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Review

Small-scale and household methods to remove arsenic from water for drinking purposes in Latin America

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ABSTRACT

Small-scale and household low-cost technologies to provide water free of arsenic for drinking purposes, suitable for isolated rural and periurban areas not connected to water networks in Latin America are described. Some of them are merely adaptation of conventional technologies already used at large and medium scale, but others are environmentally friendly emerging procedures that use local materials and resources of the affected zone. The technologies require simple and low-cost equipment that can be easily handled and maintained by the local population. The methods are based on the following processes: combination of coagulation/flocculation with adsorption, adsorption with geological and other low-cost natural materials, electrochemical technologies, biological methods including phytoremediation, use of zerovalent iron and photochemical processes. Examples of relevant research studies and developments in the region are given. In some cases, processes have been tested only at the laboratory level and there is not enough information about the costs. However, it is considered that the presented technologies constitute potential alternatives for arsenic removal in isolated rural and periurban localities of Latin America. Generation, handling and adequate disposal of residues should be taken into account in all cases.

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Contents

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1. Introduction

Arsenic removal from waters to reach levels in accordance with the regulations for drinking water purposes ($<10\,\mu g\,L^{-1}$, World Health Organization, 2004) is not an easy task. Particularly, for populations with low economical resources like those of many Latin American (LA) periurban and rural isolated communities, economical aspects are perhaps the most important factors for the selection of the technology, considering the size of the population, incidence of chronic illnesses, lack of safe water, poverty conditions, and other socioeconomic variables, including cultural and political issues, aspects that few times are taken into account.

The presence of arsenic in ground- and surface water used for drinking purposes can cause serious health problems, especially the incidence of HACRE (Hidroarsenicismo Crónico Regional Endémico in Spanish, Chronic Endemic Regional Hydroarsenicism), see McClintock et al., submitted for publication. Due to these health problems, it can be estimated that 14 million people out of the around 500 million living in LA are considered to be at risk in the LA regions. Taking into account the relevance of the problem, a large amount of scientific and technological work has been devoted in recent times to develop new economic, emergent technologies for arsenic remediation (Litter, 2002, 2006a, 2006b; Litter and Jiménez González, 2004; Litter and Mansilla, 2003; Bundschuh et al., 2010). As indicated there, efforts put to solve the problem at medium and large scale in several LA countries, i.e. those serving to more than 1000 people by centralized water networks, were successful, but only in very few cases the problem has been actually solved in isolated settlements (defined as those inhabited by less than 50 persons) or isolated houses (defined as those separated by some hundreds or thousands of meters), where centralized drinking water supply systems are absent. Although several novel simple technologies have been developed, in spite of the great health problems above mentioned, low attention by the local authorities or international agencies caused that alternative successful techniques still remain at the laboratory scale or have been tested only in few experiments in field (Cornejo et al., 2006b; de la Fuente et al., 2006). Practically no action has been implemented to ensure As-safe drinking water supplies in rural or periurban areas to accomplish the national regulations for As in drinking water, which in most of the LA countries match that of the World Health Organization (2004). A great effort should then be put to develop short-scale point-of-entry or point-of-use systems at household level appropriate for the conditions prevailing in these isolated communities (not connected to a central water supply system). Not only drinking water or water for food preparation needs to be remediated or replaced–which is

only a few percent of the total domestic water consumption–but also that used for irrigation purposes should be treated, since As can contaminate the crops.

All technologies for As removal rely on a few basic chemical processes that can be applied alone, simultaneously or in sequence: oxidation/reduction, coagulation–filtration, precipitation, adsorption, ion exchange, solid/liquid separation, physical exclusion, membrane technologies, biological methods, etc. (Litter et al., 2010a, 2010c). As it is well known, most As removal technologies are efficient when the element is present in the pentavalent state, because it forms oxianions, mainly $H_2AsO_4^-$ and $HAsO_4^{2-}$, in a pH range of 2–12, while the trivalent form is uncharged at pH below 9.2 (H_3AsO_3). This is the reason why many As remediation methods use, previously to other processes, an oxidation step to oxidize As(III), if present, to As(V). However, oxidation without help of other physical or chemical transformations does not remove As from water and have to be followed by other processes. As it is obvious, boiling of water for purification does not remove As and, on the contrary, this process increases As concentration by evaporation. This is a fact commonly ignored by most of the potentially affected people.

