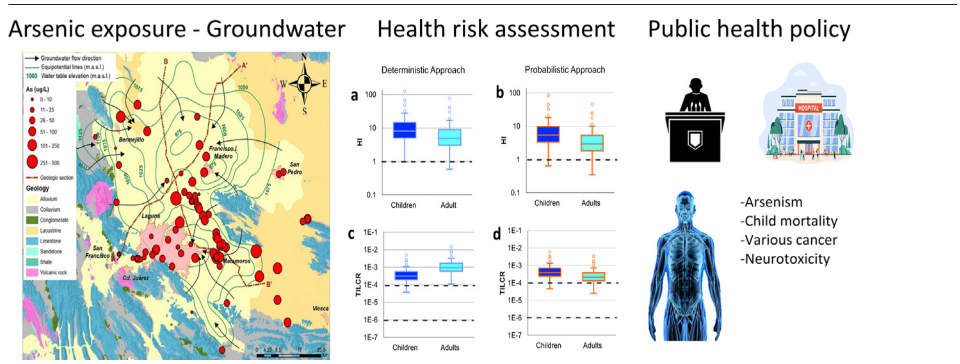
HIGHLIGHTS

- Systematical analysis of As geochemical processes, health impacts and public policy
- As pollution in the aquifer system caused by overextraction of groundwater mainly for irrigation
- Unacceptable As levels in >90 % of the study area and exposure of 2–5 times of safety levels
- Insufficient policy responses due to legal and normative dysfunctional framework evidenced
- Design of orchestrated and coherent collective actions proposed putting health in the center.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Daniel Alessi

Keywords:
Arsenic
Aquifer overexploitation
Health risk assessment
Public policy
Comarca Lagunera region
Mexico

ABSTRACT

Nearly half of the world's urban population depends on aquifers for drinking water. These are increasingly vulnerable to pollution and overexploitation. Besides anthropogenic sources, pollutants such as arsenic (As) are also geogenic and their concentrations have, in some cases, been increased by groundwater pumping. Almost 40 % of Mexico's population relies on groundwater for drinking water purposes; much the aquifers in semi-arid and arid central and northern Mexico is contaminated by As. These are agricultural regions where irrigation water is primarily provided from intensive pumping of the aquifers leading to long-standing declines in the water table. The focus of this study is the main aquifer within the Comarca Lagunera region in Northern Mexico. Although the scientific evidence demonstrates that health effects are associated with long-term exposure to elevated As concentrations, this knowledge has not yielded effective groundwater development and public health policy. A multidisciplinary approach – including the evaluation of geochemistry, human health risk and development and public health policy - was used to provide a

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http://dx.doi.org/10.1016/j.scitotenv.2022.159347
Received 24 June 2022; Received in revised form 30 August 2022; Accepted 6 October 2022
Available online 10 October 2022

current account of these links. The dissolved As concentrations measured exceeded the corresponding World Health Organization guideline for drinking water in 90 % of the sampled wells; for the population drinking this water, the estimated probability of presenting non-carcinogenic health effects was >90 %, and the lifetime risk of developing cancer ranged from 0.5 to 61 cases in 10,000 children and 0.2 to 33 cases in 10,000 adults. The results suggest that insufficient policy responses are due to a complex and dysfunctional groundwater governance framework that compromises the economic, social and environmental sustainability of this region. These findings may valuable to other regions with similar settings that need to design and enact better informed, science-based policies that recognize the value of a more sustainable use of groundwater resources and a healthier population.

1. Introduction

Nearly 50 % of the world's urban population depends on underground water sources. Yet, ever more aquifers are being polluted and overexploited by humans, sometimes with irreversible consequences (UNESCO, 2022). In semi-arid and arid agricultural regions, where groundwater is the primary source of potable water and irrigation, aquifers are commonly contaminated with geogenic contaminants such as arsenic (As) (Li et al., 2020). Chronic arsenicosis due to drinking water has affected over 300 million people worldwide (Hassan, 2018). Numerous studies suggest that chronic exposure to high levels of As significantly increases the risk of health effects. Once consumed, As acts to impair neural development and disrupt endocrine function (including diabetes mellitus), and drives cardiovascular and chronic kidney disease, reproductive disorders, and cancer of the lungs, kidney, bladder, and skin (Berg et al., 2001; Bhowmick et al., 2018; Hassan, 2018; Hong et al., 2014; Mandal and Suzuki, 2002; Naujokas et al., 2013; Palma-Lara et al., 2020; Shakoor et al., 2017). High As concentrations in aquifers also threaten groundwater-irrigated agriculture by polluting soils and reducing crop yields (Bindal and Singh, 2019; Shrivastava et al., 2017).

The Principal Lagunera Region (PLR) aquifer (12,500 km^2) represents the main aquifer within the Comarca Lagunera region in Northern Mexico, where an intensive agricultural development meets with a rapid urban expansion (Dorjderem et al., 2020; Torres-Martínez et al., 2021). The rate of groundwater extraction is approximately twice total annual recharge (CONAGUA, 2020a); this over-extraction has led to a sustained draw-down of the water table and water quality deterioration.

The PLR has long been recognized as having significant water quality problems owing to both natural factors and anthropogenic activities (Arreguín-Cortez et al., 2012; Azpilcueta Pérez et al., 2017; Brouste et al., 1997; Saldarriaga-Noreña et al., 2014). Concentrations of sulfate, nitrate, and fluoride in the PLR commonly exceed drinking water limits set by the World Health Organization (WHO) (Torres-Martínez et al., 2021). Trace elements, such as molybdenum, selenium, and manganese, also exceed WHO recommendations. Arsenic, however, is the most critical concern because of the frequency and degree with which it surpasses the WHO guideline and Mexican standard of 10 μg/L (WHO, 2017; SSA, 2021) as well as the severity in public health consequences. There is a scientific general consensus that the As is geogenic in origin but its concentrations are exacerbated by excessive groundwater pumping. However, the groundwater flow and reaction processes that mobilize the As into pore-waters are still debated (Ortega-Guerrero, 2017; Sariñana-Ruiz et al., 2017).

Adverse health effects driven by As exposure have been documented in the Comarca Lagunera Region since the 1960s (Albores-Medina et al., 1979; Cantellano-Alvarado et al., 1964; Rosas et al., 1999). The frequency of these health affects appear to have increased during the 1980s (Cebrian et al., 1983; CONAGUA, 2000; Del Razo et al., 1990, 1993; Garcia-Salcedo et al., 1984; Gutiérrez-Ávila et al., 1989). In 1993, the region had the highest incidence of genitourinary and skin neoplasms in Mexico and a higher birth defect rate than the national average (3.4 vs. 2.4 %) (Leke et al., 1993). Recent studies continue to document these adverse effects on population health (Armienta and Segovia, 2008; Fisher et al., 2017; Laine et al., 2015). Gamboa-Loira et al. (2020) found that the urinary As levels in 64 % of women in the region exceeded the WHO's guideline value. A summary of human health studies completed over the last 15 years in the region is presented in Table S1 (Supplementary Material).

Despite these long-standing and negative impacts, risk assessments conducted on communities exposed to As-rich groundwater have been insufficient. A clear characterization of health risks from human exposure to As in drinking water is crucial to design effective drinking water management strategies in the region (Saha and Rahman, 2020). Socioeconomic conditions in this region, specifically household incomes and formal access to health services (Adeloju et al., 2021; Shaji et al., 2021), occupy a central role in development policy (Hess, 2016; Todaro and Smith, 2020). Good governance and management are essential for a more sustainable groundwater use (Megdal, 2018). Governance implies actors (not only governments) designing and applying policies through institutions with a normative foundation (Bressers et al., 2016; World Bank, 2017). Specifically, groundwater management includes the actions needed to implement laws, policies and decisions involved in governance (Megdal, 2018). Water governance in Mexico is highly complicated and fragmented (CONAGUA, 2020a; Hoogesteger and Wester, 2017; Kruckova and Turner, 2017; OECD, 2012). There are complex interactions at each spatial scales (i.e., municipality, basin, and federal) (Kruckova and Turner, 2017).

The distribution and degree of As poisoning from untreated groundwater consumption to be better documented at the regional scale. The severity of the problem must be recognized by international organizatons, the general population, the health sector, and administrators (Shaji et al., 2021). The distribution of As in groundwater and the health impacts it causes must be addressed through preventative policy actions (Zuzolo et al., 2020). Therefore, the objectives of this paper are: 1) to update the geochemical model of the aquifer based on new evidence; and 2) to explain the implications of this model for policy that can address groundwater overextraction and public health.

2. Materials and methods

2.1. Study area

2.1.1. Geographical and socioeconomic framework

The PLR aquifer is located in the heart of the Comarca Lagunera region in north-central Mexico (25°00′ - 26°45′N; 102°15′ - 103°50′W; Fig. 1a). The land surface is composed of lacustrine clay deposits. As it is a desert, these clays are desiccated, and covered by xerophilous scrubs (Cervantes Ramírez and Franco González, 2010). The climate ranges from arid to semi-arid in the plain, and from semi-arid to temperate in the mountainous area (Gabriel Morales and Pérez Damián, 2010). The average annual precipitation and temperature in the plain are 260 mm and 19.0 °C, respectively (CONAGUA, 2018). The majority of precipitation falls in the wet season between June and September.

The main source of surface water in the region comes from the Nazas and Aguanaval rivers, which discharge their waters into the now dry lagoons of Mayrán and Viesca. The Nazas river is regulated by the Lázaro Cárdenas and Francisco Zarco dams (Fig. 1b). The lower reaches of the Nazas (below the dams) only transports water during the irrigation cycle (March–August) and during exceptional flood events (CONAGUA, 2018).

