



Review

Occurrence and treatment of arsenic in groundwater and soil in northern Mexico and southwestern USA

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ABSTRACT

This review focuses on the occurrence and treatment of arsenic (As) in the arid region of northern Mexico (states of Chihuahua and Coahuila) and bordering states of the southwestern US (New Mexico, Arizona, and Texas), an area known for having high As concentrations. Information assembled and assessed includes the content and probable source of As in water, soil, and sediments and treatment methods that have been applied in the area. High As concentrations were found mainly in groundwater, their source being mostly from natural origin related to volcanic processes with significant anthropogenic contributions near mining and smelting of ores containing arsenic. The affinity of As for solid phases in alkaline conditions common to arid areas precludes it from being present in surface waters, accumulating instead in sediments and shifting its threat to its potential remobilization in reservoir sediments and irrigation waterways. Factors such as oxidation and pH that affect the mobility of As in the subsurface environment are mentioned. Independent of socio-demographic variables, nutritional status, and levels of blood lead, cognitive development in children is being affected when exposed to As. Treatments known to effectively reduce As content to safe drinking water levels as well as those that are capable of reducing As content in soils are discussed. Besides conventional methods, emergent technologies, such as phytoremediation, offer a viable solution to As contamination in drinking water.

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1. Introduction

Arsenic is one of the most feared contaminants because of its high toxicity at small concentrations and ability to go undetected (NRC, 1999). It is invisible, tasteless, and odor-less. Exposure to high levels of arsenic (As) can cause problems in humans ranging from gastrointestinal symptoms to arsenicosis, a chronic disease resulting from extended exposure to As, which occurs mainly via ingestion of water containing this metal. Once it is dissolved in water and ingested, As accumulates in the body. Contamination of groundwater with arsenic (As) is a major environmental and public health problem on a global scale. The World Health Organization (WHO) guideline for As in drinking water is $10 \mu\text{g L}^{-1}$ (WHO, 2004). This limit also applies in the US (since January 2006), India, Taiwan, Vietnam, and Japan (Mondal et al., 2006; Mohan and Pittman, 2007). In Bangladesh, China, and most of Latin-American countries the maximum contaminant level (MCL) is $50 \mu\text{g L}^{-1}$, while in Germany and Canada the limit is $25 \mu\text{g L}^{-1}$ (Mondal et al., 2006). In México, the MCL has been $25 \mu\text{g L}^{-1}$ since 2005 (SSA, 1999).

Well-known As contaminated regions include Bangladesh, India (Brinkel et al., 2009); Inner Mongolia, China (Guo et al., 2006; Wade et al., 2009; Xia et al., 2009); and eastern Croatia (Habuda-Stanic et al., 2007). In 2003 it was estimated that more than 50 million people in Bangladesh were drinking water with As concentrations exceeding the national standard of $50 \mu\text{g L}^{-1}$ (Mondal et al., 2006). Other regions recently reported with a high probability of As contamination include Central Austria, New Zealand, Northern Afghanistan, and Northern Mali and Zambia in Africa (Amini et al., 2008). Arsenic concentration levels of 300–4000 $\mu\text{g L}^{-1}$ have been reported in these regions, with concentrations higher in groundwater than in surface water most of the time. Countries like Thailand and Taiwan reported a large proportion of groundwater contaminated with As at levels from 100 to over 2000 $\mu\text{g L}^{-1}$ (Ning, 2002).

At least four million people drink water with unhealthy As levels in México and the Latin-American countries of Argentina, Chile, El Salvador, Peru, and Nicaragua (Bundschuh et al., 2006). Knowledge about As concentrations in Latin American groundwater has recently increased; however, additional efforts are required to complete the map of arsenic distribution and extend the study of soils and sediments (Litter et al., 2010). Argentina and México have the largest number of dispersed populations exposed, and arsenicosis represents a public health issue (Bundschuh et al., 2006; Armienta and Segovia, 2008). In México, sources of drinking water exceeding $50 \mu\text{g L}^{-1}$ have been found in Baja California Sur, Chihuahua (Delicias, Meoqui, Julimes), Coahuila (Torreón, Santa Ana), Nuevo León (Monterrey), Durango (Gomez Palacio), Zacatecas, Hidalgo, Morelos, Guanajuato (Acámbaro), Sonora (Hermosillo, Guaymas, Obregon), and San Luis Potosi (Villa de la Paz-Matehuala) (Cebrián et al., 1983; Garcia-Vargas et al., 1991; Wyatt et al., 1998; Carrillo-Chavez et al., 2000; Alarcón-Herrera et al., 2001; Razo et al., 2004; Martin-Romero et al., 2006; Ruiz-González and Mahlnecht, 2006). Regions with high probability of As contamination in México, Chile, Argentina, and the southwestern United States have been suggested in literature (Amini et al., 2008).

The US Environmental Protection Agency (EPA) has estimated that about 13 million people in the US, mostly in the western states, are exposed to As in drinking water at levels above

$10 \mu\text{g L}^{-1}$; of those 12 millions, 2.5 million are exposed to $25 \mu\text{g L}^{-1}$ (USEPA, 2001). Comprehensive maps of groundwater occurrence of As in the US are found in Focazio et al. (1999) and Ryker (2002), who attribute As content to factors such as water-use patterns, well construction, and local geochemistry. Other factors such as pH and Eh conditions, which in turn are governed by geology, climate, drainage and topography, also affect the As content in water (Amini et al., 2008).

Arsenic is usually distributed as water-soluble species, colloids, suspended forms and sedimentary phases (Peng et al., 2009). Sediments usually act as carriers and hence potential sources for metals in aquatic environment (Peng et al., 2009). Mobilized arsenic is most likely transported by water and accumulated in downstream river sediment as a result of the great affinity of As to solid phases. The most common sources of non-naturally occurring As worldwide arises from the presence of alloys used in the manufacture of transistors, laser and semi-conductors (WHO, 2004).

Humans exposed to excessive As concentrations develop respiratory, gastrointestinal, hematologic, hepatic, renal, dermic, neurologic, and immunologic effects (Rosado et al., 2007). Arsenic can be detrimental to central nervous system and cognitive development in children (Rosado et al., 2007). It also accumulates in fingernails and hair (Choong et al., 2007). High dermic toxicity has been reported in some countries around the world (Mondal et al., 2006; Choong et al., 2007). Le et al. (2004) identified more than 20 As compounds present in the natural environment and biological systems. As (V) is a molecular analog of phosphate and inhibits oxidative phosphorylation thereby short-circuiting life's main energy-generation system (Mondal et al., 2006). As (III) binds to sulfhydryl groups impairing the function of many proteins and affects respiration by binding to the vicinal thiols in pyruvate dehydrogenase and 2-oxoglutarate dehydrogenase (Mondal et al., 2006).

According to the WHO, it is technically feasible to achieve As concentrations of $5 \mu\text{g L}^{-1}$ but this requires careful process optimization and control, while a more reasonable expectation is that $10 \mu\text{g L}^{-1}$ should be achievable by conventional treatment (WHO, 2004). In the US, the EPA reduced the MCL for As from $50 \mu\text{g L}^{-1}$ to $10 \mu\text{g L}^{-1}$ and mandated that all community and non-transient, non-community water systems comply with the new standard by January 23, 2006 (USEPA, 2003). The majority of utilities that had been previously in compliance with the EPA regulations had to install new and/or modify existing As removal systems to meet the new MCL (USEPA, 2006b).

This review focuses on the occurrence, mobility, toxicity, and treatment of As in the arid region of the states of Chihuahua, Coahuila and Durango in northern México and the bordering states of New Mexico, Arizona, and western Texas of the southwestern US (Fig. 1). Treatment methods known to be effective in reducing As content to safe drinking water levels as well as innovative technologies that have been applied in the region are also discussed.

The compilation of information on As content in water, soils, and sediments provided here is intended to produce a clearer picture of As contamination in the study region, of its source and of its threat to human health. The study area, in parts or as a whole, is commonly recognized as having high As content (Robertson, 1989; Del Razo et al., 1990; Armienta, 2003; Frost et al., 2003). Previous studies conducted elsewhere showed that As concentrations in water and soil are far from being the same throughout a large area, but are instead constrained to certain lithologies, environ-