In what follows, some small-scale technologies studied and applied in poor, isolated, decentralized rural and periurban populations of LA will be described. Some of these technologies are merely adaptation of conventional methods like coagulation–filtration, or adsorption, using very cheap materials, while other are based on novel technologies such as biological or photochemical processes. No references to studies or papers of other regions of the world will be made, but they can be consulted in several recent references on the subject (e.g., Höll and Litter, 2010; Litter et al., 2010a; Mohan and Pittman, 2007; Morgada and Litter, 2010; Ravenscroft et al., 2009).

It is important to emphasize that for the use of techniques, the population has to be trained for proper handling and disposal of the wastes, to prevent additional environmental or health risk.

2. Combined oxidation, adsorption, coagulation/flocculation methods

In LA, different technologies for single households were developed or adapted by scaling down and simplifying conventional methods used in water treatment plants for As removal, such as those described in Litter et al., 2008, 2010c. These methods use the oxidation, adsorption and coagulation sequence (Sastre et al., 1997; Esparza and Wong, 1998). Some examples follow.

A household scale low-cost As removal methodology was developed in Peru (Esparza, 2002; Esparza and Wong, 1998; Castro de Esparza et al., 2005; Castro de Esparza, 2010) using ALUFLOC, a
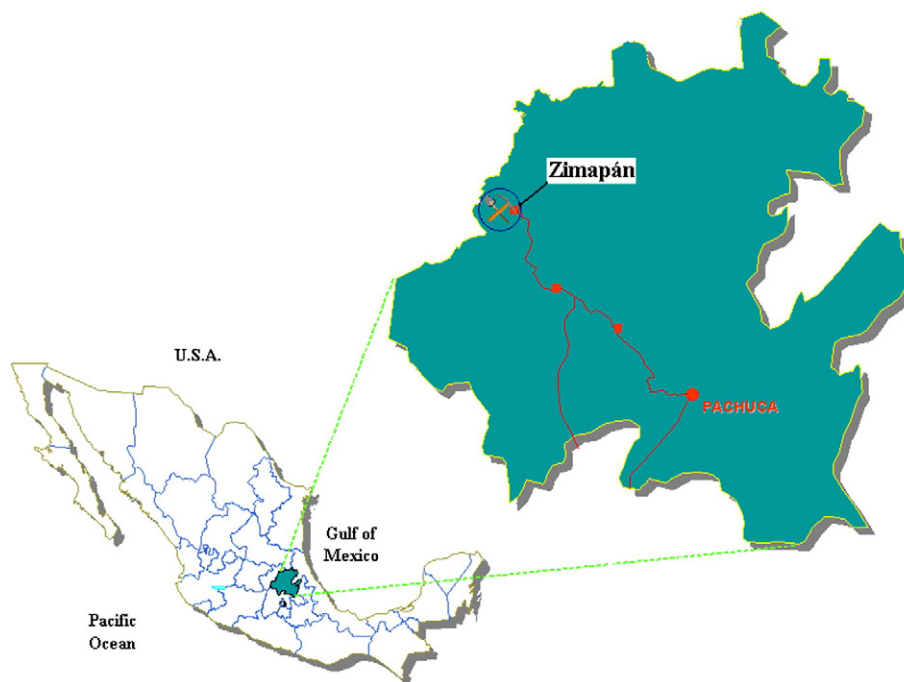
Fig. 1. Location of Zimapán, México.

mixture of an oxidant (chlorine), activated clays (acting as As adsorbents and/or ion exchangers) and a coagulant ($Al_2(SO_4)_3$ or $FeCl_3$) (Bedolla et al., 1999). This methodology was tested in Puno (Peru) and allowed to remove up to 98% of the dissolved As (As concentration in raw water: $1 \, mg \, L^{-1}$), reaching the value regulated by the Peruvian normative at that time. Training campaigns with the population have been performed.

The suitability of an activated aluminum hydroxide hydrogel, added directly to the water, for use at household-scale was tested (Lujan and Graieb, 1994, 1995; Luján, 2001). The hydrogel was prepared using hydrated aluminum sulfate, powdered calcium hypochlorite, ammonium hydroxide and demineralized water. Two hundred groundwater samples ($40–800 \, \mu g \, L^{-1}$ As) from different wells of the Tucumán province (Argentina) were tested; in all cases, final As concentrations below $10 \, \mu g \, L^{-1}$ were attained, in accordance with the national regulation (Código Alimentario Argentino, 2007).