This region's economy is composed mainly of intensive livestock (meat and dairy), irrigated agricultural production (often linked to livestock), mining, and industrial activities (Torres-Martínez et al., 2021) (Supplementary Material, Fig. S1). The region hosts 1.6 million inhabitants, distributed in nine municipalities, five in Coahuila and four in Durango. Around 86 % of the populaton lives in the La Laguna Metropolitan Zone (LMZ),

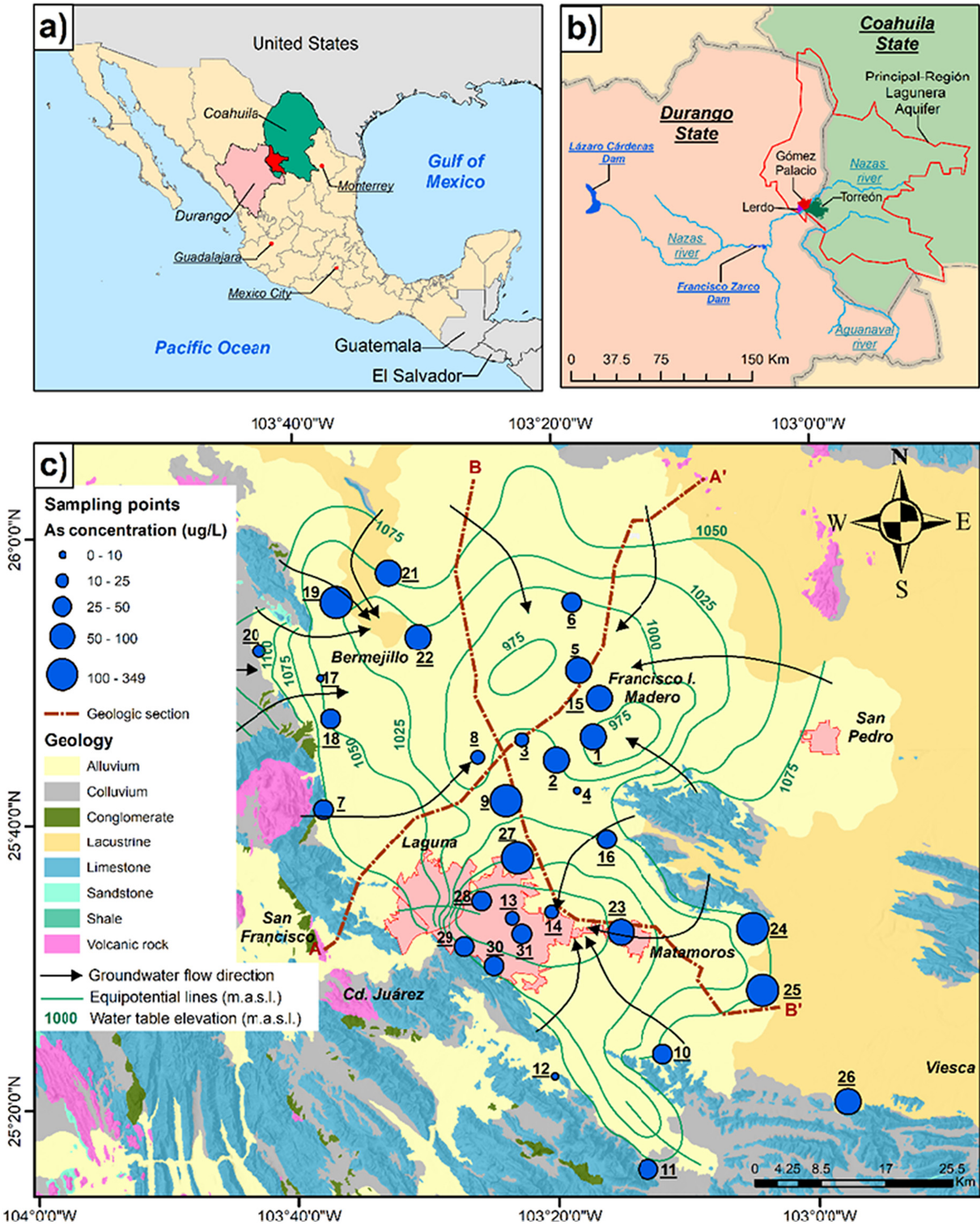

Fig. 1. a) Location of the study area and states of Durango and Coahuila in Mexico; b) Principal Lagunera Region aquifer (red outline) with Laguna Metropolitan Zone integrating the cities of Torreón, Gómez Palacio, Lerdo, and Matamoros; c) Surface geology of the study area with groundwater equipotential lines corresponding to 2018, sampling sites with As concentration. Adapted from SGM (2000, 2008) and CONAGUA (1999). The underlined numbers indicate the ID and location of wells that were sampled for the present study. The light pink areas with red outlines represent urban areas.

comprising the conurbated cities of Torreón, Gómez Palacio, Lerdo, and Matamoros (INEGI, 2020) (Fig. 1b).

2.1.2. Hydrogeological context

Most of the region is composed of a plain that is interrupted by mountain ranges trending NW-SE (Gutiérrez-Ojeda, 1995; CONAGUA, 2018) (Fig. 1c). The plain is mainly composed of clastic materials that form intercalated layers of gravel, sand, silt, and clay, which was physically eroded from the surrounding mountains and transported primarily by the Nazas and Aguanaval rivers (Gutiérrez-Ojeda, 1995; CONAGUA, 1992, 1999, 2011). Smaller mountain ranges with folded strata of marine sedimentary rocks of Mesozoic age, consisting mainly of limestone, sandstone, and shale, define the southern, western and northern limits of the aquifer (CONAGUA, 2011).

The aquifer system is comprised of poorly-consolidated sediments and fractured rocks (Table S1). The upper, granular aquifer is unconfined; however, sandy-loam and clay horizons create locally semi-confined conditions (Esquivel Victoria, 2008; CONAGUA, 2011, 2018). Most of the existing wells extract groundwater from this upper aquifer unit. Their depths are typically 500 m. The lower, fractured aquifer is comprised of limestones and dolomites from the Lower to Upper Cretaceous. The circulation of water in these rocks, mostly through dissolution ducts and caverns, appears to follow the course of geological structures, emerging as springs in fault zones or areas where permeable limestones are in contact with overlying alluvial-lacustrine filling material (CONAGUA, 1992, 1999, 2018; Molina Maldonado, 2004; Aparicio González, 2018).

Most of the recharge to the granular aquifer is derived from the Lázaro Cárdenas and Francisco Zarco dams, with local and lateral infiltrations through the alluvial deposits of the Nazas river (CONAGUA, 1992; González Hita et al., 1994; Gutiérrez-Ojeda, 1995). Other contributions are from irrigation return flows, rainwater infiltration and lateral flow from adjacent aquifers (CONAGUA, 2002; Esquivel Victoria, 2008).

2.1.3. Groundwater extraction and use

The rate of groundatger extraction from the PLR aquifer has been increasing for many years; the combination of fertile sedimentary soils, abundant sunshine, warm to hot temperatures, and water from the Nazas and Aguanaval rivers made it an attractive place for irrigated agriculture since pre-colonial times. By 1940, >1000 drilled production wells were in use, leading to the first obvious signs of water table decline (Gutiérrez-Ojeda, 1995; CONAGUA, 1999). The construction of the Lázaro Cárdenas dam in the 1940's reduced recharge to the aquifer from the Nazas river considerably (ECL, 2019; SARH, 1986). The subsequent lining of >4000 km of irrigation channels to reduce irrigation losses further reduced recharge (Gutiérrez-Ojeda, 1995). By the early 1960s, approximately 3000 production wells were in use (Gutiérrez-Ojeda, 1995; CONAGUA, 1999; Hoogesteger, 2018).

In 2020 an estimated 1088.5 hm^3/yr of permitted water was extracted from approximately 2350 active deep wells. This is approximately twice the recharge rate (534 hm^3) (CONAGUA, 2020a). The extracted volume is primarily used for agriculture (81.2 %), drinking water supply (11.6 %), and livestock (4.6 %). An additional 440 hm^3 of non-permitted (i.e., illegal) groundwater was pumped for agriculture (ECL, 2019).

Under pre-development conditions the regional groundwater flow direction was parallel to the Nazas and Aguanaval rivers (CONAGUA, 1992, 1999) from SW to NE (Fig. 1b). However, since 1975, cones of depression have been reported in zones with a high density of wells pumping in the central part of the aquifer (Molina Maldonado, 2004) causing groundwater to flow concentrically towards those the center of the aquifer (Gutiérrez-Ojeda, 1995). Today, these cones of depression continue to expand and merge (Fig. 1c). The average annual rate of the water table decline is 1.6 m/yr (CONAGUA, 2020b).

2.1.4. Groundwater governance

Groundwater governance in the Comarca Lagunera region reflects the national framework, plus additional complexities that are common in shared government jurisdictions. Article 27 of the Mexican constitution confers ownership of water to the nation. According to the National Water Law (Ley de Aguas Nacionales - LAN), water users need a concession (i.e., permit) from the national water authority, CONAGUA, to extract groundwater (Hoogesteger and Wester, 2017; SARH, 1992). Users pay a water fee that varies with the permitted extraction volume and intended use. According to article 115 of the Mexican constitution, municipalitites are responsible for providing domestic water. Different federal entities regulate water quality. These are covered under environmental regulations set by the Secretariat of the Environment (Secretaría de Medio Ambiente y Recursos Naturales - SEMARNAT), public health rules set by the Secretary of Health (Secretaria de Salud), and regulatory risks functions (e.g. food safety, environmental protection) are set by the Federal Commission for Protection of Sanitary Risks (COFEPRIS). Lastly, the human right to water is contained in Article 4 of the Constitution.