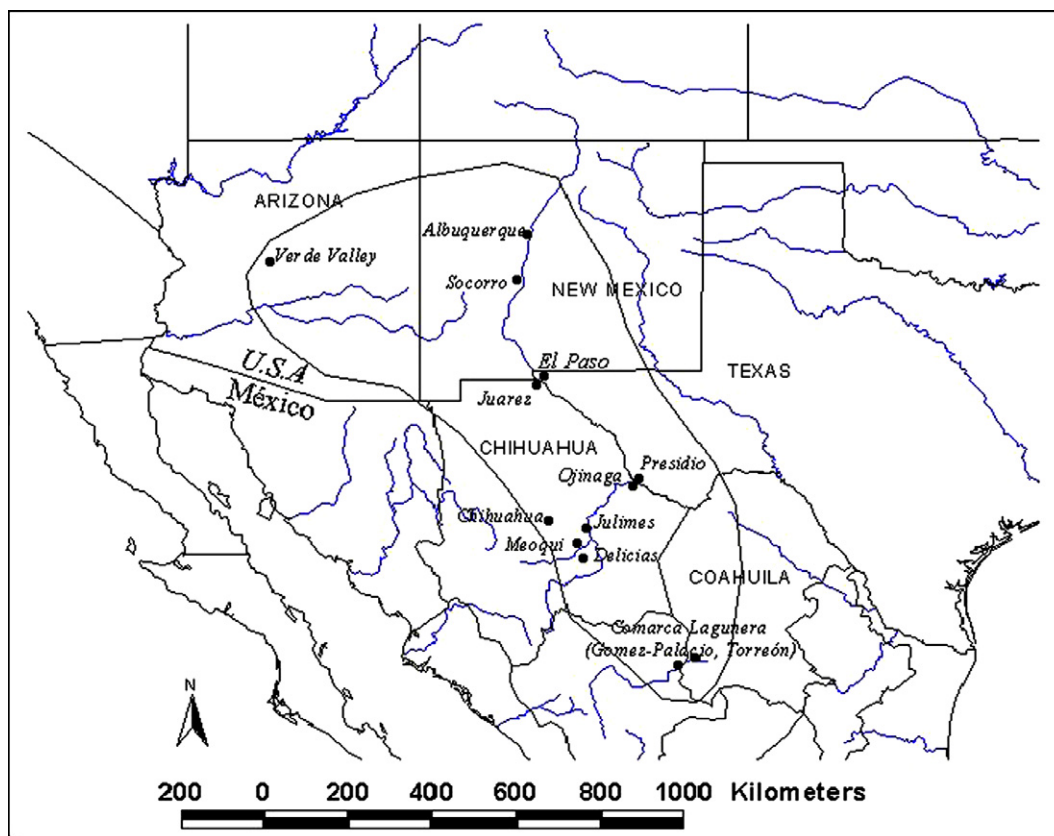


Fig. 1. Study area showing locations of interest.

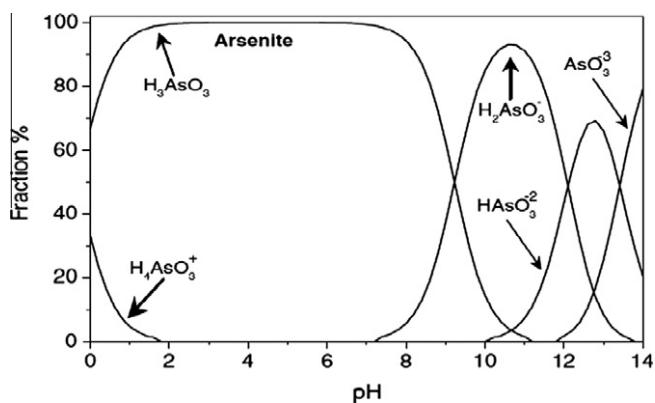


Fig. 2. Arsenic speciation as a function of pH for total As(III) (concentration 50 mg L^{-1}) (Vaklavikova et al., 2008).

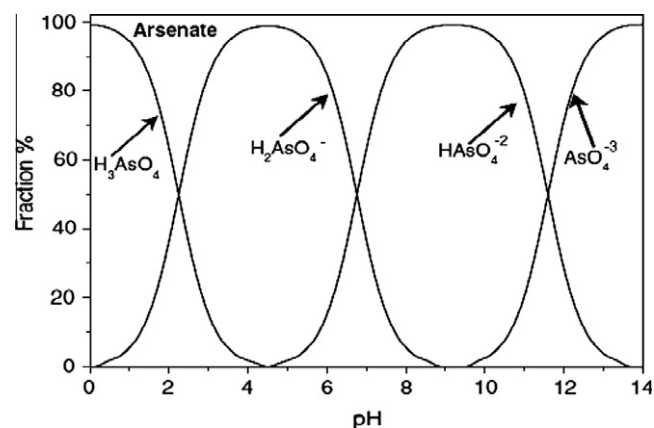


Fig. 3. Arsenic speciation as a function of pH for total As(V) (concentration 50 mg L^{-1}) (Vaklavikova et al., 2008).

mental conditions, and human contributions (Welch et al., 2000a; Smedley and Kinniburgh, 2002). Population and industrial growth during the past few decades has put a strain in the already scarce water resources of the region (HARC, 2000; Kelly, 2001; Bexfield and Plummer, 2003). Increased stream water diversion for water supply and the lowering of water tables may lessen the flow of streams enough to affect their dilution effect, therefore renewing concerns about As contamination in the region.

Within the region of study two areas of reportedly high As content are the Comarca Lagunera in México and the central part of the state of New Mexico. Both areas of study have similar geological and physiographical characteristics throughout. They are largely located within the Chihuahuan Desert, an elevated plateau with vast plains and low precipitation. Although various other sites

within these regions have also been tested for As content in either water or soil, they have not been studied as thoroughly as the two aforementioned regions, resulting in significant gaps of knowledge about the extent of As contamination. Soto et al. (2010) identified three main hydrogeological environments in México for arsenic's groundwater occurrences, namely alluvial aquifers (northern México), areas of mining activities (north-central México) and geothermal waters associated with recent volcanic rocks (central México).

1.1. Arsenic chemistry

The dominant forms of As present in the environment are arsenite (As(III)) and arsenate (As(V)) (Haque and Johannesson, 2006). They can form inorganic or organic compounds. Inorganic com-

pounds include hydrides, halides, oxides, acids, and sulphides. The most common organic compounds in water are methyl and dimethyl compounds (Vaklavikova et al., 2008). As(V) is most commonly found in surface water while As(III) is most frequently present in anaerobic ground waters (Vaklavikova et al., 2008; USEPA, 2000). Redox and pH conditions control the speciation of As (Sharma and Sohn, 2009). Typical As(V) species present under oxidizing aerobic conditions are the mono- and divalent oxyanionic forms H_2AsO_4^- and $\text{H}_2\text{AsO}_4^{2-}$ (Fig. 2), respectively, while the neutral As(III) species HAsO_2 is present in waters of pH lower than 9.0 (Fig. 3). At a pH of 5.0–8.0, which is typical of natural waters, As(V) exists as an anion, while As(III) remains fully protonated and is present as a neutral species (Fierro et al., 2009); thus As(V) is better sorbed on most media than As(III) (Mohan and Pittman, 2007; Welch et al., 2000a). Sharma and Sohn (2009) gives a comprehensive description of concentrations and speciation of As as affected by pH, Eh, organic matter, key inorganic substances, such as phosphate and sulfide, and adsorbents. The As(III) species predominates under moderate reducing conditions in sediments, groundwater and soils. The toxicity and mobility of arsenic depends on its speciation; As(V) is less toxic to humans and more readily removed than As(III).

1.2. Arsenic mobility

Arsenic mobility depends on the redox properties of the aquatic environment. Reducing aquatic environments are typical in regions rich in organic matter. Under this conditions it is common to find high microbial activity and low oxygen concentrations (Amini et al., 2008). Reducing regions may contain high sulfate concentrations and therefore low dissolved arsenic due to microbial sulfate reduction that will provide the conditions to precipitate As as a sulfide.

Oxidizing aquatic environments are found in arid and semi-arid regions, like the region of study, with high evaporation rates. High salinity and high pH are typical in these regions (Amini et al., 2008). The presence of soluble arsenate (AsO_4^{3-}) is common in oxidizing regions (Amini et al., 2008). Both reducing and oxidizing environments may be found within an aquifer. Arsenic dissolves in the presence of water. In the absence of oxygen, i.e., deeper aquifers, it occurs in the reduced form As(III) (also reported as H_2AsO_3^- or arsenite) while in the oxidized part of the aquifer As takes the less toxic form of As(V) (HAsO_4^{2-} , arsenate) (Das et al., 1994; Smedley and Kinniburgh, 2002). The difference between the two As forms has been attributed to an oxygen double bond in the As(V) molecule which influences its ability to dissociate and cause an increase in pH (Prabhu et al., 2003).

The presence of organic compounds and oxidation–reduction products affect the dissolution and sorption of As (Robertson, 1989; Sracek et al., 2004; Peng et al., 2009). Water table fluctuations also affect As mobility (Das et al., 1994). Since the mobility of As is determined by its oxidation state, the behavior of As will change depending on the biotic and abiotic conditions of the water (Vaklavikova et al., 2008). At the same time, inorganic As forms are more mobile than organoarsenic species.

Arsenic has a strong affinity for solid phases (i.e., soil) and binds readily to iron oxides. Surface water with low carbonate concentration that subsequently comes in contact with groundwater that has high carbonate content results in bicarbonate ions displacing HAsO_4^{2-} sorbed in aquifer oxyhydroxides (Szramek et al., 2004). Surprisingly, no correlation between As and Fe was found in either study. This lack of direct correlation suggests that other processes besides sorption and desorption of As in iron oxides are involved in the mobilization of As. Some of these may be the As-release mechanism from iron oxides that has been related to chelating agents secreted by bacteria (Kalinowsky et al., 2000), the ability

of As(III) to form complexes with extremely rich-carbonate waters (Neuberger and Helz, 2005), and the presence of natural organic matter in soils and sediments (Wang and Mulligan, 2006). Different arsenic mobilization theories based on the combination of chemical, physical and microbial factors are presented elsewhere (Mondal et al., 2006).