3. Geological and other low-cost materials as adsorbents

3.1. General

Geologic materials including diverse types of rocks and minerals such as iron oxides and hydroxides, clays, limestones, and feldespates have the capability of retaining As at the pH values of natural waters. Remediation with natural geological materials (soils or sediments) is an emerging solution for poor people in remote rural settlements at household level, especially if the materials are locally available and can be collected by the population.

Various natural Fe- and Al-rich minerals such as iron oxyhydroxides, hematite (α-Fe_2O_3), goethite (α-$FeO(OH)$), gibbsite (γ-$Al(OH)_3$)), iron/manganese oxides like "greensand" and soils or sediments including these minerals (e.g., oxisols, laterite), indigenous limestone, zeolites, iron-coated zeolites, clay minerals (montmorillonite, bentonite) were tested for As removal in LA either with laboratory-prepared or natural waters with rather good success (Alvarez-Silva et al., 2009; Armienta et al., 2009; Bundschuh et al., 2011; Claesson and Fagerberg, 2003; Dávila-Jiménez et al., 2008; Deschamps et al., 2003, 2005; Esparza and Wong, 1998; Garrido et al., 2008; Litter,

2006a, 2006b; Litter and Mansilla, 2003; Litter and Jiménez González, 2004; Ladeira and Ciminelli, 2004; Lindbäck and Sjölin, 2006; Mejía Zamudio et al., 2009; Mellano and Ramirez, 2004; Muñoz et al., 2005; Ongley et al., 2001; Petkova-Simeonova, 1999; Rivera and Piña, 2000; Simeonova, 2000; Storniolo et al., 2005).

Some cheap adsorbents such as granular iron hydroxide (GFH®), Bayoxide® (GFO), and commercial titanium dioxide, all of them proved to be effective materials for As removal (Höll and Litter, 2010; Litter et al., 2010a), have been also tested in some waters of the Chacopampean plain in Argentina. Although the use of these materials is very simple, does not require chemical addition and can be useful at community or household levels, to our knowledge, results have not been very good. However, they are still submitted to research in some laboratories of the region.

In what follows, the use of local adsorbents in Zimapán, a mining zone of Mexico, a "family filter" developed for the Lagunera region in Mexico and studies performed using derivatives of kaolin in Salta, Argentina, will be presented in more detail.

3.2. Arsenic removal by indigenous limestone in a mining zone of Mexico

Zimapán (Hidalgo state, Mexico, 20°40′N and 99°22′N, Fig. 1) is a historical mining site where health-effects due to consumption of As-polluted water have been observed in the population (Armienta et al., 1997a; Valenzuela et al., 2007). The geology of Zimapán is composed by Mesozoic-age limestones and Cenozoic-age volcanic and continental deposits (Armienta et al., 1997b). In spite of the presence of several tailing deposits without environmental protection regulations, dissolution of As-bearing minerals naturally present in the fractured limestone aquifer is the main groundwater pollution source (Armienta et al., 2001; Sracek et al., 2010). A well used as drinking water source until 1995 contained $1100 \, \mu g \, L^{-1}$ As. Currently, although part of the population is supplied with better quality water, due to the climate conditions and lack of superficial water, a highly polluted well ($430 \, \mu g \, L^{-1}$ in January, 2010) is still connected to the municipal water supply system.

Based on preliminary tests and its ubiquity in the area, diverse experiments were performed with rock samples from the Soyatal

(lutitic limestone) formation, to evaluate its suitability to remove As. Samples from Soyatal used in the experiments carried out by Ongley et al. (2001) showed chemical composition variations, from 20 to 80% calcite, 20 to 65% quartz and from less than 1 to 15% clay.

A pilot study of As removal was carried out in this place. Preliminary research identified hematite as an alternative sorbent for removal, and the results in field confirmed the effectiveness of the materials, which allow reaching concentrations from 3 to 28 µg L^{-1} in the treated water, lower than the Mexican Regulation of 50 µg L^{-1} at that time (Official Mexican Standard, 1994; Petkova-Simeonova, 1999).

Laboratory experiments performed to increase the knowledge about the capacity of indigenous limestones to remove As were also developed (Romero et al., 2004). Samples containing a high proportion of calcite (80%) showed a pzc between 9.3 and 9.5, in agreement with that found by other authors (Sadiq, 1995), indicating the potential to retain As at the slightly basic pH of the contaminated water. This capacity was confirmed by batch sorption experiments developed with artificial As solutions at various pH values, and with natural groundwater. A high As removal was determined at pH 7.4–11.2. At this pH range, calcite is responsible for As sorption, as previously proved (Romero et al., 2004; Román-Ross et al., 2006; So et al., 2008). About 75% As was removed ($[As]_0 = 4220$ µg L^{-1}) using a 20:10 water:solid ratio at pH 7.4. Coprecipitation of complex Ca arsenates or As adsorption onto calcite or clay may have been responsible for As retention.