Coahuila and Durango states have their own state water commissions. These are supposed to regulate and support the provision of water services (drinking water, sanitation), which is implemented by municipal water and sanitation utilities (Wilder et al., 2020). A major irrigation district (Distrito de Riego 017) covering ~52,000 ha is also located within the limits of PLR aquifer unit. The different levels of government that have overlapping roles to manage water sourcing, waste water treatment and safeguard water quality and quantity, makes groundwater management a very difficult task and explains some of the rampant overexploitation and widespread human exposure to toxic levels of As today. This is because the responsibility to address As concentrations and evaluate their implications for health and development policy is also fragmented across several governing bodies. The official plans and responses as well as the legal norms and regulations are not coordinated between these bodies.

2.2. Methods

2.2.1. Geochemical work

Thirty-one groundwater samples were collected in November 2018 (Table S2). The sites were selected to span a range of land-uses, potential contamination sources, and hydrochemistry based on the national monitoring network of Comisión Nacional del Agua (CONAGUA). Physicochemical parameters (pH, temperature, ORP, salinity, and electrical conductivity), major cations (Na^+, K^+, Ca^{2+}, Mg^{2+}), anions (Cl^-, F^-, SO_4^{2-}, NO_3^-), minor and trace elements, and water isotopes (δ^2H and $\delta^{18}O$). Water isotopes were expressed as delta per mil (‰) which represents the deviation of the ratio of heavy to light isotopes from the Vienna Standard Mean Ocean Water (VSMOW) reference standard. Detailed laboratory methods and used equipments are provided in Section S1.

Collinearity amongst the hydrochemical parameters was assessed using the non-parametric Spearman rank correlation analysis since the values of many variables were not normally distributed. Significant association between any two variables was assessed at both $p = 0.01$ and $p = 0.05$. To examine whether As concentrations were significantly different across categories, the non-parametric Kruskal-Wallis test was used with a significance level of $p = 0.05$.

To determine if a trend of dissolution or precipitation of minerals was predominant, saturation indices were calculated. The saturation index (SI) is defined as SI = log (IAP/K) where IAP is the ion activity product and K is the equilibrium constant. Equilibrium is indicated when SI = 0; the groundwater is oversaturated with respect to the particular mineral when SI > 0, which means that the mineral phase may precipitate to achieve equilibrium. If SI < 0, the groundwater is undersaturated with mineral phase, which means that dissolution is required to reach equilibrium. The geochemical code PHREEQC (Parkhurst and Appelo, 1999) was applied for this exercise.

2.2.2. Assessment of human health risks from As contaminated water

The human health risk from exposure to As in well water was performed using an US Environmental Protection Agency (EPA) method (USEPA, 1996, 2004). Ingestion and dermal contact were considered to be the main routes of exposure to As in water to adults and children. Initially, point estimates of deterministic risk were calculated. Next, a probabilistic health risk was simulated for a more complete and realistic estimation by integrating the uncertainty and variability of the risk calculation parameters. Data from 91 wells were combined from this and another recent study (Sariñana-Ruiz et al., 2017) (Table S3). Water As concentrations were used to calculate the average daily intake for ingestion and dermal contact (Eqs. (1) and (2)):

$$ADI_{ing} = \frac{CW \times IR \times EF \times ED}{BW \times AT} \quad (1)$$

$$ADI_{derm} = \frac{CW \times SA \times Kp \times EF \times ED \times ET \times CF}{BW \times AT} \quad (2)$$

where ADI_{ing} and ADI_{derm} are the average daily intake of As through ingestion (drinking water) and dermal exposure, respectively (μg kg^{-1} day^{-1}); *CW* is the measured groundwater As concentration (μg/ L). The population was assumed to use untreated groundwater for drinking, as field observations found the water is rarely treated for As removal; *IR* is the rate of water intake (L day^{-1}); *EF* is the frequency of exposure (days year^{-1}); *ED* is the exposure duration (years); *BW* is the average body weight (kg); *AT* is the average time of exposure (days); *SA* is the exposed skin surface area available for contact (m^2); *Kp* is the dermal permeability constant (cm h^{-1}); *ET* is the exposure time (h day^{-1}); and CF is the unit conversion factor (=10).

The carcinogenic risk, expressed as the incremental lifetime cancer risk (ILCR), is estimated as follows (Eq. (3)):

$$ILCR = ADI \times CSF \tag{3}$$

where *ILCR* is the probability that an individual will develop cancer as a result of exposure to a contaminant, and *CSF* is the cancer-causing slope factor (μg kg^{-1} day^{-1}). To estimate the total carcinogenic risks from combined ingestion and dermal exposures, the calculated ILCR values for each (Eq. (3)) were summed and expressed as total incremental lifetime cancer risk (TILCR). For reference, the EPA adopts carcinogenic risk critical points between 1×10^{-6} (i.e., 1 case in 1 million people), a risk management goal (point of excess cancer risk), and 1×10^{-4} (i.e., 1 case in 10,000 people), an unacceptable risk (health hazards, which need some sort of intervention and remediation) (USEPA, 2012). Here, results were compared with the latter, the unacceptable risk level.

The hazard quotient (HQ), determines the risk of non-carcinogenic effects due to ingestion or dermal exposure to As in water. It is calculated as follows (USEPA, 2004) (Eq. (4)):

$$HQ = \frac{ADI}{RfD} \tag{4}$$

where *RfD* is the As reference dose (μg kg^{-1} day^{-1}). To estimate the total hazard risk from combined ingestion and dermal exposures, the calculated HQ values from ingestion and dermal contact (Eq. (4)) were summed and expressed as hazard index (HI). When the HI is >1, there is a potential non-carinogenic health risk (USEPA, 1989; Zhang et al., 2019). The RfD and CSF for As were obtained from IRIS database (USEPA, 2022).

After the initial development of a deterministic model, probabisitic modeling was performed with a Monte Carlo simulation using the software Crystal Ball (Oracle, 2017). The previously calculated point estimates were complemented with probability distribution assumptions to calculate a distribution of the output. The fitting distribution functions for input data were tested through goodness-of-fit tests. The description of all the parameters and their values used in both the deterministic and probabilistic risk estimation is given in Table S4.

2.2.3. Poverty and access to formal health services

As mentioned above, it was assumed that the whole population consumes untreated groundwater. As mentioned previously, this assumption is supported by field observations despite the existence of different water treatment technologies for absorption of As (Adio et al., 2017; Alswat et al., 2016; Saleh, 2020, 2021; Saleh et al., 2022). However, unofficial estimates from locals who were asked suggest that 60–70 % of the population consume bottled water from private establishments known as water houses (*casas de agua*). These water houses are supposed to be overseen by local water authorities, the municipalities and the health secretariats. In practice, however, this does not seem to be the case. Therefore, the human health risks calculated here are only valid for the share of population without access to bottled water, i.e. the rural and urban poor. Poverty levels and formal access to health services were analyzed against access to water, the As concentrations and health risks, as well as the access to formal health services (CONEVAL, 2021).

2.2.4. Development and public health policy in relation to high As concentrations

To assess whether As concentrations and health risks are considered in existing development and public health policy, the current development plans of the states of Coahuila and Durango and their two largest municipalities, Torreón, Coahuila (GMT, 2019) and Gómez Palacio, Durango (GMGP, 2019) were analyzed (GECZ, 2017; GED, 2016). The goal was to determine if these plans addressed the issues of groundwater overexploitation, As concentrations, and health impacts, in both the diagnosis and policy sections.

3. Results

3.1. Water quality and standards

The physicochemical water properties are shown in Table S5. The statistical summary of this data is presented in Table 1. The pH varied from 6.0 to 8.6 (average 7.6), indicating a slightly acidic to alkaline range. The water temperature ranged from 23.7 to 33.6 °C (average 28.5 °C); 72 % of the groundwater wells produced water with a temperature that exceeded mean air temperature (30 °C), suggesting a mild geothermal influence. The EC and TDS values ranged from 332 to 5957 μS/cm (average 1424 μS/cm) and 190 and 3894 mg/L (average 885 mg/L), respectively, which spans fresh to brackish water. The most mineralized groundwater was located in the northern and northwestern portion of the study area.

Six physicochemical parameters (pH, TDS, SO_4^{2-}, NO_3^-, As, F^-) exceeded the Mexican standard for safe drinking water in different percentages of samples ranging from 6 to 90 % (Table 1). Most remarkably, two samples had As concentrations that reached ~35 times the WHO/Mexican standard for safe drinking water.

The relative order of ion abundances and correlation analysis between elemental concentrations within the well water samples are effective tools to help reveal physical processes and chemical reactions that drive the chemical composition of the aquifer. Groundwaters in the study area are chemically evolved as they are rich in Na^+ and SO_4^{2-}. Arsenic concentrations are associated with F and temperature which suggests a geogenic origin. The relative order of cation abundance was $Na^+ > Ca^{2+} > Mg^{2+} > K^+$, while the order of anion abundance followed $SO_4^{2-} > HCO_3^- > NO_3^- > Cl^- > F^-$.

A correlation matrix of physicochemical parameters is given in Table S6. Significant positive Spearman rank correlations ($p < 0.01$) were observed between Ca^{2+} and Mg^{2+} ($r_s = 0.88$), Ca^{2+} and SO_4^{2-} ($r_s = 0.87$), and between Na^+ and SO_4^{2-} ($r_s = 0.73$), Cl^- and Ca^{2+} ($r_s = 0.77$). NO_3^- was significantly correlated with halides Br^- ($r_s = 0.74$) and Cl^- ($r_s = 0.74$), Ca^{2+} ($r_s = 0.69$), Mg^{2+} ($r_s = 0.67$), and K^+ ($r_s = 0.63$), while pH and HCO_3^- showed a highly inverse association with NO_3^- ($r_s = -0.62$ and -0.56, respectively). Trace elements Sr^{2+} and U are strongly associated with alkali metals (Li^+,Na^+, and K^+) and earth alkalies (Mg^{2+}, Ca^{2+}) as well as SO_4^{2-}, NO_3^-, halides (Cl^-, Br^-) and salinity. Arsenic is correlated ($p = 0.01$) with temperature ($r_s = 0.65$), F^- ($r_s = 0.66$), and Al^{3+} ($r_s = 0.62$), and has a negative correlation with NO_3^- ($r_s = -0.54$). At a reduced significance level of $p = 0.05$, As is positively correlated with pH ($r_s = 0.39$), and negatively correlated with HCO_3^- ($r_s = -0.39$), Ca^{2+} ($r_s = -0.37$) and K^+ ($r_s = -0.36$).