Difficulty in identifying As sources and understanding its mobility in groundwater and its chronic poisoning capability have renewed the interest in this toxic element worldwide (Das et al., 1994). Numerous studies on the overall occurrence, toxicity, and remediation of As have been conducted in recent years; noteworthy are the review articles by Smedley and Kinniburgh (2002), Sofuoglu et al. (2003), Duker et al. (2005), Arsenic Exposure and Health Effects Conference Proceedings (Abernathy et al., 1997; Chappell et al., 1999, 2001, 2003), Proceedings of The International Congress on Natural Arsenic in Groundwaters of Latin America (Bundschuh, 2006), and Proceedings of the Second Arsenic International Congress on Arsenic in the Environment, Valencia, España, May 2008.

With the exception of a few studies (i.e., Robertson, 1989; Razo et al., 2004), the mobility of As in the area of study has not been studied as thoroughly as in other areas of known high-As content. Although the mobility has been studied extensively for a variety of geologic materials in arid regions elsewhere, more investigations are needed to obtain a better insight on the local processes affecting the transport and fate of As.

2. Arsenic occurrences in the area of study

The most influencing environmental variables of As occurrences are soil pH, hydraulic conductivity, soil type, aquifer type, well depth, elevation, sand, silt and clay fractions, mean annual precipitation, temperature, and land use (Twarakavi and Kaluarachchi, 2006). Other factors such as evaporation, phosphate concentration, and presence of iron oxides and sulfides are also important. In the area of study the effect of these variables will depend on the source of As, i.e. naturally occurring As in water and soil and As from anthropogenic origin.

2.1. Natural occurrences

The natural occurrence of As in the study area has been attributed to a set of geological conditions rather than a particular type of geologic formation (Welch et al., 1988). These conditions consist of one or more lacustrine sediments, As-bearing mineral deposits, and volcanic rocks; the later along with cold and hot springs related to volcanic activity.

Arsenic bearing rocks are an important source of As in this area (Robertson, 1989; Rosas et al., 1999; Bexfield and Plummer, 2003) and contribute to the As content of groundwater. Many of the surrounding mountains contain Cretaceous to Early Miocene volcanic rocks where hydrothermal deposits formed as a result of magmatic-hydrothermal processes (Clark et al., 1982). These deposits contain lead and zinc mineralization with some gold and silver to which As shows a close association (Smedley and Kinniburgh, 2002).

The presence of As in the Comarca Lagunera, México, is attributed either to a hydrothermal system with high contents of lithium, boron, Arsenic, and fluoride or to mobilization of As from the aquifer clay to the groundwater, due to desorption of As retained on clay (Armienta and Segovia, 2008). Geochemical modeling conducted by Gutiérrez-Ojeda (2009) showed that, under natural conditions, the evaporated surface water carried by the Nazas and Aguanaval rivers may have contributed to the elevated As concentrations found in the lower parts of the alluvial aquifer in the Lagunera region of northern México.

Table 1
Total arsenic in water on selected locations.

Location	Sampling medium, No. samples	Mean, range of values ($\mu\text{g L}^{-1}$)	Reference
Comarca Lagunera, MX	Groundwater, 129	^a n/a, 8–624	Del Razo et al. (1993)
Comarca Lagunera, MX	Groundwater, 73	41, 7–740	Rosas et al. (1999)
Chihuahua, MX	Hot spring, 60	n/a, 110–191	Flores-Tavizón et al. (2003)
Meoqui, MX	Well water, one batch	n/a, 75–134 n/a, 50–277	Piñón-Miramontes et al. (2003)
Julimes, MX	Groundwater, 10	n/a, 10–376	Espino-Valdés et al. (2009)
Meoqui, MX	Groundwater, 20	n/a, 15–130	Espino-Valdés et al. (2009)
Rosales, MX	Groundwater, 12	n/a, 15–72	Espino-Valdés et al. (2009)
Delicias, MX	Groundwater, 19	n/a, 300	Espino-Valdés et al. (2009)
Chihuahua, MX	Well water, one batch		Fierro et al. (2009)
Río Puerco, NM	River water, 4	111, 21–193	Branvold and Branvold (1990)
Río Salado, NM	River water, 3	346, 190–503	Branvold and Branvold (1990)
Socorro Basin, NM			
	Groundwater, 74	9.5, <2–43	Branvold (2001)
Middle Rio Grande Basin, NM	Groundwater, 288	5.2 ^b , <1–600	Bexfield and Plummer (2003)
Travertine Mound and Soda Dam, NM			
	Hot spring, 6	1302, 680–1880	Reid et al. (2003)
Jemez River, NM	Surface water, 26	57.9, 2–300	Reid et al. (2003)
Hillsboro mining district, NM	Mine waste rock pile, 4 ^a	n/a, 2–180	Munroe et al. (1999)
Verde Valley, AZ	Groundwater, 40	n/a, 10–48	Foust et al. (2004)
Verde Valley, AZ (Montezuma well)	Spring, 1	210	Foust et al. (2004)
Verde Valley, AZ	Groundwater, 456	16, <2–1300	Robertson (1989)
El Paso-Presidio, TX	River water, 7 ^c	6.9, 4.7–10.1	IBWC (1997)
El Paso-Presidio, TX	River water, 8 ^c	6.1, <0.9–11.0	IBWC (2004)
El Paso, TX	Surface water, 7	n/a, 4–22	Rios-Arana et al. (2003)
Carrizo Sand Aquifer, TX	Groundwater, 10	n/a, 0.12–1.99 nmol/kg	Haque and Johannesson (2006)

^a n/a = not available.

^b Average of 13 water quality zones.

^c No. of stations.

Arsenic is a naturally occurring element in the southwestern US state of Arizona. Its presence may be due to natural dissolution/desorption reactions in geothermal water and to mining activities. The state was ranked highest in As risk by the National Human Exposure and Assessment Survey (NHEXAS) because of high As residue concentrations in tap and non-tap drinking water (Jones and Joy, 2006). Groundwater As concentrations greater than $50 \mu\text{g L}^{-1}$ have been identified in the interior of many basins in southern and central Arizona, including the Safford and San Pedro basins in southeastern Arizona (Robertson, 1989). In the Socorro Valley of Central New Mexico, high concentrations of As in groundwater are associated with thermal springs (Branvold, 2001).

2.2. Arsenic in water

Studies reporting high concentrations of As in water within the study area are listed in Table 1. Most of the studies on water report concentrations of less than $10 \mu\text{g L}^{-1}$ As in all but few locations. Locations showing high As content have been studied in detail to determine the source and the mobility of As. Since wells supply most of the drinking water to this area, most large cities are equipped with filtration and reverse osmosis treatment systems, and, thus, As and other contaminants are removed to safe levels. In contrast, rural wells, farms, and small communities generally have inadequate, if any, treatment systems, thus exposed to a higher risk of developing As related health problems.

High levels of As have been found in several natural sources in the northern arid and semi-arid regions of México. The state of Chihuahua, among others, is one in which As has been detected in sources of natural drinking water (Flores-Tavizón et al., 2003). Within the Comarca Lagunera, arsenicosis cases that appeared in the 1960's prompted numerous studies which pointed to the local groundwater as the immediate source of As (Cebrián et al., 1994; Hernández-Zavala et al., 1998). Del Razo et al. (1993) reported As concentrations above $50 \mu\text{g L}^{-1}$ in 50% of the wells sampled, and similar results were found by Rosas et al. (1999). In both studies, the predominant species was As(V). The trivalent form (As(III)) is

more toxic than the pentavalent form (As(V)). Although it is reasonable to assume that As(V) would be present in most water supply sources due to aeration and chlorination practices (USEPA, 1985), toxic trivalent forms have been found with the pentavalent forms in the Comarca Lagunera (Del Razo et al., 1990).

Another study conducted in the Comarca Lagunera (Del Razo et al., 2002) reported that the water exceeded eight times the As MCL established in México ($25 \mu\text{g L}^{-1}$) and that hot beverages (hot plain water, tea and/or coffee) had an As concentration 20% greater than plain tap water, even when the boiling process did not transform the inorganic form of As to an organic one. Del Razo et al. (2002) suggest that As present in food is dependent on water amount and cooking time. A survey of 58 wells of the region reported As concentrations higher than $700 \mu\text{g L}^{-1}$ (Molina, 2004).

In Chihuahua, high As concentrations have been reported for the Delicias–Meoqui and Jimenez–Camargo aquifers. A study conducted by Comisión Nacional del Agua (CNA) reported 50% of the wells having As concentrations higher than $50 \mu\text{g L}^{-1}$, with the highest concentrations found in the towns of Julimes and Aldama (CNA, 1996). Wells in nearby Delicias contained up to $450 \mu\text{g L}^{-1}$ As (Ruiz-González and Mahlknecht, 2006). Faults present in the area with a NW–SE trend were believed to be the conduit for the As found in the underlying geologic material. These faults are parallel and extend over an area that includes Delicias, Meoqui, Jiménez, and Camargo, covering an approximate length of 200 km (Vega-Gleason, 2001). Recently Espino-Valdés et al. (2009) reported As concentrations higher than the MCL of $25 \mu\text{g L}^{-1}$ in 72% of 61 drinking water wells tested in the Meoqui–Delicias aquifer (Table 1). This aquifer is the main water source for Julimes, Meoqui, Rosales, and Delicias. The presence of the metal is attributed to geogenic sources related to the recharge flow coming from arsenopyrite mineral deposits of surrounding mountains and the contact with sediments accumulated in the aquifer as well as to the upflow of geothermal water.