The extent of As removal from a solution prepared equilibrating water with local tailings was tested with distinct rock:water ratios, particle size, reaction time, and shaking periods in batch experiments. The highest removal was achieved with rocks from the Soyatal limestone formation. Continuous shaking during 12 h with Soyatal rock particles decreased As from 768 µg L^{-1} to 55 µg L^{-1}. Eight, 12 and 24 h reaction times removed 63%, 64% and 78% of As, respectively, from a water sample containing 322 µg L^{-1} As. Increase in the rock:water ratio augmented also the As removal. As expected, due to a larger surface area, a higher removal was obtained with smaller rock particles. However, crushed and powdered rocks had similar effectiveness to remove As at a 1:5 rock:water ratio. Other experiments demonstrated a decrease of As concentration in native water from 500 µg L^{-1} to less than 30 µg L^{-1} using a 1:10 rock:water ratio (Ongley et al., 2001).

The capacity of rock samples to treat various batches of fresh polluted water was also determined in subsequent tests. The same rocks may be used at least 5 times without decreasing their As removal capability. When 10 g rock particles (<0.5 mm size) were tested in one liter of a water sample from a well used for drinking water purposes, a 90% decrease in the As content (460 µg L^{-1}) was reached. Flow-through columns packed with crushed Soyatal rocks showed also good As removal. Experiments were performed with rock particles of different size to assure adequate flow and good As retention. Garden polyethylene hose (1.27 cm diameter) packed with rocks with diameters ranging 0.84 to 1 mm decreased As content from 200 µg L^{-1} to 25 µL^{-1} in the first 2 min. However, the concentration in the outflow increased up to 100 µg L^{-1} As after one hour (Armienta et al., 2009; Flores et al., 2009).

Soyatal rock samples may be used as an in-house method to treat As-polluted water in a very simple way that does not require any particular skill. A bucket is filled with one to 2 kg of crushed rock of Soyatal and 20 L of water containing up to 50 µg L^{-1} As. The mixture should be stirred as frequently as possible during 24 h, allowed to settle, and filtered through several layers of cloth (Ongley et al., 2001). Results from column experiments showed also the potential to use the materials to treat the water flowing from a contaminated well still connected to the potable water pipeline. Waste limestone may be disposed in the tailings dams located outskirts of Zimapán town, whereby As retention on iron oxyhydroxides and formation of more stable As-bearing minerals such as beudantite have been observed to control As mobility (Méndez and Armienta, 2003; Romero et al., 2006).

The proposed system has been informed to the population and authorities of Zimapán (including handling of wastes) but it has been not yet put in practice by the inhabitants.

3.3. Uptake of arsenic by 4-Na-mica, a derivative of kaolin, in Salta, Argentina

Clay minerals are very abundant in different regions of LA; particularly, in Salta (northwest Argentina), the predominant clays are illite and kaolins. Lorenzo et al. (2008) studied As removal from water with a low-cost synthetic 4-Na-mica and its product of mechanical treatment (4-Na-micaMT), obtained from kaolin. The 4-Na-mica (Na$_4$Si$_4$Al$_4$Mg$_6$O$_{20}$F$_4$.xH$_2$O) was synthesized from kaolin following the method of Kodama and Komarnemi (1999). A portion of the obtained mica was ground in an oscillating mill during 300 s to obtain the 4-Na-micaMT. The products of synthesis presented a considerably higher capacity of cationic exchange and a higher pzc than the original kaolin, which improves the ability for adsorption of anions. In particular, the product of mechanical treatment developed an altered structure with a considerable number of aluminum ions exposed in the edges of the particles that could act as potential sites to improve As removal.

Arsenic adsorption studies were made in batch at room temperature using an orbital agitator at pH 7.0. Suspensions of 50 mg mineral in 50 mL of osmosis water containing As at 550 µg L^{-1} were prepared and stirred during periods from 30 min to 4 days. Then, the adsorbent was filtered and the remaining As concentration was analyzed by atomic absorption spectrophotometry connected to a hydride generator. Adsorption isotherms were obtained using a solution containing 1650 µg As L^{-1} and varying the mass of adsorbent between 10 and 450 mg.