3.2. Geochemical processes controlling groundwater chemistry

The main geochemical processes that explain the distribution of As concentrations across the PLR aquifer as constrained by isotopic and chemical information are depicted in Fig. 2. The isotopic $\delta^{18}O$ and δ^2H values of groundwater samples ranged from −9.97 ‰ to −7.18 ‰, and −71.54 ‰ to −52.89 ‰, respectively. According to the Craig diagram in Fig. 2a, the groundwater is of meteoric origin and falls below the global meteoric water line (GMWL; Rozanski et al., 1993). The linear trend $\delta^2H = 5.168\delta^{18}O - 19.25$ reflects evaporation during groundwater recharge. As a slope of the evaporation line of >5 is atypical for a semi-arid climate, the observed trend suggests the mixing of evaporated soil water with infiltrated rainwater/recirculated agricultural water (Clark and Fritz, 1997;

Table 1

Statistical summary of physicochemical parameters. Note: All data are in mg/L, except where otherwise indicated. SD means the mean standard deviation. WHO means World Health Organization safe water drinking guideline (WHO, 2017). NOM-127 represents the Mexican standard for drinking water.[c]

Parameter	n	Min	Mean	SD	Median	Max	WHO (2017), mg/L	% > WHO	NOM-127 (2021), mg/L	% > NOM-127
Temperature (°C)	31	23.70	28.65	2.51	28.40	33.60	–	N/A	–	N/A
DO	31	1.29	4.23	1.77	4.06	7.31	–	N/A	–	N/A
pH (−)	31	6.00	7.55	0.53	7.64	8.56	–	N/A	6.5–8.5	6
TDS	31	189.50	786.98	601.95	630.50	2957.50	–	N/A	1000	26
EC (μS/cm)	31	332.10	1278.17	960.67	1029.00	4704.00	–	N/A	–	N/A
Ca	31	10.10	133.62	137.02	81.10	526.00	–	N/A	–	N/A
Mg	31	0.09	19.15	21.89	8.01	99.50	–	N/A	–	N/A
Na	31	22.00	126.53	120.82	115.00	691.00	–	N/A	–	N/A
K	31	0.85	3.73	2.05	3.34	8.98	–	N/A	–	N/A
HCO_3	31	104.92	170.23	42.36	162.26	280.97	–	N/A	–	N/A
SO_4	31	32.10	415.57	541.20	308.00	2630.00	–	N/A	400	32
Cl	31	3.51	44.75	44.44	29.60	158.00	–	N/A	–	N/A
NO_3 as N	31	0.01	7.74	11.59	2.42	45.00	11[a]	23	11	23
Si	31	11.70	19.46	4.63	18.70	31.10	–	N/A	–	N/A
Al[b]	31	0.00	0.00	0.02	0.00	0.10	–	N/A	0.2	0
As	31	0.01	0.07	0.08	0.05	0.35	0.01	90	0.01[a]	90
B	31	0.07	0.35	0.29	0.30	1.54	2.4	0	–	N/A
Br	31	0.05	0.51	0.55	0.29	2.43	–	N/A	–	N/A
F	31	0.12	0.88	0.68	0.64	3.09	1.5	16	1.0	26
Fe[b]	31	0.0001	0.008	0.011	0.005	0.052	–	N/A	0.3	0
Li	31	0.02	0.10	0.05	0.09	0.27	–	N/A	–	N/A
Mn	31	0.00001	0.010	0.017	0.001	0.070	–	N/A	0.15	0
Mo	31	0.002	0.022	0.054	0.009	0.306	–	N/A	–	N/A
Sr	31	0.02	2.94	4.94	0.91	24.60	–	N/A	–	N/A
U	31	0.001	0.009	0.011	0.005	0.049	0.03	10	–	N/A
$\delta^{18}O$ (permil)	31	−9.97	−8.22	0.64	−8.07	−7.18	–	N/A	–	N/A
δ^2H (permil)	31	−71.54	−61.61	3.66	−61.20	−52.89	–	N/A	–	N/A

[a] Equivalent to 50 mg/L NO_3 (as NO_3^-).

[b] Not detected values of Fe and Al were censored (0.5 * lower detection level).

[c] The Mexican drinking water standard (SSA, 2021) prescribes a diferenciated reduction of permissible level of As to 10 μg/L, depending on the size of the population of urban centers.

Mahlknecht et al., 2008; Torres-Martínez et al., 2021). The deuterium excess varied between 1.2 and 10.7 ‰, which infers a post-condensation evaporative effect. The combination of a wide range of TDS (199–2957 mg/L) and relatively stable $\delta^{18}O$ values (≈ − 8.0 ‰) suggests that processes other than evaporation were mainly responsible for increasing salinity of groundwater. The relatively stable $\delta^{18}O$ over groundwater depth also infers important mixing processes.

Bivariate plots suggest that dissolution of evaporites such as gypsum drives the evolution of groundwater chemistry. The Gibbs diagram in Fig. 2b confirms the dominance of water-rock interactions controlling major ion chemistry. The Piper plot (Fig. 2c) reveals that samples matched Ca- to Na-type (left triangle) regarding cations, while with respect to anions the majority plotted as SO_4-type and the remaining as HCO_3-SO_4 mixed type. Na-normalized molar ratios of HCO_3^- (0.05–4.2), Ca^{2+} (0.1–2.9) and Mg^{2+} (0.0015–1.02) in groundwater are presented in Fig. 2d,e and plot along a mixing line that spans silicate, evaporite and carbonate mineral compositions as end-members (Gaillardet et al., 1999).

Groundwater chemistry evolution is mainly controlled by incongruent silicate and evaporite weathering. Water samples with a Na^+/Cl^+ molar equivalent ratio of 1 indicates halite dissolution. The samples display a range of this ratio from 1.3 to 26.3, which suggests that plagioclase/albite weathering was the main process and halite dissolution was insignificant in the study area (Fig. 2f) (Appelo and Postma, 2005). The combined concentration of Ca^{2+} and Mg^{2+} was equal to, or lower than, the combined concentration of SO_4^{2+} and HCO_3^- (Fig. 2g). According to Hounslow (1995), this could mean that the source of Ca^{2+} is silicate weathering, but other processes such as pyrite oxidation and calcite precipitation could not be excluded.

To evaluate ion exchange processes, a plot of (Ca + Mg)-(HCO_3 + SO_4) versus (Na + K- Cl) was included (Fig. 2h). The expression on the vertical axis, (Ca + Mg)-(HCO_3 + SO_4), represents the amount of Ca and Mg gained or lost relative to that provided by gypsum, calcite and dolomite; while the term (Na + K-Cl) represents the amount of Ca and Mg gained or lost in relation to chloride salt. If ion exchange were a dominant process in the system, the should plot close to a line with a slope of −1. Since many points lie far away from this cation exchange line, this indicates either that most Na^+, Ca^{2+} and Mg^{2+} do not participate in ion exchange reactions or that ion exchange processes are altered by other processes. Saturation indices depict a trend from undersaturation to saturation with increasing salinity/transit time regarding gypsum and anhydrite phases, which confirms the hypothesis of dissolution of these evaporites (Fig. S2).

3.3. Increasing As contamination and current situation

A long record of As measurements in well water within this study area exists over the past 60 years (Table S7), albeit with a sampling bias. Studies with small sample sizes would have the most variability in cumulative histograms (most uncertainty). These results consistently exhibit the longstanding existence of high As concentrations. Whereas the maximum As concentration in the aquifer value has not increased over the past decades, the median concentration has. Arsenic contamination has become endemic in the PLR aquifer over this period (Section S2).

The greatest increases in As concentrations are located in three geographic areas (Fig. 1c; Fig. S3); the first is located in alluvial sediments in the regional cones of depression north of La Laguna metropolitan area towards Francisco I. Madero. Here the water table elevation is below 1000 masl. The second area lies within lacustrine sediments in the southeast, revealing the highest As concentrations of the region; and the third is in the northern portion close to Bermejillo. These three areas bundle the samples with elevated As concentrations.