Farther south in Valle del Guadiana, in the state of Durango, 59% of wells supplying drinking water contained more than $50 \mu\text{g L}^{-1}$, some with concentrations of up to $167 \mu\text{g L}^{-1}$. Almost all wells in

this region exceeded the WHO MCL of $10 \mu\text{g L}^{-1}$ As (Alarcón Herrera et al., 2001). The presence of As is attributed to the geological composition of the aquifer, which is mainly of volcanic origin.

In central New Mexico, surface and groundwater levels exceeding $50 \mu\text{g L}^{-1}$ have been reported (Stanton et al., 1999; Dunbar et al., 2002; Bexfield and Plummer, 2003); they attributed mainly to the input of geothermal waters or derived from As-rich volcanic rocks such as those found in the Jemez Mountains (Reid et al., 2003). It is estimated that 16% of wells in New Mexico exceed the MCL of $10 \mu\text{g L}^{-1}$ As (BEG, 2005). Most of the As contamination is focused in the Middle Rio Grande Basin. In Texas, groundwater As contamination is widespread, with approximately 6% of wells exceeding the EPA MCL (BEG, 2005). In the El Paso (US)–Juárez (México) border area, wells from the Hueco Bolson aquifer tested below the $10 \mu\text{g L}^{-1}$ MCL while wells in the adjacent Mesilla aquifer had an average of $20 \mu\text{g L}^{-1}$ As, a value similar to the average of $16.6 \mu\text{g L}^{-1}$ found in Juárez (Benitez-Marquez, 2005). About 20% of the wells in the El Paso region exceeded the $10 \mu\text{g L}^{-1}$ MCL for drinking water, with As found in both the trivalent (As(III)) and pentavalent (As(V)) forms (Benitez-Marquez, 2005). In Arizona, values reported for well water (10 – $210 \mu\text{g L}^{-1}$) suggest that groundwater is less affected by mining activities than by naturally occurring As present at specific locations (Foust et al., 2004).

Although less relevant for human health, because they does not represent a large percentage of drinking water supply, surface waters in the area have also been tested for As content. The alkaline conditions of most surface waters and the affinity of As to attach itself to solid phases at these conditions result in a small concentration of As in most rivers and water bodies of the area, with As $> 10 \mu\text{g L}^{-1}$ in only about 10% of the sampled sites along the Rio Grande (International Boundary Water Commission (IBWC), 1994, 1997, 2004).

Even though the majority of the studies conducted in the area of interest indicate high As concentrations, a few studies report As levels below the established MCL. For example, studies conducted by Haque and Johannesson (2006) on As concentration and speciation analyses in production wells along a flow path in the Carrizo Sand aquifer in Southeastern Texas found that the metal concentrations (0.37 – 2.5 nmol L^{-1}) were below the levels recommended by the World Health Organization (WHO) and the MCL established by the US EPA in drinking water ($10 \mu\text{g L}^{-1}$). They found that As(V) predominates in groundwater within the first 15 km and beyond 66 km of the flow path, whereas As(III) predominates between 15 and 59 km. The presence of As(V) in the upper 15 km of the aquifer was attributed to the reduction of Fe(III) oxyhydroxides/oxides and subsequent release of adsorbent and/or coprecipitated As(V), followed by reabsorption of As(V) in the mid-reaches of the aquifer. The observed increase in As(V) further down-gradient was attributed to a pH related desorption as pH increased to 8.5. The Carrizo Sand aquifer is considered a relatively pristine and unconsolidated sedimentary aquifer (Haque and Johannesson, 2006). Arsenic concentrations ranging from less than 5 to $50 \mu\text{g L}^{-1}$ have been reported in the Rio Verde Basin, which is located in the southeastern section of the study area (Planer-Friedrich et al., 2001).

A more recent study reported elevated groundwater As levels widespread in the southern part of the Southern High Plains (SHP-S) aquifer in Texas as compared to the As levels in the north part (SHP-N) (Scanlon et al., 2009). The current EPA MCL of 10 was exceeded by 47% of the wells in the SHP-N, with maximum concentrations of $164 \mu\text{g L}^{-1}$, whereas EPA MCL was exceeded by only 9% of the wells in the SHP-N, with maximum concentrations of $43 \mu\text{g L}^{-1}$. In the study data provided by different sources for a total of 1522 was analyzed. The analytical method used most of the times was Inductively Coupled Plasma Spectroscopy (ICP-MS), following EPA method 200–7. The contrast in As levels between the

north and south was attributed to changes in total dissolved solids (TDS) from 395 mg L^{-1} to 885 mg L^{-1} , respectively. Based on speciation analysis, As(V) was present in this oxidizing, semi-arid system and even though it was correlated to the groundwater TDS, it could not be attributed to evaporative concentration, a property typical of this type of environment. Groundwater was classified as oxidizing based on the presence of dissolved oxygen (DO), SO_4^- and NO_3^- . High TDS in the SHP aquifer was related to upward movement of saline water from the underlying Triassic Dockum aquifer. High pH levels typical of semi-arid regions could not be attributed to the mobilization of As in the aquifer since it was found to be near neutral (7.0–7.6). The most possible reason for the regional As distribution and correlation with TDS was attributed to the counterion effect caused by a change from Ca- to Na-rich water which is related to the upward movement of saline water from the underlying Dockum aquifer. The primary source of As in this area was attributed to the presence of either volcanic ashes in the SHP aquifer or original source rocks in the Rockies; the secondary source was attributed to sorbed As onto hydrous metal oxides that are competing for sorption sites (Scanlon et al., 2009). Salinization of the Rio Grande, with TDS concentrations as high as 1000 mg L^{-1} , has been considered a water-quality problem of greatest concern (USGS, 2010). Concentrations of dissolved solids from season to season in the Rio Grande between Caballo Dam and El Paso are commonly twice as high during the nonirrigation seasons as compared with concentrations during irrigation seasons (USGS, 2010). An increase in concentrations from 60 mg L^{-1} to 2000 mg L^{-1} was reported on the Rio Conchos at its headwaters upon receiving irrigation drain returns; however, the salinity decreases as the Rio Conchos confluences with the Rio Grande (Gutierrez and Borrego, 1999). A study conducted elsewhere suggests that As in groundwater wells has a positive correlation not only with sodium and alkalinity (HCO_3^-), but also with fluoride and trace elements such as uranium, molybdenum and vanadium (Rango et al., 2010).

2.3. Arsenic in soils and sediments

The spatial distribution of As in rock, soil and sediment can be found in digital data sets: the National Uranium Resource Evaluation Database (NURE), from the US Department of Energy, for the US and in a series of data sets for $1^\circ \times 2^\circ$ quadrangles, available from the Servicio Geológico Mexicano, previously known as Consejo de Recursos Minerales, for México. NURE geochemical data was originally collected for uranium resources within the US, but later broadened to include concentrations for 59 elements found in stream sediments, soils, surface waters and ground waters (Hoffman and Buttleman, 1994). The NURE data is however restricted to the eastern part of the Southern High Plains aquifer in Texas (2009, Scanlon).

Permissible As levels for soil vary according to the use of the soil and as set by the preliminary remedial goals (PRG) for contaminated soils (USEPA, 2004a). For As, these PRGs are 0.39 mg kg^{-1} for residential soil and 1.6 mg kg^{-1} for industrial soil with direct contact exposure. Values for soil screening are reported based on their dilution attenuation factor (DAF), which is the ratio of the concentration of pollutant in the soil and in the groundwater at the water supply well. The lowest possible value of DAF is 1.0, which means that there is no dilution or attenuation at all. High values of DAF correspond to a high degree of dilution and attenuation.

Studies reporting high concentrations of As within the area of interest are listed in Table 2. Sediments of the Rio Grande between El Paso and Presidio (Texas) were analyzed for As at five locations in 1992, 1995 and 1998, and concentrations ranged between 1.1 and 14.3 mg kg^{-1} (IBWC, 2004). Rios-Arana et al. (2003) measured

Table 2
Arsenic content in soil and sediments on selected locations.

Location	Sampling medium, No. samples, extractant	Median, range of values ($\mu\text{g g}^{-1}$)	References
Comarca Lagunera, MX	Soil, 50 HNO_3 – H_2SO_4 mixture	<i>n/a</i> , 11–30	Rosas et al. (1999)
Comarca Lagunera, MX	Roadside dust, 19 HNO_3	113, 79–297	Benin et al. (1999)
Chihuahua, MX	Roadside dust, 19 HNO_3	32, 14–96	Benin et al. (1999)
Julimes & San Diego de Alcalá, Chih., MX	Soil surrounding hot spring, HNO_3	46, 40–116	Flores-Tavizón et al. (2003)
Villa Matamoros, Chih., MX	Mine tailing site, HNO_3	63–82	Flores-Tavizón et al. (2003)
Río Conchos Basin	Sediment, 1, aqua regia	<i>n/a</i> , 7.3–24.9	Gutierrez et al. (2009)
El Paso-Presidio, TX	Sediment, 5 ^a SEM/AVS conc. HCl	7.2, 1.1–8.9	IBCW (2004)
El Paso-Presidio TX	Sediment, 6 ^a SEM/AVS conc. HCl	5.4, 1.9–14.3	IBWC (1997)
Río Conchos, MX	Sediment, 2 ^a SEM/AVS conc. HCl	8.1, 8.0–8.2	IBWC (1997)

^a Phase III; field samples collected in November 1998.