The mechanically treated mica adsorbs a larger amount of As, needing contact times of six days to reach the maximum removal capacity, whereas the non-ground material needs only 2 h. The higher efficiency of 4-Na-micaMT can be attributed to the fact that the treated material presents a higher quantity of negatively charged border sites that adsorb protons of the solution producing a continue increase of pH. As the contact time increases, the ground mica decreases its charge and, consequently, the electrostatic repulsion, being able to adsorb a higher quantity of As species. The removal capacities obtained were 60.7% for 4-Na-mica and 96.5% for the mechanically activated mica, showing the potentiality for its application in the treatment of waters contaminated with As.

Both materials were also tested with natural contaminated waters (ca. 200 µg As L^{-1}) from the Chacopampean plain (Salta, Argentina), at laboratory scale. Although the removal capacity decreased, the sorbents were still very efficient for As retention. More detailed studies are needed before the materials can be employed in filters in rural areas.

4. Electrochemical technologies: application of capacitive deionization for arsenic removal from water for human consumption in the Lagunera region, Mexico

Some simple electrochemical technologies can be applied for As oxidation and removal from water. In this section, the use of capacitive deionization to remove As from water of the Lagunera region in Mexico will be described (Garrido et al., 2008, 2009; Garrido Hoyos et al., 2010).

In some parts of the Lagunera region, central part of northern Mexico, chronic As poisoning is endemic (Cebrián et al., 1994). Most of this region is reputed to have a substratum rich in As, producing high As levels in well water, in the 0.080–624 µg L^{-1} range (Del Razo
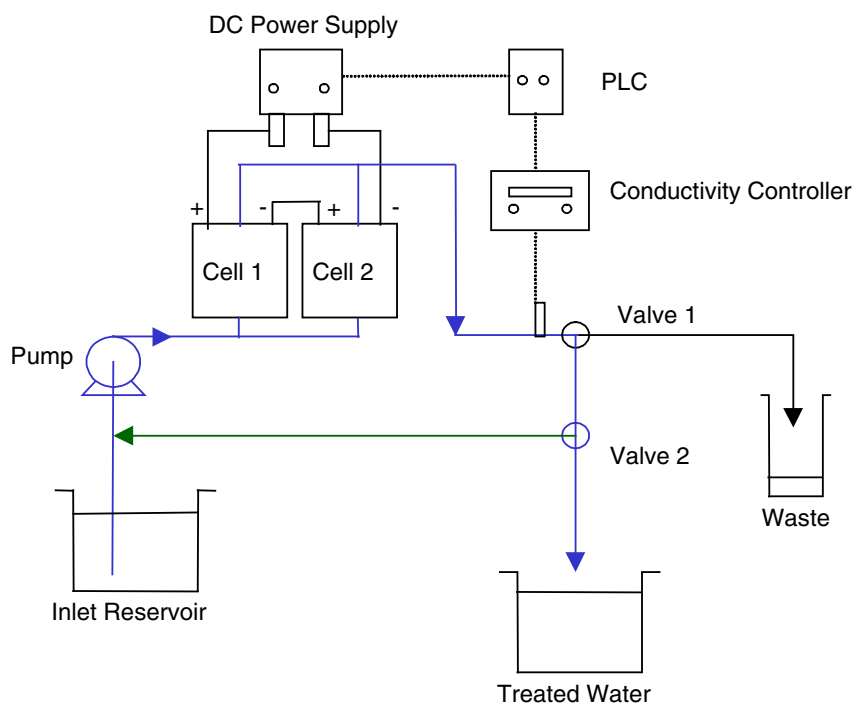
Fig. 2. Typical scheme of a capacitive deionization system. Nominal flow: 3.3 L min^{-1}, operation pressure: 1.0 kg cm^{-2}, and initial TDS: 1000 mg L^{-1}.

et al., 1990). It has been estimated that more than half a million people consume water with As concentrations superior to 50 μg L^{-1}.

The technology of capacitive deionization is an alternative to reverse osmosis with low operational cost, minimal maintenance and easy operation. A Canadian advanced electrochemical technology based on this process was applied to evaluate As removal from a well from the Lagunera region (located in Gómez Palacio, 25°26′45″N, 103°26′18″W, Coahuila State, Mexico). The system works with an electrostatic charging cell constituted by carbon electrodes, behaving as a capacitor. The process works through three stages. During the first stage, the capacitor is energized by direct current, creating positive and negative charged surfaces. Arsenic ionic species are attracted and electrostatically adsorbed onto the surface of the electrodes. To regenerate the electrode surfaces, the polarity of the cell is automatically reversed during stage two, causing the capacitor to release the contaminants into the cell channels. The third stage is the removal of the contaminants from the cell by flushing with a small quantity of liquid, forming a concentrated solution.