Arsenic concentrations varied across land uses, municipalities and outcropping geology type, however, the small number of samples in each category limited demonstrating statistical significance with the Kruskal-Wallis test at $p = 0.05$. Concentrations found in agricultural well water

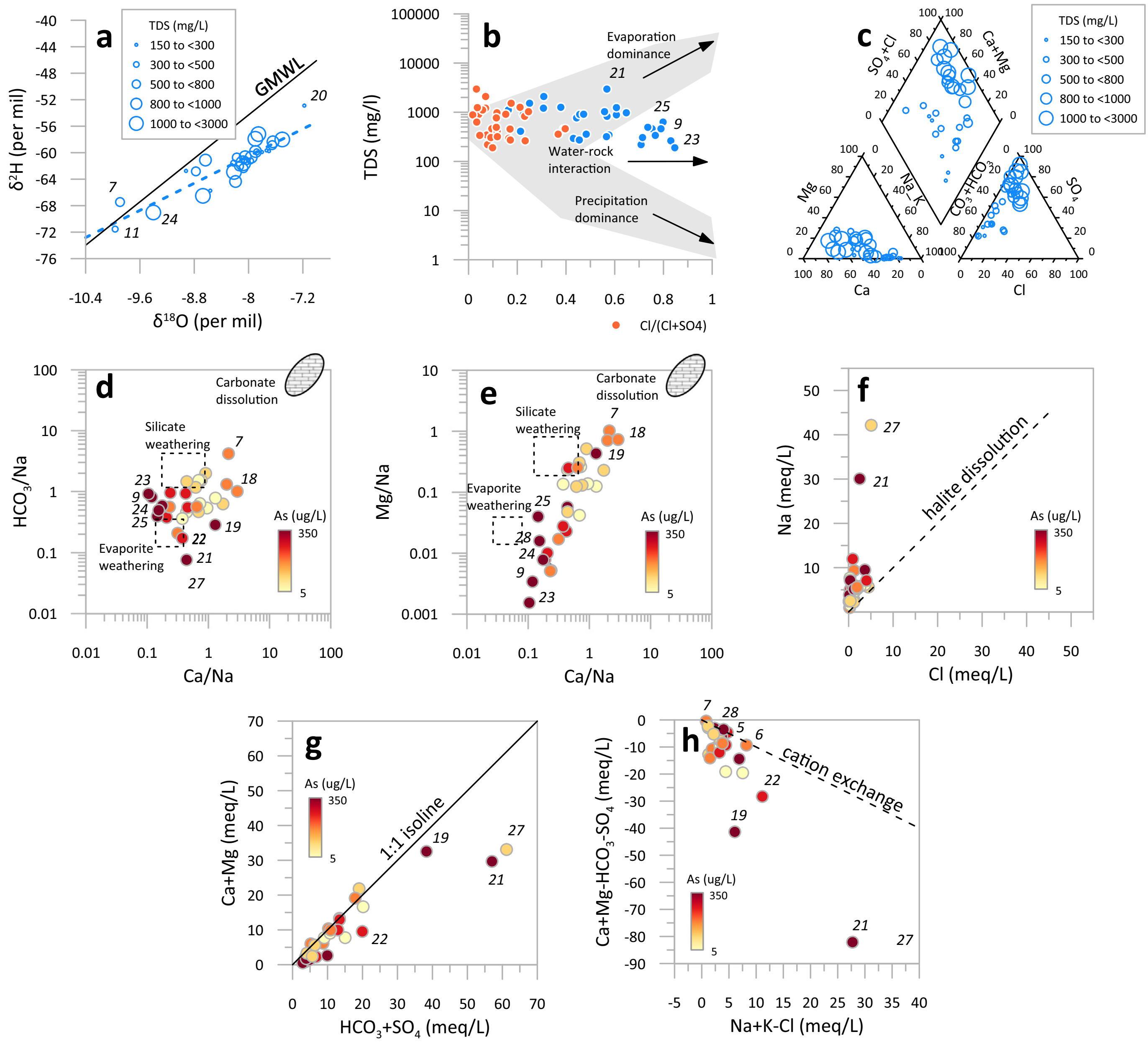

Fig. 2. (a) Values of $\delta^{18}O$ and $\delta^{2}H$ in groundwater of the PLR aquifer and Global Meteoric Water Line (GMWL, Rozanski et al., 1993), showing that evaporation is not driving As concentration; (b) Gibbs plot indicating water-sediment interaction as the primary process controlling the hydrogeochemistry in the study area; (c) Piper plot showing the evolution of major ion geochemistry in the groundwater, and the dominance of sulfate; (d) Na-normalized HCO_3 versus Na-normalized Ca, suggesting the predominance of silicate weathering and evaporite dissolution, and the insignificant contribution of carbonate dissolution; (e) Na-normalized Mg versus Na-normalized Ca; (f) Na versus Cl relationship; (g) Equivalent (Ca + Mg) versus (HCO_3 + SO_4) relationship; (h) Plot for evaluation of ion exchange.

(mean 85.8 μg/L, n = 16) tended to be higher than those in urban well water (57.5 μg/L, n = 9) and more than twice than those in wells from undeveloped shrubland (37 μg/L, n = 6) (Fig. S1 and Table S5). On average (mean water As), the most contaminated municipalities were Gómez-Palacio (105.4 μg/L, n = 4) and Viesca (104.5 μg/L, n = 6), while the least polluted were Torreón (48.7 μg/L, n = 7) and Mapimi (20.5 μg/L, n = 3). Finally, water samples from alluvium sediments demonstrated higher As concentrations (76.1 μg/L, n = 26) than limestones (38.3 μg/L, n = 5).

3.4. Assessing potential health risks using a probabilistic approach

The results of deterministic and probabilistic risk analysis are given in Table S8 and summarized in Fig. 3. Overall, the results show that there is a significant non-carcinogenic health risk (expressed as hazard index, HI) for both children and adults. The probability of exceeding the reference dose was 97 % for children and 91 % for adults using the probabilistic model. These were similar to values estimated using the deterministic model (100 % for children and 96 % for adults). Dermal exposure contributed only minimally (<1 %) to the HI compared to the risks from ingestion. The median exposure dose for children and adults was 5.4 and 3.9 times the reference dose, respectively.

The probability of exceeding the acceptable incremental lifetime cancer risk (TILCR) level was 96 % for children and 83 % for adults using the probabilistic model. These were somewhat different than the values estimated using the deterministic model for children (91 %) and adults (100 %). The difference between the deterministic and probabilistic approach is due to the distribution assumptions in the Monte Carlo simulation. The

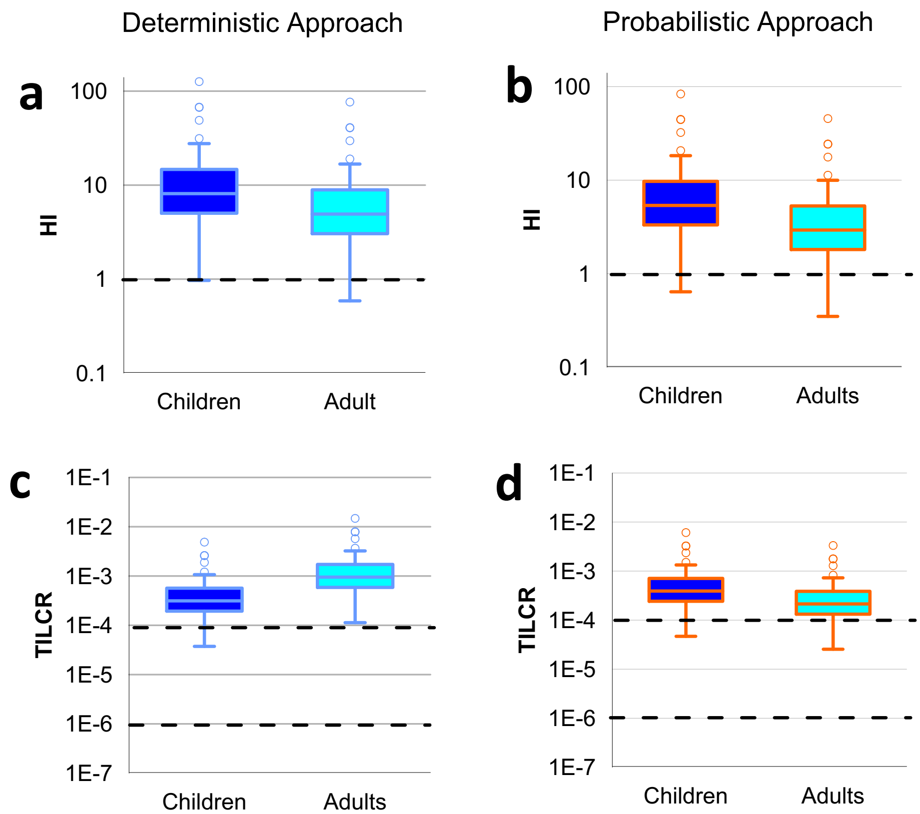

Fig. 3. Box plots showing hazard index, HI (a and b) and total incremental lifetime cancer risk, TILCR (c and d) estimated by deterministic (a and c) and probabilistic approaches (b and d) for children and adults exposed to groundwater As (n = 91 wells) through ingestion and dermal contact. Note 1: Horizontal dashed lines indicate safety levels commented in the method section: the upper represents the unacceptable risk level and the lower the risk management goal. Note 2: Data points lying outside of the upper and lower inner fences of the box plots (Q1 ± (1.5 * interquartile range)) were considered outliers.

contribution from dermal exposure to the TILCR was insignificant (<1 %). The median TILCR for both children and adults was 3.9 and 2.1 times the acceptable value.

The potential health risk distribution obtained by combining the human health risk assessment model with the spatial data analysis model is shown in Fig. 4a,b, the carcinogenic risk according to the probabilistic approach varied between $4.7 \cdot 10^{-5}$ and $6.1 \cdot 10^{-3}$ for children and between $2.5 \cdot 10^{-5}$ and $3.3 \cdot 10^{-3}$ for adults. It shows an unacceptable level of carcinogenic risk in >95 % of the study area for adults. The non-carcinogenic risk for children varied between 0.6 and 83.5 and between 0.35 and 45.5 for adults (Fig. 4c,d). Again, the risk category was higher for children than for adults, and the threshold of 1.0 was typically exceeded for both age groups. For both carcinogenic and non-carcinogenic risk, the most exposed municipalities were Matamoros (east), Viesca (north), Gómez Palacio (west), San Pedro (south) and Tlahualillo (southwest), while Mapimi, Lerdo, Francisco I. Madero and Torreón were less exposed. The results of the spatial distribution using the deterministic approach were similar (Fig. S4).

3.5. Poverty and formal access to health services

The current and potential health risks are framed within a regional context of widespread poverty and low formal access to health services. Table 2 shows that over 650,000 (or 41 %) of the aquifer's population fall below poverty line (CONEVAL, 2021). Although with fewer inhabitants than the metropolitan areas, non-metropolitan, smaller municipalities - like San Pedro, Viesca, Mapimí and Tlahualilo – have higher percentages of households in poverty of nearly 50 %. This figure is similar to the national average. However, in absolute terms, the LMZ concentrates most people in this condition (approximately 542,000 inhabitants or 83 % of all); Torreón and Gómez Palacio alone concentrate two thirds.