As in sediments of the Río Grande in the El Paso–Juárez region and found the highest amount, 2.32 mg kg^{-1} , at the American Dam which was attributed to past smelting activities. Higher values were reported for roadside dust near smelters and for soils surrounding hot springs. Soils within the Comarca Lagunera contained up to 30 mg kg^{-1} As; however, only 12%, or less, of the As was extractable (Rosas et al., 1999). The occurrence of As in soils from this area has been attributed to volcanic hydrothermal processes (Rosas et al., 1999; Armienta, 2003), although As in some areas is clearly influenced by human activities (smelters, refineries, fertilizers, and sewage) as described below.

Sediments from the Río Conchos Basin in Northern México were found to contain As levels in excess of the guideline value of 22 mg kg^{-1} for agricultural soils (Gutierrez et al., 2009) and also close to the 33 mg kg^{-1} limit for the onset of negative impact to aquatic organisms. The basin comprises the Conchos River and its tributaries, the Chuvíscar and San Pedro Rivers. Arsenic content in the basin is mainly associated with the presence of As-rich mineralization present in volcanic rocks. It is suggested that this basin may act as a sink for As, raising concern about potential future release of the metal from sediment into the water column (Gutierrez et al., 2009).

2.4. Anthropogenic sources

Anthropogenic sources of contamination occur when As is mined or concentrated and displaced by human controlled processes to locations where it is unwanted. Products such as wood preservatives, paints, alloys, semi-conductors, fossil fuel combustion, mine wastes, smelting, landfilling, sewage, and agricultural applications (pesticides and fertilizers) may also introduce As into the environment (USDHHS, 2000; Choong et al., 2007; Vakilovikova et al., 2008). Arsenic originating from anthropogenic activities is frequently detected in both sediments and aquifers (Peng et al., 2009).

Although industrial water constitutes a small percentage of the total water usage in the study area (Kelly, 2001; Dunbar et al., 2002), pesticides and sewage add to the total amount of As present in surface waters (Smedley and Kinniburgh, 2002). Moreover, As in pesticides and other agricultural and industrial products add to its accumulation in sediments of rivers and reservoirs from where, under the right conditions, it may remobilize back to the water column (Vázquez-Montiel et al., 1999; Norman and Dilley, 2002). Historically, the study area has had an important agricultural component with several irrigation centers (Río Grande Valley in New Mexico, and Districts of Juárez, Delicias, and the Comarca Lagunera) (Vázquez-Montiel et al., 1999; HARC, 2000; Kelly, 2001; Norman and Dilley, 2002). Organic arsenicals used as herbicides for cotton and other crops may be an important anthropogenic contribution of As to the environment in this region (USDHHS, 2000; BEG, 2005). Norman and Dilley (2002) studied irrigated soils in San Acacia, approximately 75 km south of Albu-

querque, New Mexico, and found that the soil contained 1.6 – 2.7 mg kg^{-1} labile As with As(V) as the primary species. They also reported anomalous concentrations of As(III) in drain water and attributed its presence to the application of phosphate-based fertilizers and/or insecticides. Phosphate ions replace sorbed As(V) ions and once the surface becomes saturated, As breakout occurs, resulting in water with high As concentrations (Norman and Dilley, 2002). High As concentrations in groundwater of northern Texas have been attributed to disposal of cotton gin waste, which has been found to contain As at levels up to 240 mg kg^{-1} (Aurelius, 1988). Groundwater at one site in Texas had As concentrations exceeding $2 \times 10^6 \mu\text{g L}^{-1}$ (Welch et al., 2000b).

Arsenic concentrations higher than the levels recommended by Mexican and other international agencies were reported in the San Pedro River in Chihuahua, México (Gutiérrez et al., 2008). This river is a tributary of the Conchos River, which joins the Río Grande/Río Bravo in Chihuahua, on the Mexican side and in Texas on the American side. The Río Grande/Río Bravo is a natural boundary between the United States of America and México. It is suggested that the human activities associated with the Conchos River (i.e., wastewater discharges and waste products from agriculture and industrial activities) are the main cause for the As contamination of the San Pedro River (Gutiérrez et al., 2008).

Sewage from Albuquerque, New Mexico, has also been identified as a significant source of As to the Río Grande (Dunbar et al., 2002). Interestingly, in the Mexican state of Aguascalientes, sewage from a major city diverted into a small reservoir enriched the As content in the sediments to a maximum of 12.6 mg kg^{-1} (Lara and Ortiz, 1999). In the Juárez Valley, it has been a practice to mix raw wastewater with water from the Río Grande before the water is further used downstream for irrigation. Vázquez-Montiel et al. (1999) found low concentrations of As and other metals in the water column of wastewater canals and suggested that metals in the water column must have sorbed onto suspended sediments which then settled, enriching the metals content of sediments in canals and rivers.

Studies of the impact of As in soils are scant within the area of interest, although concern about long-term impacts of smelter operations is apparent (Benin et al., 1999). Several mining and ore processing operations containing As are present in the area (Foust et al., 2004). Hard-rock mining activities at Taos, New Mexico, dating from the 19th century were found to adversely affect the Río Grande and its tributaries (USDHHS, 2005).

Elevated As levels in soil of the Anapra neighborhood in Juárez, México, were attributed to a lead smelter in El Paso, Texas, that operated until 1985. Measurement of As in the soil showed higher concentrations of 25.2 mg kg^{-1} , 21.4 mg kg^{-1} , and 19.5 mg kg^{-1} , compared to concentrations in soil located 25 km away from the smelter which had a mean concentration of 8.6 mg kg^{-1} (USDHHS, 2005). Benin et al. (1999) analyzed samples of roadside surface dust from residential neighborhoods near metal smelters in Torreon and Chihuahua and near a metal refinery in Monterrey,

México. They concluded that all three posed a health threat to people living nearby. Soils and dust collected in houses near smelters had concentrations that varied between 50 and 788 mg kg⁻¹, with a median of 113 mg kg⁻¹ (Table 2). The study reported that 37% of the samples in Chihuahua, 35% of the samples in Monterrey, and 89% of the samples in Torreon exceeded the maximum Superfund cleanup goal of 5–65 mg kg⁻¹ for As in soil (USEPA, 1997).

Munroe et al. (1999) conducted a study in four waste rock piles representing different types of mineral deposits in the Hillsboro mining district in Sierra County, New Mexico, to characterize their geochemistry. Mine waste rock piles (MWRPs) are anthropogenically created landforms in mining areas as a result of open pit and underground mining activities. He reported As concentrations within a range of 2.0 mg L⁻¹ to more than 150 mg L⁻¹, and found that the metal concentration increased as the size fraction of the waste rock pile decreased. He attributed this behavior to the mineralogical nature of the material, including clay and silt size particles containing high cation exchange capacity, and the high surface area available for sorption of ions. A potential run-off impact may exist in the area due to the inability of the ground to sorb the rain at the rate which it falls, thereby increasing the erosion of the MWRP and enabling higher chemical reactivity between the increased surface area of the pile and rain fall (Munroe et al., 1999). The annual mean precipitation at the Hillsboro station is 327 mm, with the highest rainfall amounts occurring during July, August, and September. Sampling procedures used in the Munroe et al. (1999) study to characterize the MWRP accounted only for horizontal, and not for vertical, chemical heterogeneities.

The toxicity characteristic leaching procedure (TCLP) and the synthetic precipitation leaching procedure (SPLP) were applied to a commercially waste-derived soil amendment marketed as Ironite, which is a mining waste from a former silver mine in the state of Arizona. SPLP concentrations of five samples exceeded the US hazardous waste toxicity characteristic (TC) limit for As (5 mg L⁻¹). The greater As leachability in the SPLP (1.9–8.2 mg L⁻¹) relative to the TCLP (4.8 mg L⁻¹) was explained as a result of lower pH conditions in the SPLP (Dubey and Townsend, 2004).

Total phase As concentrations as high as 22 µg L⁻¹ were found in Manada Creek, a urban tributary of the Rio Grande located in an industrialized area of Laredo, Texas, where warehouses, a major railroad, and a decommissioned antimony (Sb) smelter were located (Baeza et al., 2010). Samples were analyzed for the dissolved and total phases using ICP-MS. In this study six sites were selected along the creek with two located upstream and four downstream of the Sb smelter. In the southern High Plain elevated As concentrations in near surface soils related to nitrate and phosphate concentrations indicate a fertilizer or arsenical pesticide source (BEG, 2005).

Food is also a potential anthropogenic source of As contamination in the area of interest (Mora et al., 2001; Schmitt et al., 2005). Rice with high As concentrations has been found in the United States and around the world (Abedin et al., 2002; Zavala and Duxbury, 2008; Zavala et al., 2008). A study conducted by Zavala and Duxbury (2008) evaluated 204 commercial rice samples purchased in retail stores in upstate New York and supplemented with samples from other countries. They combined their data set with literature values to derive a global “normal” range of 0.08–0.20 mg kg⁻¹ for As concentration in rice. The study reported that the wide variability found in US rice grain was primarily influenced by region of growth rather than commercial type. Rice grown in Texas, a state within the area of study, showed significantly high mean As concentrations (0.258 and 0.19 mg kg⁻¹), with a Texas distributor having 75% of the samples above the global “normal” range. The authors suggest that the rice in that particular region is being produced in an As contaminated environment. Arsenic incorporation into the milk production chain in the Region Lagu-

nera was reported by Rosas et al. (1999). They reported As concentration in milk from cow from this region ranged from below 0.9 to 27.4 ng g⁻¹.