Fig. 2 shows the flow diagram of the capacitive deionization plant (Model DesEL-4 k, EnPar, Canada), where the pilot tests were performed. The two capacitive deionization cells of this plant were connected electrically in series and hydraulically in parallel. Each cell was supplied with a peak voltage of 1.2 VDC and a maximum peak current of 375 A.

Working at six different configurations, in which desorption time was kept constant, and starting from an As initial concentration of 100 μg L^{-1}, As removals between 93–97% were obtained. Other ionic compounds like NH_4^+, NO_3^-, PO_4^{3-}, SO_4^{2-}, Ca^+, K^+, Mg^+ and Na^+ were removed simultaneously. The value of the specific energy consumption was 1.321 kWh m^{-3}. The rejected water produced had high values of ionic compounds, especially for As, 3420 μg L^{-1}. Removal of As from reject water was investigated in a further research, using goethite as adsorbent (Garrido et al., 2008).

The results of the studies indicate that capacitive deionization is an efficient technology, potentially economical: a consumption of only 1.3–1.7 kWh m^{-3} was estimated. The cost of the treatment was calculated as around 0.15 USD kWh m^{-3}. It should be remarked that

high temperatures may affect the proper operation. Trained operators are needed and upscaling for flows higher than 10 L/s have been not attempted.

5. Biological methods

5.1. General

Relatively little is known about the use of biological removal of As from water, although these methods show a great potential due to its environmental compatibility and possible cost-effectiveness. Microbial activity can remove, mobilize, and immobilize As through sorption, biomethylation–demethylation, complexation, coprecipitation, and oxidation–reduction processes. *Ex-situ* bioleaching can effectively remove As from contaminated soils, helped by biostimulation, e.g., addition of carbon sources and mineral nutrients. Bioadsorption, i.e., adsorption of pollutants by a biomass or biofilm of living or dead organisms such as algae, bacteria, aquatic macrophytes or vegetal organisms and biopolymers, can be used either *ex-situ* or *in-situ*; coprecipitation with biogenic solids or sulfides can be also tried. Both As(III) and As(V) can be efficiently adsorbed and precipitated onto biological flocs built by iron bacteria (Litter et al., 2010a; Teixeira and Ciminelli, 2005).

In LA, different types of low-cost natural biological materials such as cellulose, milled bones, sedges, sorghum biomass or waste biomass have been tested for their suitability to remove As from water: cellulose and activated carbon filters (Muñiz et al., 2009), activated waste biomass with high fibrous protein content obtained from chicken feathers (Teixeira and Ciminelli, 2005), non-immobilized sorghum biomass as a sorbent (Porras, 2006), biosoils (Ascar et al., 2008) and natural biogenic hydroxyapatite from cow-charred bones (Czerniczyniec et al., 2007; Litter et al., 2008).

5.2. Phytofiltration

A promising emerging alternative to conventional cleanup methods is phytofiltration, a very environmentally friendly and low-
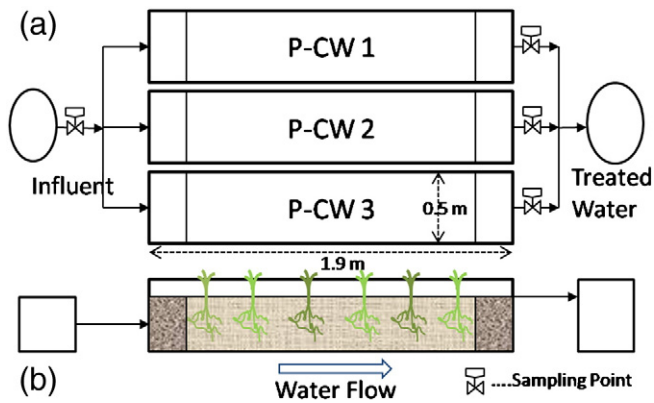
Fig. 3. Plant view (a) and section (b) of the constructed wetland prototypes for rhizofiltration by *Eleocharis machrostachya* in Chihuahua, Mexico.