A large part of the population in the aquifer area lacks formal access to health services. Over 370,000 people or 23 % of the entire population living above the aquifer are in this situation. This percentage increases to 25 % as an average for non-metropolitan municipalities or 58,000 people. Thus, a slightly larger proportion of inhabitant in rural municipalities lack formal access to health services than metropolitan areas, however, the metropolitan contain the greatest population without access to these services (84 % of the whole region). For example, Torreón and Gómez Palacio alone house two thirds of the people in the region lacking access to health services (Table 2).

3.6. Development and health policy

The previous findings have profound implications for development and health policy. As previously highlighted, Gómez Palacio is the most exposed municipality to carcinogenic risks. Viesca is also a municipality exposed to high risks. These spatial variations are worth considering in the design of a comprehensive public policy that links the quality of groundwater, risks to health, poverty and access to health services. However, these links are almost absent in formal development and health planning for the PLR.

The two state development plans of Coahuila and Durango are elaborated in compliance with federal and state constitutions and planning laws. Although the Comarca Lagunera region is shared by these two states, joint planning is rarely conducted. Documents from both states recognize the right of the population to a healthy environment, and that a lot of work is needed to achieve this goal. The case of As contamination and its impacts on health of population does not have a high standing in these two plans (GECZ, 2017; GED, 2016). Though these plans call for more comprehensive strategies, they do not refer specifically to the issues of excessive extractions through over-permitting and illegal extractions of water of the PLR aquifer.

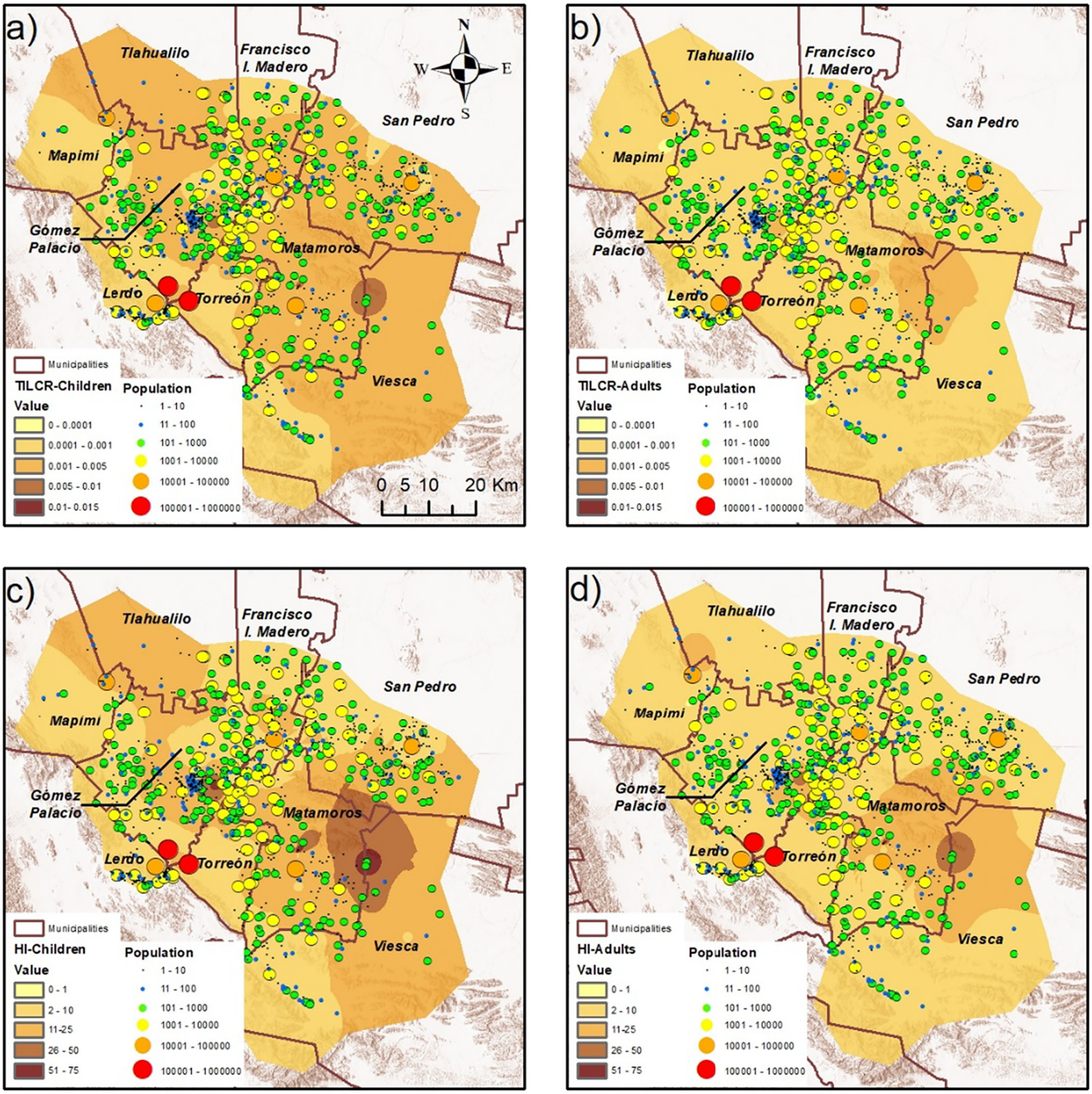

Fig. 4. Spatial distribution of risks of ingestion of As-concentrated water according to the Monte Carlo model: carcinogenic risk for children (a), carcinogenic risk for adults (b), non-carcinogenic risk for children (c), and non-carcinogenic risk for adults (d). It shows that in general children were at a higher risk than adults. The most exposed municipalities were Matamoros (east), Viesca (north), Gómez Palacio (west), San Pedro (south), and Tlahualillo (southwest). Mapimi, Lerdo, Francisco I. Madero, and Torreón were less affected. Note: The spatial distribution of risks according to the deterministic model is shown in Fig. S4.

The case of health policy is even more striking. For instance, out of the 21 strategies included in the Coahuila state development plan for the health sector (GECZ, 2017, pp. 79–80), there is not a single one focused on As contamination in this region. Durango's development plan is not much different; in the diagnosis and vision for the future there is neither an explicit reference to the As problems nor to its impacts on population health. In the health policy section, this issue is addressed very generally.

This disregard for health risks and policy also applies to the municipal planning sphere, as shown in the cases of Torréon, Coahuila (GMT, 2019), and Gómez Palacio, Durango (GMGP, 2019), the two largest municipalities considered here to illustrate this point. In Torreón, the overexploitation of the PLR aquifer is recognized, but there is no reference to As contamination and its health impacts. In Gómez Palacio, nothing is mentioned about As concentrations and public health; not even water quality is mentioned. Meanwhile, health specialists have documented that these concentrations drive patterns in disease in the population (ECL, 2019; Moran Martínez and García Salcedo, 2016) (Table S9). In fact, the national medical profession (Núñez et al., 2015) and the academic world have long considered the Comarca Lagunera region as suffering from the highest levels of As in groundwater and human exposure in Mexico (e.g., Ortega-Guerrero, 2003).

Table 2
Access to water, risk exposure to As ingestion, poverty line and health services in the municipalities of the Principal Lagunera Region aquifer, 2020.

Municipalities of the PLR aquifer (according to whether they are metropolitan or not)	Population	Inhabitants without access to water from a public network (within housing units)[a]	Population without access to water from a public network (within housing units, in %)	Population without access to water from a public network (within housing units, as % of total the aquifer)	Number of inhabitants with income below poverty line[c]	Population with income below poverty line (in %)	Population with income below poverty line (as % of total for the aquifer)	Number of inhabitants without formal access to health services[a]	Population without formal access to health services (in %)	Population without formal access to health services (as % of total for the aquifer)	Average cancer risk exposure (children) due to As ingestion (per 10,000)[b]	Average cancer risk exposure (adults) due to As ingestion (per 10,000)[b]	Expected cancer cases (adults and children) due to water ingestion[d]
Laguna Metropolitan Zone													
Torreón (Coah)	720,848	1442	0.2	9	269,818	37.4	41.4	157,866	21.9	42.5	3.0	1.7	67
Gómez Palacio (Dgo)	372,750	1118	0.3	7	161,012	43.2	24.7	78,278	21	21.1	2.3	1.3	27
Lerdo (Dgo)	163,313	980	0.6	6.1	66,402	40.7	10.2	49,828	25	13.4	1.5	0.8	7
Matamoros (Coah)	118,337	1302	1.1	8.2	44,656	37.7	6.8	27,573	23.3	7.4	3.3	1.8	12
Subtotal / Average	*1,375,248*	*4842*	*0.35*	*30.3*	*541,888*	*39.4*	*83.1*	*313,545*	*22.8*	*84.4*	*2.5*	*1.4*	*113*
Non-Metropolitan Area													
San Pedro (Coah)	101,041	6366	6.3	39.9	52,999	52.4	8.1	27,483	27.2	7.4	4.6	2.5	14
Francisco I. Madero (Coah)	59,035	1063	1.8	6.7	23,781	40.3	3.6	12,988	22	3.5	5.0	2.6	9
Mapimi (Dgo)	26,932	1077	4	6.7	12,945	48.1	2	7164	26.6	1.9	1.5	0.8	1
Tlahualilo (Dgo)	21,143	571	2.7	3.6	9794	46.3	1.5	4736	22.4	1.3	7.1	3.9	5
Viesca (Coah)	20,305	2051	10.1	12.8	10,719	52.8	1.6	5645	27.8	1.5	4.8	2.7	3
Subtotal / Average	*228,456*	*11,128*	*4.9*	*69.7*	*110,238*	*48.3*	*16.8*	*58,016*	*25.4*	*15.6*	*4.6*	*2.5*	*32*
Total / Average (aquifer)	1,603,704	15.97	1	100	652,126	40.7	100	371,561	23.2	100	3.7	2.0	145

*Sources:*Population municipalities of Coahuila: http://www.cuentame.inegi.org.mx/monografias/informacion/coah/poblacion/default.aspx?tema=me&e=05
Population municipalities of Durango: http://www.cuentame.inegi.org.mx/monografias/informacion/dur/poblacion/default.aspx?tema=me&e=10
Access to water and health services: IRS 2020 - anexos (CONEVAL, 2021).
Risk exposure to ingestion of As: Authors (findings from this study, Fig. S3, Supplementary material).
Poverty Line: https://municipal-coneval.hub.arcgis.com/pages/bienestar-economico. The sources were consulted during the period 15.12.2021 to 27.12.2021.