3. Arsenic toxicity in the area of study

Arsenic exposure to drinking water has been associated with relative risks of health. Arsenic carcinogenic can be present in nine different modes including chromosomal abnormalities, oxidative stress, altered DNA repair, altered DNA methylation patterns, altered growth factors, enhanced cell proliferation, promotion/progression, gene amplification, and suppression of p53 (Siegel, 2006). Only 10–30% of the As eliminated in urine by humans is in inorganic form, with 55–75% and 10–20% eliminated as dimethylarsenic acid (DMA) and monomethylarsenic acid (MMA), respectively. From this As forms the inorganic species As(III) is considered as the most toxic because it can bind to and inhibit the activity of at least 200 proteins (Siegel, 2006). The toxicity of As(III) is related to its high affinity for the sulfhydryl groups of biomolecules such as glutathione (GSH). Sulfide-activated As(V) could bind to the sulfhydryl group of GSH and cause toxic effects as well (Sharma and Sohn, 2009). DMA can act as either a promoter or complete carcinogen for bladder cancer (Siegel, 2006).

In the area of study health effects due to arsenic exposure are being reported in different ways. Symptoms of As poisoning observed in México are cutaneous manifestations (skin pigmentation changes, keratosis and skin cancer), peripheral vascular disease, gastrointestinal disturbances, and alteration in the coporphyrin/uroporphyrin excretion ratio. Although As exposure at high levels in the Region Lagunera, México, was associated with increased cancer incidence (Rosales-Castillo et al., 2004), a study conducted in Arizona showed no evidence of altered DNA oxidation and repair associated with tap water exposure to As at levels below 40 µg L⁻¹ (Burgess et al., 2007). Another study was conducted by Rosales-Castillo et al. (2004) in Region Lagunera, México, to assess the relationship between chronic As exposure, human papilloma virus (HPV) contact and non-melanoma skin cancer (NMSC). In this region arsenicosis is endemic. The study suggested that HPV infection could constitute an additional risk factor for NMSC development in humans chronically exposed to As. Evidences that inorganic As exposure may be diabetogenic are reported in a study conducted in Coahuila, México, by Coronado-González et al. (2007). An interesting application of As in humans was shown by Tollestrup et al. (2005), who utilized information provided by area dermatologists to detect incidence of As-induced skin problems and attributed its source to drinking water. Results on As exposure via drinking water and characterization of urinary As excretion among adults in the Yaqui Valley, Sonora, México was presented by Meza et al. (2004). Population in the Valley have been chronically exposed to As concentrations of 4.3, 3.1, and 2.5 times above the US EPA drinking water standard of 10 µL L⁻¹. Arsenic exposure was estimated through water intake over 24 h and As excretion was assessed in the first morning void urine. As (III), As(V), as well as monomethyl As (MMA) and dimethyl As (DMA) were determined by high performance liquid chromatography/inductively coupled plasma mass spectroscopy (HPLC/ICP-MS). Meza et al. (2004) found a positive correlation between total As intake by drinking water/day and the total As concentration in urine, with inorganic As as the second major As species present in the urine. Considerable variations in metabolism of As was found within the group which were attributed to polymorphism of the methylation enzymes.

The impact of As toxicity on children 6–8 years of age exposed to areas with reportedly high As concentrations in the area of interest have been reported (Rosado et al., 2007). Based on the available

20-year dataset, a case study was conducted on the communities of a 2.8 million-acre Native American reservation in southwestern Arizona to evaluate the impact of reducing the As MCL from 50 to 10 $\mu\text{g L}^{-1}$ at the local level for several small, rural, tribal, community systems. The study concluded that 17 out of 67 community wells on the reservation have As concentrations above the MCL (10 $\mu\text{g L}^{-1}$) for the Revised Arsenic Rule (RAR) established by the EPA in 2001. The wells are located in 14 communities distributed along the region (Jones and Joy, 2006).

Rosado et al. (2007) conducted studies in elementary schools located near a metallurgical smelter complex in Torreon, México, these studies demonstrated that, independent of socio-demographic variables, nutritional status, and levels of blood lead, association exists between urinary As concentration and several cognition tests such as Visual-Spatial Abilities with Figure Designs, the PPVT, the WISC-RM Digit Span Subscale, and the Visual Search and Letter Sequencing Tests. The researchers showed that the deficit in cognitive performance among school-age children is associated to As, even at low exposure levels, and affects complex cognitive processes such as memory and problem solving which in turn could affect performance at school. Additional studies related to the effect of As on children from the Lagunera region are presented by Pineda-Zavaleta et al. (2004) and Trejo-Acevedo et al. (2009). It is reported that it is unlikely that nutrition has a substantial role in the etiology of As-related diseases (Mondal et al., 2006). Studies conducted elsewhere reported that children exposed to arsenic levels greater than 50 $\mu\text{g L}^{-1}$ in drinking water achieved significantly lower Performance and Full-Scale scores than did children exposed to lower water arsenic levels. Reduction of children's intellectual function was found even after adjustment for socio-demographic covariates in a dose-response manner (Chen et al., 2009).

Using standardized incidence ratios (SIR) for population exposed to As in New Mexico, Siegel (2006) found dependence of the choice of the reference population. He found that the SIR was significantly less than 1.0 when the rates from the US standard population were used as reference but was significantly greater than 1.0 when the adjacent unexposed counties were used as a reference population. To conduct this study Siegel (2006) defined exposed population as white males living in counties in which the majority of the population consumed drinking water from regulated community wells with mean arsenic concentrations of 10 $\mu\text{g L}^{-1}$ or greater.

In the US, 5% of all community water supply systems have to take corrective action to lower As to the current MCL (Mondal et al., 2006). The shallow groundwater of the western US is more vulnerable to As contamination than in the eastern US. Even though Arizona is considered a hotspot for As contamination due to the exposure through untreated groundwater consumption (Kumar et al., 2006), few counties of Arizona were identified with high risk from As exposure through untreated groundwater (Twarakavi and Kaluarachchi, 2006). Mondal et al. (2006) have summarized a global scenario of As poisoning, reporting that in general the range of total As concentrations in wells in México and the US is 8–624 $\mu\text{g L}^{-1}$ and 1–490 $\mu\text{g L}^{-1}$, respectively.

In a recent study conducted elsewhere, chronic arsenic exposure through drinking water was associated with an increase in the mortality rate in that region. The study recommended urgent solutions and resources to mitigate the resulting health effects of arsenic exposure (Argos et al., 2010).

4. Treatment technologies in the area of study

Treatment technologies for As removal may be classified in three main groups: conventional, enhanced and emerging technol-

ogies. Conventional methods include precipitation, coagulation, flocculation, adsorption, ion exchange (Zhang et al., 2007; Elizande-González, 2001), and membrane filtration (Shih, 2005; Ning, 2002). Conventional technologies to clean As-contaminated water have two main limitations related to cost and waste generation, especially for small treatment systems (Elless et al., 2005). The first limitation is due to the requirement of chemicals for pre- and post-pH adjustment for maximal effectiveness. The second is due to the generation of large volumes of backwash water and spent media or sludge that must be disposed of (Elless et al., 2005). Enhanced methods are modifications of already existing removal processes. In the case of adsorption, modification occurs by producing new adsorbents materials (Hristovski et al., 2007; Maji et al., 2007). Emerging technologies are mostly focused on phytoremediation and bio-filtration (Mondal et al., 2006). Phytoremediation is the use of plants to extract, sequester, or detoxify pollutants (Peng et al., 2009). A comparison of best available technologies to phytofiltration for removal of As from drinking water is presented by Elless et al. (2005). Wang and Zhao (2009) summarize recently reported research studies on As bioleaching from soils, and As removal from waters by biosorption. Some of these technologies have been applied in the area of interest and are presented in the next section.

4.1. Conventional arsenic removal methods

Treatments reported for water supply systems in the area of interest consist mainly of reverse osmosis and coagulation (Cadena and Kirk, 1995). Reverse osmosis is a high-pressure membrane process by which dissolved contaminants are removed from a feed stream through diffusion (USEPA, 2000). Other pressure-driven processes (Huxstep and Sorg, 1988) include microfiltration and ultrafiltration. Reverse osmosis is effective in removing As from a solution but generate large volumes of unusable wastewater due to the presence of dissolved minerals (Vogels and Johnson, 1998). In the past few years, within the state of Chihuahua, 88 reverse osmosis treatment facilities have been installed to supply safe drinking water to 70 towns within 23 counties and comply with the existing norm NOM 127 (World Water Forum, 2006). Piñón-Miramontes et al. (2003) used cake alum to treat drinking water in Chihuahua and Ghurye et al. (2004) reported As removal to <2 $\mu\text{g L}^{-1}$ for groundwater in Albuquerque using iron coagulation followed by direct microfiltration. Also in Albuquerque, Thomson et al. (2003) tested a family-size filter consisting of commercially available alumina to reduce As to safe drinking water levels.