cost alternative technique with an increasing development during the last two decades. The discovering of tolerating, or even hyperaccumulating plants, shows that it is possible to phytoremediate As-contaminated substrates. The process involves several steps, starting with the selection of the most promising plants capable of removing the contaminant from water and retaining it in their roots. The plants are then transplanted into a constructed wetland, where As from the polluted water will be removed. The plants in this process mainly absorb and concentrate the metals in their roots, but also translocate some low quantities to their shoots (Dushenkov et al., 1995; Salt et al., 1998). Only in recent years, plants able to hyperaccumulate As were discovered, like *Pteris vittata* (Ma et al., 2001) and other ferns (Francesconi et al., 2002; Srivastava et al., 2005; Zhao et al., 2002). Aquatic macrophytes have been particularly considered for removal of As from contaminated surface water bodies. In LA, some studies for removal of As from contaminated surface water bodies were performed using different species of aquatic macrophytes: Water Hyacinth (*Eichhornia crassipes*) and Lesser Duckweed (*Lemna minor*) (Alvarado et al., 2008), dried algae (*Lessonia nigrescens*) (Hansen et al., 2004) and dried macro-algae (*Spyrogira* spp.) (Bundschuh et al., 2007). Other examples are given in Section 5.5.

Other groups of plants, including trees, have been noticed to tolerate As until certain levels of toxicity, making them suitable for phytostabilization of As-contaminated sites (Gonzaga et al., 2006).

Three more detailed examples follow: i) experiments with *Eleocharis machrostachya*, performed in Chihuahua, Mexico, ii) phytoremediation to reduce As contamination due to gold mining activities in Minas Gerais State, Brazil, and iii) studies performed in Argentina testing the potential of aquatic macrophytes.

5.3. Arsenic removal in Mexico through rhizofiltration by Eleocharis machrostachya

Alarcón-Herrera et al. (2007, 2009) evaluated the As tolerance of two Cyperaceae species, *Schonoeplectous americanus* and *E. machrostachya*, collected near the towns of Naica and San Diego de Alcalá (Chihuahua state, northern Mexico). Since 97% of the plants survived the As exposure, and since plant growth was not visibly affected, it was concluded that both species are tolerant to As and can be used for rhizofiltration to remove As from drinking water.

A further study aimed at determining the behavior of *E. machrostachya* in a constructed wetland prototype under the local environmental conditions of Chihuahua, a semi-arid region. Plants were collected from native environments, propagated and transplanted into pots with coarse sand as substrate, always covered with a depth of 5 cm of water to simulate flood conditions for 50 days. Then, the plants were transplanted into the constructed wetland prototypes and acclimated for 5 weeks. The study was conducted over

7.5 months in a system consisting of three prototypes of constructed wetland (Fig. 3).

The units (P-CW1, P-CW2, P-CW3) operated in parallel with subsurface flow. The support medium was lime sand. P-CW1 and P-CW2 were planted with *E. machrostachya*, with a plant density of 4000 plants m^{-2}; P-CW3 was a control, with a similar lime sand substrate but no plants. Two influx As concentrations (100 and 500 $\mu g\,L^{-1}$, prepared with water from underground water wells of Chihuahua City) and two retention times (2.3 and 1.0 d^{-1}) were used. Water samples were collected at the entrance and exit of the wetland units daily. At the end of the experiment, the total As content retained by soil and plants (aerial parts and roots) was measured. For this purpose, both sections of the plants were washed, dried and submitted to digestion with HNO_3 for 12 h, followed by digestion in a microwave oven (CEM, 2002). Analyses of the samples with low As quantities (<1000 $\mu g\,L^{-1}$) were performed by atomic absorption spectrophotometry with hydride generator. A plasma emission spectrophotometer (ICP-OES) was used for the soil and plant analysis, as well as for samples with high As concentrations.

The number of *E. machrostachya* individuals in the wetland units doubled during the experiment and reached in average a length of 1.6 m. The average As concentrations in the roots of the plants and in the soil were 47 and 13 mg kg^{-1}, respectively, indicating that the plant retained 3.7 times the amount of As retained by the soil around it. These results indicate that *E. machrostachya* acted during the experiment as an efficient rhizofiltrator, retaining a great quantity of As in its roots.