[a] These figures are calculated based upon the percentages given. Official statistics come in percentage and not in absolute terms.
[b] Exposure to carcinogenic risks of children and adults. In general children are more exposed than adults.
[c] A person is below the poverty line when income is not enough to acquire a basket of basic food, goods and services.
[d] Calculated based on the total incremental lifetime cancer risk for adults and children, and assuming that 45 % of the population is drinking raw water.

4. Discussion

4.1. Processes driving As concentration

The origin of As in the study area has been debated by scholars for the past 30 years (Table S8). In this study, As concentrations were confirmed to be significantly and positively correlated with several parameters (Table S6). The inverse relationship between As and NO_3^- concentrations may be indicative of mixing between shallow, recently infiltrated water affected by surface processes with deeper, mineralized and geothermally impacted, older and hotter groundwater. Water that has abundant NO_3^- suppresses As concentrations through dilution and vice versa. Several thousand long-screened wells likely connect different geological formations and enable mixing processes driven by differential hydraulic heads between layers. The geographical coincidence of zones with elevated As concentrations (i.e. east of Matamoros and between the La Laguna Metropolitan area and Francisco I. Madero) with regional cones of depression below agricultural lands and elevated temperatures above 30 °C confirms the influence of geothermal fluids. Evidence of mixing in long-screened wells have been observed also in other Mexican aquifers and elsewhere (Huang et al., 2022; Izbicki et al., 2015; Knappett et al., 2020; Mayo, 2010).

Mejía et al. (2018) hypothesized that the reductive dissolution of Fe-oxyhydroxides containing As and higher As concentrations in deeper aquifer layers, may be considered as potential factors inducing the dissemination of As in this alluvial aquifer (Mora et al., 2021). Ortega-Guerrero (2003, 2017) argued that evaporation in the paleolakes in the southeastern and eastern portion and infiltration towards the aquifers is the main mechanism of As enrichment, while adsorption and co-precipitation of As on Fe-oxides, clay minerals and organic carbon represent the main controls.

Based on previous hypotheses and new evidence, a comprehensive geochemical conceptual model was developed, which explains the processes driving As accumulation in different parts of the aquifer system (Fig. 5). In the source areas in the western and northwestern portion (Bermejillo), high As concentrations were associated with the dissolution of calcareous sedimentary rocks (limestone, gypsum) and possibly oxidation of sulfide minerals (for example, samples 19, 21, and 22). In these areas, As concentrations correlate with salinity and are independent from pH. These waters provide a regional flow component. Meanwhile, in the basin-fill deposits underlying the Nazas river and irrigated areas (recent infiltration), groundwater has a lower salinity, and As concentrations were related to the absorption/desorption from ferric hydroxides and clayey-rich sediments (for example, samples 2, 3, 5, 9 and 23).

Here, the As concentrations are generally lower but increase with depth and pH. This is consistent with the findings of Aparicio González (2018) who observed a gradually increasing As concentration with depth along production wells as the aquifer condition transitioned from unconfined to semiconfined. The As concentration increase is due to the upwelling or regional flow from calcareous rocks induced by heavy pumping, and mixing with groundwater from the granular aquifer. Additional As enrichment is possible locally where the regional flow component in the fractured aquifer comes in contact with geothermal, silica-rich fluids. Finally, thick lacustrine, fine-grained sediments of the former Viesca and Mayran paleolakes adjacent to the eastern portion of the aquifer areas represent an important additional source of highly enriched As (<5000 mg/L) porewater due to past evaporative accumulation and desorption from clays and oxihydroxides.

In general, geothermal admixture and dissolution of evaporites represent the most important As sources, while ferric hydroxides and clayey-rich sediments control have a more minor influence on As concentrations in the aquifer. Steepening lateral and vertical hydraulic gradients caused by increasing groundwater abstractions may drive the continued release and migration of As to more parts of the aquifer.

4.2. From elevated As concentrations to health risks

An analysis of historical data demonstrates a gradual increase of median As concentration in groundwater from the region during the past six decades. On average, the most polluted municipalities are Gómez-Palacio and Viesca, while the least polluted municipalities are Torreón and Mapimi. As a result of As groundwater contamination, children and adults drinking raw water may be exposed to As concentrations up to almost 84 and 46 times the reference dose (dose assumed not to be associated with health

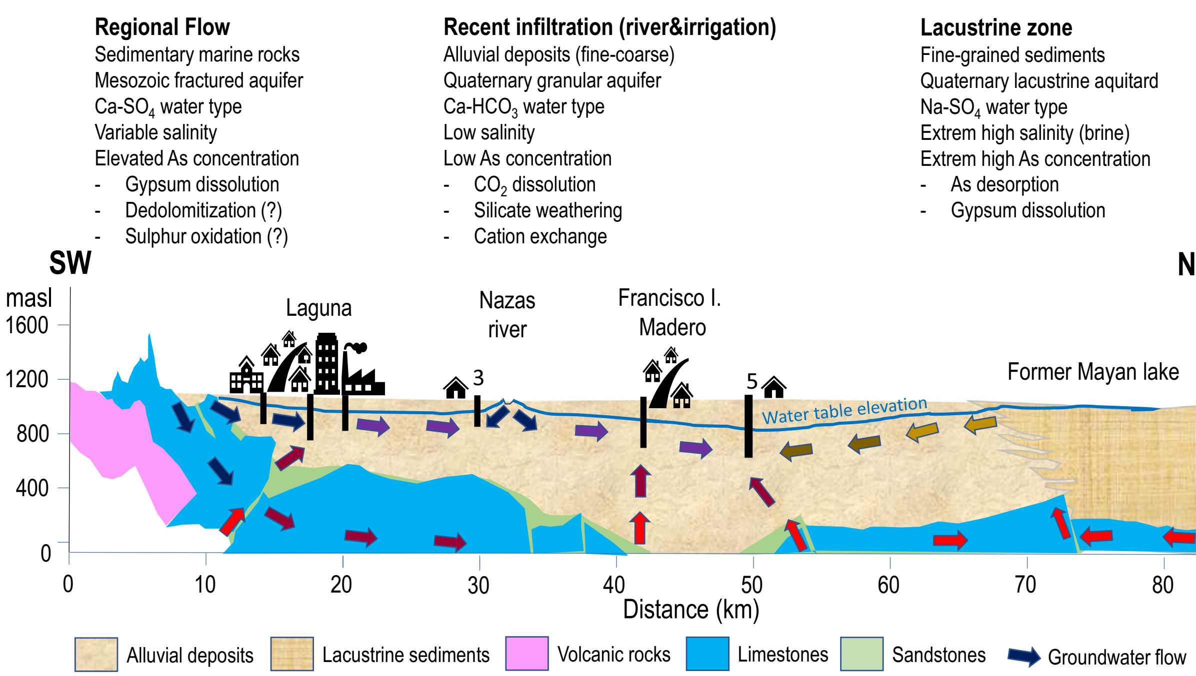

Fig. 5. Conceptual geochemical model of the PLR aquifer. Adapted and updated from Molina Maldonado (2004), Ortega-Guerrero (2017), and Aparicio González (2018). Note: The colour of the different flow pathways indicate the circulation/origin: blue = local and intermediate flow, red = regional flow with higher temperature, ocher = local and intermediate flow originated from lacustrine sediments.

effects), respectively. Furthermore, As showed important carcinogenic and non-carcinogenic health risks for both children and adults.

Carcinogenic risks were unacceptable in >90 % of the study area, with median exposures of 2 to 5 times the safety levels. According to the probabilistic model, children have higher non-carcinogenic and carcinogenic health risks derived from water As ingestion and dermal contact, which is in agreement with previously published evidence (Landrigan and Goldman, 2011). A combined exposure and socioeconomic needs approach showed that Gomez Palacio is most affected municipality due to both, highest health risks from drinking As polluted water, the largest number of potentially exposed people along with the greatest population of impoverished people without access to formal health services.

4.3. Poverty and health risks

It is relevant to consider the health risk of As exposure in the light of the poverty status of a large part of the population. When linking water provision, As health risk, poverty and health services, a metropolitan versus non-metropolitan pattern seems to emerge (Table 2 and Fig. 4). Non-metropolitan municipalities have less access to water, in general have higher As-driven carcinogenic risks, and a greater share of their population is below the poverty line and without formal access to health services. Furthermore, as found by a relatively recent study of the United Nations Development Programme, smaller municipalities are less able to cope with this adverse context (UNPD, 2019). This study found that Tlahualilo (Durango) and Viesca (Coahuila) have the highest risk levels (Table 2); about 50 % of their population live in poor conditions and thus less capable of buying bottled water. A jug of water (typically 20 L) in a rural area costs twice as much as in the cities (approximately 1 USD compared to 0.5 USD, respectively).

In Torreón, it is estimated that around 60–70 % of the population consumes bottled water from private establishments (*water houses*); this share is expected to be higher in rural areas. The consumption of bottled water has been an unregulated practice of 20 years (personal communications from local government officials). The municipality hosts between 400 and 600 *water houses*. In theory, the health sector and the municipalities should oversee the operations of these establishments but this does not happen in practice. Government agencies – from the federal to the local - have neither the capabilities nor the commitment to do so.