Adsorption is another common method used for As removal. It is used as a finishing process in the treatment of drinking water, usually performed after C/F. In adsorption, a substance is transferred from the liquid phase to the surface of a solid where it becomes bound by chemical or physical forces. Some adsorbents include silica, bauxite, kaolinite, and synthetic resins (Prabhu et al., 2003). When activated alumina is used as adsorbent, its oxidized surface sorbs contaminant ions contained in the feed water at an optimal pH of 6.0 (USEPA, 2000). Deng et al. (2006) conducted a bench demonstration in Columbus, New Mexico, using a novel mesoporous activated-alumina adsorbent to remove As and fluoride from groundwater. Groundwater in the Columbus/Palomas border region contains significantly high levels of As (50 $\mu\text{g L}^{-1}$) and is used as the primary drinking water supply (Deng et al., 2006). Some of the factors that made the adsorption technology feasible are its superior performance at removing multiple contaminants, ease of use, and minimal waste generation (Deng et al., 2006).

Westerhoff et al. (2006) presented a case study for evaluating a cost-effective approach for As removal. His approach was applied at a well in Valle Vista and Rimrock, Arizona for a source water

containing 34.8–47.6 $\mu\text{g L}^{-1}$ (USEPA, 2004b, 2006a) and 48.3–81.4 $\mu\text{g L}^{-1}$ (USEPA, 2005a) of total arsenic respectively, with As(V) as the predominant species. The approach was also applied in Desert Sands, New Mexico, with reduction of raw water concentrations of 20.7–30.1 $\mu\text{g L}^{-1}$ and having As(III) as the predominant species (USEPA, 2005b). Treatments included adsorption and filtration with iron-modified activated alumina or granular ferric oxide (USEPA, 2005a,b, 2006a).

4.2. Enhanced methods

Modifications to enhance conventional As treatments methods are numerous elsewhere (Mondal et al., 2006; Mohan and Pittman, 2007; Choong et al., 2007; Dermont et al., 2008). Attempts have been made to develop more efficient As(III) oxidation processes to meet new stringent As drinking water standards. Adsorption has proven to be a more efficient method to remove As, when using alumina and granular ferric hydroxides or ferric salts (USEPA, 2006b) as sorbents. Activated alumina adsorption is best if applied when the initial As concentration is not much greater than the target level; however, the recovered adsorbed As is only 50 to 70% (Vogels and Johnson, 1998). Adsorption using granular ferric hydroxide akaganeite as an adsorbent was also found effective, combining efficiency and a small residual mass of the coagulation-filtration process (Cadena et al., 2002). Iron exchanged natural zeolites have been reported as promising materials for As removal (Elizalde-González et al., 2001; Dávila-Jiménez et al., 2008).

The majority of the treatment methods for the removal of As involve chemical precipitation with lime and ferric salts followed by coagulation and filtration processes at pH values of 7–12. One of the disadvantages of working at these pH levels is the sludge generation caused by hardness and ferric hydroxide precipitation, which makes the process inefficient from a waste minimization point of view. Working at pH 3–4 achieved removal efficiencies of over 99% and produced less sludge (Cadena and Kirk, 1995) but the amount of iron needed for equivalent As removal increased (Vogels and Johnson, 1998). Vogels and Johnson (1998) proposed converting the ferrous iron into ferric form by oxidation with a strong agent to decrease the iron requirement, which further minimized sludge production. Song et al. (2006) reported an enhanced coagulation process with ferric ions and coarse calcite. They reported that the enhanced coagulation followed by conventional filtration achieved over 99% removal from water with high-As concentrations (5 mg L^{-1}). It is suggested that attractive electrical double layer interactions between small As-borne coagulates and calcite particles eliminate the potential energy barrier between the heterogeneous particles. Parga et al. (2005) removed As with an efficiency of more than 99% from contaminated groundwater in a field pilot scale study in the Comarca Lagunera by using a modified electrochemical process with air injection. This allowed reduction of the reactive retention time, minimizing the residual mass and, therefore, reducing the operating costs of the process.

Natural adsorbent materials, such as zeolites (Elizalde-González et al., 2001) and akaganeite (Cadena et al., 2002) have been used for As contamination treatment. Elizalde-González et al. (2001) used zeolite of the clinoptilolite type produced in México to investigate its sorption efficiency for various redox species of arsenic. The zeolite was capable of reducing the concentration of both As(V) and As(III) to WHO's recommended maximum levels. Modified natural adsorbents have also been proposed for As removal. Sullivan et al. (2003) used hexadecyl-trimethylammonium bromide (HDTMA-Br) surfactant to modify a clinoptilolite zeolite from Winston, New Mexico to remove As from soil leachate. They found that the surfactant modified zeolite (SMZ) can effectively adsorb As from high pH leachate solutions while simultaneously removing the large concentration of dissolved organic carbon typically gen-

erated by alkaline soil washing processes. It is assumed that during the process exchange sites in the SMZ originally occupied by Br^- and OH^- are replaced by the anionic forms of As. It is reported that SMZ adsorbs all major classes of water contaminants (anions, cations, organics, and pathogens) thus making it amenable to a variety of water treatment applications (Bowman, 2003). Cadena et al. (2002) conducted experimental studies using granular ferric hydroxide akaganeite for the treatment of groundwater contaminated with arsenic. Akaganeite is a microscopic iron oxyhydroxide and is considered as a very effective adsorbent for the removal of arsenate from drinking water. This technology takes advantages of the efficiency and small residual mass of the coagulation-filtration process combined with the simple processing of the fixed bed adsorption on activated alumina. Novel adsorbent materials, such as titanium oxide, iron oxide nano-particles, granular ferric oxide, zirconium oxide, modified bone char, iron coated resin, and coated silicate have also been implemented (Aragon et al., 2007a,b,c; Siegel et al., 2007; Siegel et al., 2008). In all the cases, the adsorption performances of the adsorbent materials depended on the physical and chemical nature of the media and the test methodology.

4.3. Emerging technologies

Some emerging technologies have been applied in the area of study, the majority of them oriented to the emerging phytoremediation. In most of the cases only laboratory scale experiments are presented and no cost-efficiency analysis was presented. The use of selected metal-accumulating plants has been presented as an efficient alternative for the removal of As (Gardea-Torresdey et al., 2001, 2004; Flores-Tavizón et al., 2003; Elless et al., 2005; Aldrich et al., 2007). Elless et al. (2005) conducted a pilot scale demonstration in Albuquerque, New Mexico, using an As-hyperaccumulating fern (*Pteris vittata*) to remove As from drinking water and obtained an As concentration below the detection limit (2 $\mu\text{g L}^{-1}$) at a water flow rate as high as 1900 L d^{-1} . They reported an average bioaccumulation factor (BAF) exceeding 16,000 and proposed this system as the basis for a solar-powered hydroponic technique for small scale treatment of As-contaminated water. Plants classified as As hyperaccumulators require a concentration in their biomass of at least 1000 $\mu\text{g g}^{-1}$ in dry weight, and a bioconcentration capacity in their tissues of more than 100 times (Flores-Tavizón et al., 2003). Also, the metal concentration in shoots should be greater than that in roots and the enrichment coefficient should be greater than 1 (Haque et al., 2008). The significantly higher efficiency of As phytofiltration by As-hyperaccumulating fern species is associated with their ability to rapidly translocate absorbed As from roots to shoots (Malik et al., 2009). Peng et al. (2009) suggest that the direct uptake capacity of phytoremediation for heavy metals can be achieved by most plant species able to grow in the contaminated soil. Indirect reactions, such as stimulation of microbial activity, redox reactions, and formation and precipitation of insoluble metal compounds in the rhizosphere, may play a relatively important role in the phytoremediation process. The hyperaccumulator *Pteris Vittata* has been used to remove As from soils elsewhere (Shelmerdine et al., 2009). The use of engineering genetic strategies to increase the As-hyperaccumulating capacity of plants has been proposed by Wang and Zhao (2009).

Flores-Tavizón et al. (2003) identified As-tolerating plants, such as *only Eleocharis sp.*, *Brickellia veronicaefolia*, *Nicotiana glauca*, and *Baccharis salicifolia*, from mine sites and hot springs in the semi-arid regions of Chihuahua, while Salt et al. (1995) discussed the biological mechanism of toxic metal uptake by native plants in arid contaminated soils. Flores-Tavizón et al. (2003) identified *only Eleocharis sp.* as an indigenous plant from the State of Chihuahua, capable of tolerating As with a maximum tolerating capacity of 342 $\mu\text{g g}^{-1}$. The plant showed a bioconcentration factor (BCF) and

a translocation factor (TF) of 5.22 and 7.37, respectively. The BCF is defined as the ratio of the metal concentration in the shoots and in the soil, while the TF is the ratio of the element concentration in the shoots and the roots.

Gardea-Torresdey et al. (2001) studied the distribution of As concentrations in creosote bush and soil samples collected near a smelter in El Paso, Texas. He found that creosote bush is capable of actively absorbing As from the environment through different parts of its system. The As values encountered for the roots, stem, and leaves were 0.9–10 mg kg⁻¹, 0.26–4.61 mg kg⁻¹, and 4.3–12 mg kg⁻¹, respectively. Gardea-Torresdey et al. (2004) applied phytoremediation and the use of biosorbents for the removal of toxic heavy metals in water treatment.