Even though metropolitan populations seem to be in better conditions regarding access to water, the bulk of the population below poverty line and with no formal access to health services reside in the metropolis. These findings suggest a combined approach is needed to address the exposure and risks of the most vulnerable population consuming water with As.

Preliminary results in Table 2 suggest that at least 145 cases are associated with arsenic exposure via drinking water, of which about 80 % are living in the metropolitan area; Torreón and Gómez Palacio are the most affected municipalities due to their high health risk from drinking As polluted water combined with a large number of people potentially exposed.

Given the context in the Comarca Lagunera region, perhaps here more than anywhere else in Mexico, a fundamental human right is bottled (Pacheco-Vega, 2017; Yaniz, 2016). This fundamental right to accessible, safe, and affordable water – as is stated in the Constitution and other legal norms – is not guaranteed. Buying bottled water has been the way inhabitants cover a basic need and protect their health. It has also been the authorities' strategy to address, without truly solving, a long-standing problem. People who can't afford bottled water, continue drinking supplied water with high As concentrations; it's either drinking it or drinking no water at all. As the local leading newspaper put it over ten years ago: people are more concerned about the lack of water than the presence of As (*El Siglo de Torreón*, October 10, 2010). These findings are consistent with the high consumption of bottled water found nationally in view of the poor quality of domestic water supply (Greene, 2018; Greene and Morvant-Roux, 2020; Pacheco-Vega, 2015, 2019).

4.4. Health risks of As into development and public health policy

Arsenic concentrations and their implications for development and health policy have remained largely disassociated, as clearly seen in the analysis of current federal, state and municipal development plans. This fact contrasts sharply with the official constitutional and other normative mandates towards a clean environment and good health for population.

While health authorities do not seem to play a substantial policy role in addressing the As problem, several civic organizations - such as *Encuentro Ciudadano Lagunero* and *Alianza Laguna por el Agua* - have taken approximately 300 judiciary procedures against different authorities, pressuring for more and better water quality (El Siglo de Torreón, 2022). Some federal judges have already ruled in favour of some of these organizations, forcing local water authorities to supply clean water. The National Commission of Human Rights and the Interamerican Commission of Human Rights are knowledgeable about these procedures, too.

4.5. Hard choices about normative and policy decisions

It has been difficult to translate the scientific evidence over As concentrations into policy design and implementation (Fisher et al., 2017). The country has recently established new drinking water standards, in line with the WHO guideline of 10 μg/L, to save lives. The new Mexican standard (SSA, 2021) prescribes a differenciated reduction of the permissible level of As in drinking water to 10 μg/L: for locations >500,000 habitants by May 2023, between 50,000 and 499,999 habitants by May 2025, and <50,000 habitants by May 2028.

Nonetheless, setting a stricter standard alone will not solve the problems presented here; the enforcement issue deserves special attention. Even if this stricter standard could be established, it has a limited chance of reducing human exposure to As in drinking water, as there are dilemmas to be addressed. Ultimately hard choices must be made. This situation has long been recognized by the federal Government itself (CONAGUA, 2009).

The extreme difficulties to regulate groundwater pumping and its quality could be seen as governance failure. The case of Guanajuato, central Mexico, for example has demonstrated that the effective regulation of groundwater use under the current legal, political and economic systems is intricate and improbable (Hoogesteger and Wester, 2017). Economic and regulatory incentives encourage overexploitation of groundwater. Even programs oriented to agricultural modernization end up generating more output and hence more - not less- water use. To a large extent, this is also the experience in the Comarca Lagunera region.

Population growth has been a driving force behind increasing water demand, but it is not the largest contributor. Sealing some wells used for domestic consumption, without touching the legal and illegal pumping for agriculture – by far the largest user – will accelerate the overdrafting of groundwater. This would be the continuation of the *business-as-usual* policy of the last seven decades. However, reducing this overpumping could have adverse economic impacts and face fierce political opposition by the largest and most powerful water users. In the end, the evidence points out to deficient groundwater management and the need for complementary coherent approaches.

In overexploited aquifers - like the PLR - the state has the authority to implement drilling bans (*veda*) and, in theory, the legal right to deny new water use permits, according to the national water law (SARH, 1992). Although the existing legal framework allows for strict state groundwater regulation, in practice, CONAGUA is unable to implement and supervise these regulations due to a lack of funds and personnel capacity, corruption and often contradicting policies (Hoogesteger and Wester, 2017).

Inadequate or insufficient funding for municipal water utilities has been an issue, in both urban and (especially) rural communities (Godinez Madrigal et al., 2018; McCulligh et al., 2020; Molle et al., 2018). There has been a traditional dependence on external support, mainly from state and federal government (through CONAGUA), for infrastructural investments to expand and maintain the supply networks and the deepening and/or repositioning of wells in overdrawn aquifers (Hoogesteger, 2018;

Molle and Closas, 2020a). But state government finances are below par, and the budget of CONAGUA has been decreasing over time (Casiano Flores et al., 2019; Molle and Closas, 2020b). Very little attention and funds have been allocated to deal with water quality problems beyond chlorination (Greene, 2018; Wilder and Romero Lankao, 2006).

The policy shortcomings found in the Comarca Lagunera region are consistent with those at the national level. The Mexican Congress Audit Office (ASF, 2019) on the country's drinking water sector for the period 2012–2018, found an absence of a sound, coordinated policy. This is the restrictive context for the presidential initiative around the program *Agua Saludable* (Healthy Water) (CONAGUA, 2022). *Agua Saludable* intends to conduct, store, and treat 200 Mm^3 from río Nazas, to supply As-free water to the region over the next 25 years. Critics point out that *Agua Saludable* is highly vulnerable because it relies on water from dams in a region prone to droughts. There are also financial and environmental concerns which have led to social and political unrest, leading to legal demands by non-governmental organizations to stop this program (El Siglo de Torreón, 2022).

5. Concluding remarks

This study analyzed the occurrence, sources and geochemical processes, as well as health impacts of As in the main aquifer of semiarid, agricultural Comarca Lagunera region. Extensive information about the long-standing groundwater As contamination problem has been analyzed, mapped and integrated. There seems to be a trend indicating that As contamination in the large aquifer system is exacerbated by the overextraction of groundwater, mainly for irrigation and to a minor extend for urban water supply, which started in the last century and is unceasingly reducing the volume of remaining fresh water. Heavy pumping provoked the mixing of deeper and geothermally influenced goundwater flows with more recently recharged water as well as the advancement of saline waters from adjacent paleolakes.

The main sources of As are likely calcareous sedimentary rocks (evaporites), while ferric hydroxides and clayey-rich sediments exert a lesser control on the release of As. To anticipate future changes and further constrain the geochemical processes responsible for high As concentrations, a seasonal time series sampling program should be conducted across a wide variety of well locations and screen depths for many years.

Arsenic exposure and health effects in different subpopulations of the Comarca Lagunera region have been well documented by researchers. The unacceptable As levels in >90 % of the study area and median exposures of 2 to 5 times the safety levels observed in this study are congruent with previous findings from field studies. Preliminary results suggest that at least 145 cases are expected to be associated with arsenic exposure via drinking water; the metropolitan area has a share of 80 % of this figure.

The high concentrations of As in the Laguna region – and their impact on public health is a case in point to illustrate the shortcomings of groundwater policy in Mexico. By delivering insufficient or contaminated drinking water, several entities do not comply with mandates contained in federal and state constitutions and planning laws, as well as those in the municipal sphere. It is very likely that the introduction of the new drinking water standard will only exacerbate the no-compliance case.

The intricated institutional landscape evidences disperse responsibilities, often in contradictory terms. There is a national legal and normative dysfunctional framework that is clearly observed in this case study. This makes difficult to design and implement coherent policy towards a more sustainable groundwater use and the implications this has on public health. Long-term solution to the As problem requires a more integrated water resources management and more orchestrated and coherent collective action.

A likely pathway would be to design a coherent transition program, from the current situation to a more responsible one, which places the health of the population and development at the center, making sure that the spatial differentiation between and within rural and metropolitan settings are taken into account. More specifically, this study's results provide a starting point to prevent and mitigate exposure to As and its health effects, since they pinpoint hotspots within the region and can serve to prioritize management based on the magnitude of the risk and the size of the affected population. In view of an overwhelming situation in the PLR that has led to a paralysis, the fragmentation of the problem along with solid diagnostic data can lead to a motivation of the stakeholders to engage in partial but achievable and corrective actions.

CRediT authorship contribution statement

Herewith we state that all authors participated in the development of the manuscript. In the following an accurate and detailed description of their diverse contributions to the work:

Jürgen Mahlknecht: Conceptualization, Study area description, Data curation, Data analysis, Investigation, Writing-Original draft preparation, Funding acquisition.

Ismael Aguilar-Barajas: Conceptualization, Investigation, Data curation, data analysis, Writing-Original draft preparation, Reviewing and editing.

Paulina Farias: Data analysis, Investigation, Writing-Original draft preparation.

Peter S. K. Knappett: Reviewing and editing, Language editing.

Juan Antonio Torres-Martínez: Visualization, Validation, Reviewing and editing.

René H. Lara: Data provision, Reviewing and editing.

Jaime Hoogesteger: Reviewing and editing, Language editing.

Ricardo A. Ramírez-Ambrosio: Validation, Reviewing and editing.

Abrahan Mora: Supervision, Validation, Reviewing and editing, Project administration, Funding acquisition.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The logistical support from Organismo de Cuencas Centrales del Norte (North Central Basin Authority) of CONAGUA during the sampling campaign and providing information from previous studies is acknowledged. Sistema Nacional de Investigadores (CONACyT) provided financial support to the Mexican authors. The authors would also like to thank Cristina L. Moreau-Tuero and Rogelio Ledesma-Ruiz for participation in the fieldwork.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.159347.

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