Studies have been conducted elsewhere related to the As (III) and As(V) sorption onto different adsorption materials. Among these materials, amorphous iron oxides, magnetite, and goethite have been used. In the majority of the cases these adsorbent show high adsorption capacity for both As(III) and/or As(V) (Gimenez et al., 2010). In the area of study, phytoremediation has been applied to remove As(III) as well as As(V) species from drinking water. Haque et al. (2009) identified Sorghum (*Sorghum bicolor*), a plant grown in West Central Texas under hydroponic media, as a potential hyperaccumulator of As contaminated soil and water. They found that sorghum plants can accumulate approximately 30 times more As when the supplied solution is As(V) rather than As(III). The uptakes by roots, stems, and leaves were 2400, 810, and 300 mg kg⁻¹, respectively. The study demonstrated that the uptake of As by sorghum plants depends completely on As speciation.

Aldrich et al. (2007) studied the mechanism by which As is taken up by the root, stem, and leaf portions of a mesquite plant, a plant native to the area of interest. He found that the uptake of As(V) by the plant was preferential as compared to As(III), but once As(V) reached the roots of the plant it was reduced to As(III) which was then transported and stored in the leaves. Aldrich et al. (2007) reported the potential application of mesquite (*Prosopis* spp.) to remove As from contaminated soils in arid regions. Mesquite plant is a common small tree of the Desert Southwest of the United States and Northern Mexico. The plant is characterized by having highly drought and salt resistance and a high transpiration rate. They used X-ray adsorption spectroscopy (XAS) to prove that As(V) was reduced to As(III) in the plant by building sulfur ligands. The authors reported that mesquite plants exposed to As(V) concentrated about 770, 326, and 119 mg kg⁻¹ As in roots, stems, and leaves, respectively. Lopez et al. (2008) found that in the roots of plants treated with 50 mg L⁻¹ As(V) both species (As(III) and As(V) are present; however, As(III) was found to be the predominant species in the stem, demonstrating that mesquite fully reduce As(V) to As(III). Further studies conducted by Mokgalaka-Matlala et al. (2009) using mesquite plants (*Prosopis* spp.) showed that As speciation plays a role in the formation of low molecular weight thiols (LMWT) in response to As toxicity. They showed that the roots of mesquite plant exposed to As (III) produced significantly higher LMWT levels than the levels produced by the roots exposed to As(V) at the same concentration of As(III) applied. At high As(III) concentrations no LMWT were produced by the plant, forcing all defense mechanism to shut down and causing eventual plant death. Thiol groups play a key role in arsenic detoxification in plants (Mokgalaka-Matlala et al., 2009).

More recently, As accumulation on the Desert willow (*Chilopsis linearis* sweet), a deciduous desert tree native of southwestern US and northern Mexico, was studied (Castillo-Michel et al., 2009). Castillo-Michel et al. reported that the Desert willow grows in alkaline soils with a pH range of 6–9 and tolerates full sun exposure and drought conditions that have also been found in copper mine tailing at Globe, Arizona demonstrating its adaptability to

sites contaminated with As. They found that *Chilopsis linearis* was tolerant to concentrations of up to 40 mg L⁻¹ when As was present as As(V). AsO₄³⁻ species were found in roots, stems, and leaves of the plant. The plant was also capable of partially reducing As(V) to As(III) in the tissues. In this study, inductively coupled plasma optical emission spectroscopy (ICP-OES), and X-ray absorption spectroscopy (XAS) was used to study the speciation and local coordination environment of As inside the plant.

An emerging remediation technology has been developed by Arienzo et al. (2001) for the removal of As(III) from artificially contaminated aqueous solutions, attaining a removal efficiency of 98%. The technique consists of the electrochemical peroxidation of As with direct electric current applied to a steel electrode and a small addition of H₂O₂. Electrocoagulation has also been used to reduce As concentrations to below 10 µg L⁻¹. In this process, As(III) is oxidized to As(V), which is subsequently removed by adsorption/complexation with metal hydroxides (Kumar et al., 2004).

Innovative technologies include the use of new flocculent and adsorbent materials. Piñón-Miramontes et al. (2003) combined cake alum and a polymeric anionic flocculent (PAF) and removed 99% of As from two wells at Meoqui, Chihuahua, México. They found that the increase in As removal is directly related to the reduction of soluble Al but independent of pH. Fierro et al. (2009) prepared an iron-doped activated carbon adsorbent material by ferric chloride forced hydrolysis and removed 94% of the As present in a groundwater well from the State of Chihuahua, México. Only 14% As was removed using a commercial activated carbon (AC) adsorbent. The higher adsorption capacity of the iron-doped AC adsorbent was attributed to the existence of iron (hydr)oxides nano-particle agglomeration, which once hydrated prevented the arsenate (HASO₄⁻) diffusion toward the inner surface of the AC grain. In a previous study conducted by the same research group (Muñiz et al., 2009), they found that for a given doping method, increasing the iron content in the iron-doped activated carbon increased the corresponding As uptake. They also found that metal removal performance depends on the way the doping is carried out, with small and highly dispersed iron-oxide-based particles being the ones that lead to the highest As removal efficiency (Muñiz et al., 2009). Recently, fly ash from a Mexican mineral coal-fired plant (MFA) was used in one-step process as a raw material in producing zeolite for further application in As removal from aqueous solutions (Medina et al., 2010). The authors explored two routes in the synthesis of the zeolite. The first, direct MFA zeolitization, which resulted in the formation of W zeolite with KOH and analcime with NaOH; the second, a MFA fusion route which resulted in the formation of zeolite W or chabazite with KOH and zeolite X or P with NaOH. The chemical nature of the W zeolite surface was modified with aluminum sulfate to force the adsorption of anionic As(V) species instead of the cationic species that zeolites naturally adsorb. The new material was capable of removing 99% As from an aqueous solution originally containing 740 µg L⁻¹.

Nanoscale iron particles are an innovative extension of conventional zero-valent iron technologies. However, studies on the use of nanoscale iron particles for As removal are still limited (Yuan and Lien, 2006). In the area of study, few studies have been reported. Fierro et al. (2009) removed 94% of the 300 µg L⁻¹ As present in a groundwater well from the State of Chihuahua (México) using iron-doped commercial activated carbon (AC) while the commercial AC removed only 14%. Activated carbon is one of the most widely used adsorbents for As removal from drinking water; however, its adsorption capacity is not always satisfactory. Its efficiency depends on the physicochemical properties of the solution and on As speciation. Iron-based nanoparticles, homogeneous in size and well-dispersed in the carbon matrix were responsible for the increase of As removal. The AC nanoparticles were obtained

after six hours of ferric chloride forced hydrolysis. Nitrogen adsorption–desorption isotherms at 77 K, transmission electron microscopy (TEM), and atomic adsorption spectrometry (AAS) were used to characterize the modified material and to quantify As removal (Fierro et al., 2009). Muñoz et al., 2009 found that Fe(II) increased the iron content in the iron-doped AC, as compared to Fe(III), leading to high As uptake.

Biological processes to remove, mobilize, and contain As include sorption, biomethylation-demethylation, complexation, coprecipitation, and oxidation–reduction (Katsogiannis and Zouboulis, 2004). From these technologies, bioremediation has been applied in the area of interest and is reported as a potential cost-effective and efficient technology. Bioremediation based on microbial degradation has been applied as a remediation strategy at superfund sites. The implication of using bioremediation for the treatment of groundwater contaminated with arsenic was presented elsewhere (Islam et al., 2004; DeLemos et al., 2006). The natural process of bacterial degradation can occur whenever groundwater is contaminated by most commonly occurring contaminants. When microbial degradation of organics occurs under reduced conditions, groundwater may become contaminated by naturally occurring As. DeLemos et al. (2006) found a strong correlation between elevated As levels and organic contamination in groundwater systems and suggested that As contamination caused by natural degradation of organic contaminants may be a widespread phenomenon. Reducing as well as oxidizing environments can be found within a groundwater well (Amini et al., 2008). Reducing environment conditions are present when the supply of oxygenated water to the subsurface is diminished (DeLemos et al., 2006) and under these conditions the As mobility commonly increases.

5. Conclusions

Occurrence and treatment of As in the arid region of northern Mexico (states of Chihuahua and Coahuila) and the bordering states of the southwestern US (New Mexico, Arizona, and western Texas) were reviewed in this article. Main sources of As in the study area were found to be primarily of natural origin and associated with erosion of volcanic rocks followed by deposition in deeper parts of former drainage basins. However, sources of non-naturally occurring As included pesticides, sewages and smelting plants.

Potential future As release from sediments of the Rio Concho Basin in northern México into the water column is a cause of concern. Gaps in information abound in pointing depth and the aerial extent of local As-rich formations of concern. Exposure to As contamination has shown to be detrimental to the central nervous system and for cognitive development in children, independent of socio-demographic variables, nutritional status, and levels of blood lead.

A variety of methods to treat contaminated water and soil are available. Among them, reverse osmosis, coagulation, adsorption, and electrochemical methods have been applied in the area of interest to remove As. New innovative adsorbent material has also been applied. Some emergent technologies stand out, especially phytoremediation. Sorghum (*Sorghum bicolor*), desert bloom (*Baccharis sarothroides* Grey), fern (*Pteris vittata*), and only *Eleocharis* sp. plants growing in the states of Texas, Arizona, New Mexico and Chihuahua, respectively, may be considered as potential hyperaccumulators for the removal of As from contaminated soil and water in the region. In the area of study, enhanced and emerging technologies have been applied in most of the cases only at laboratory or bench scale. Pilot-scale applications are still needed to estimate the cost-efficiency and determine the most appropriate parameter conditions for process implementation.

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