E. machrostachya complied with most of the requirements to be an ideal plant for rhizofiltration: it was able to accumulate and tolerate significant amounts of As and offered easy handling. Remaining to be analyzed are the maintenance and disposal costs. Arsenic removal from the treated water showed an efficiency of 99% for the highest As feedwater concentration (500 $\mu g\,L^{-1}$), and 89% efficiency for the lowest As concentration (100 $\mu g\,L^{-1}$), indicating that higher entry concentrations lead to higher As removal efficiency. The As removal efficiency from water of the wetland with no plants (P-CW3) was of only 23% at the end of the experiment, which shows the inherent role of the plants in As removal.

Once the system becomes saturated, constructed wetlands may produce considerable amounts of contaminated wastes, in the form of biomass and sludge, and then convert a water-based pollution problem to one of solid waste management. Treatment and disposal of the solid wastes (roots and sludge) can become the major component of the overall treatment costs, and therefore should be taken into consideration. Further studies are required to investigate the As saturation level of the wetland system here analyzed (plants and soil). Based on the As breakthrough point, final As concentration

Table 1
Soil available As (Mehlich-3 extractor) that provided a reduction of 50% of roots and shoot dry matter (SAs50%) observed for different species in greenhouse experiments.

Species	SAs50% (mg dm^{-3})	
	Roots	Shoot
Pigeon pea (*Cajanus cajan* (L.) Millsp.)	3.5	2.3
Lead tree (*Leucaena leucocephala* (Lam.) de Wit)	70.2	74.9
Wand riverhemp (*Sesbania virgata* (Cav.) Pers.)	317.6	67.0
Eucalyptus grandis Maiden	307.6	156.2
Eucalyptus cloeziana F. Muell.	86.7	92.4
Velvetbean (*Stilozobium aterrinum* Piper & Tracy)	93.0	72.9
Jack bean (*Canavalia ensiformis* L.)	32.3	37.4
Crotalaria (*Crotalaria spectabilis* Roth.)	50.9	52.9
Townsville stylo (*Stylosanthes humilis* H.B.K.)	57.8	25.0
Forage peanuts (*Arachis pintoi* Krapov. & Gregory)	21.6	39.5
Oat (*Avena strigosa* Schreb)	57.5	79.8
Ryegrass (*Lolium multiflorum* Lam.)	62.3	50.0

and potential leachability of the residues, recycling or remediation technologies should be designed for its safe handling and disposal.

5.4. Phytoremediation as mitigation approach to reduce arsenic contamination due to gold mining activities in Minas Gerais State, Brazil

The history of Minas Gerais State (southeastern Brazil) is closely related to gold exploitation, beginning at the end of the 17th century (Meneses and Matschullat, 2007), with a present 48% of the total reserves of gold of Brazil (>30 Mg annual production; PORMIN, 2008). Most of this production comes from the *Quadrilátero Ferrífero* region (QF, Iron Quadrangle in English), located in the southern part of the Minas Gerais State. The QF is also known due to a derived environmental problem: As contamination coming from arsenopyrite ($AsFeS_2$), a common present mineral in those sites (Bundschuh et al., 2008; Vial et al., 2007). Mining operations produce large deposits of rock wastes, where these sulfide minerals are oxidized and can cause acid drainage and As solubilization (Mello et al., 2003). The As/Au ratio in QF gold ores varies considerably from 300 to 3000 and it was estimated that, throughout the three centuries of gold mining, at least 390,000 Mg of As was discharged into the drainage system, increasing As concentration in the different environmental compartments (Borba et al., 2000).

Usually, trying to mitigate acid drainage, large amounts of lime are applied to the mine waste, which is settled on an impermeable layer to avoid direct contact with the soil. Afterwards, the tailing surface must be revegetated to reduce the free energy of the system, fixate carbon, mitigate sulfide oxidation and improve conditions for the site colonization by other plants and animals. Therefore, by revegetating mine tailings and spoils, it is possible to reduce As contamination levels. However, revegetating sulfide substrates is not easy (Dias and Assis, 2008). In addition to the high As and heavy metal levels of contamination, the substrates are usually very acidic, have extremely low levels of organic matter and mineral nutrients and also unfavorable physical properties (Silva et al., 2004).

Liming, scarification, returning of the topsoil and organic residues application are some of the most common techniques of ameliorating the substrate planting conditions (Campello, 1998; Dias et al., 2007). Nevertheless, given the large extensions of mine tailings and spoils, these operations are very costly and may be impracticable due to the lacking of materials (litter, organic residues, topsoil, etc.). Therefore, success in the revegetation of sulfide substrates depends largely on the use of species that tolerate such inhospitable conditions, based on
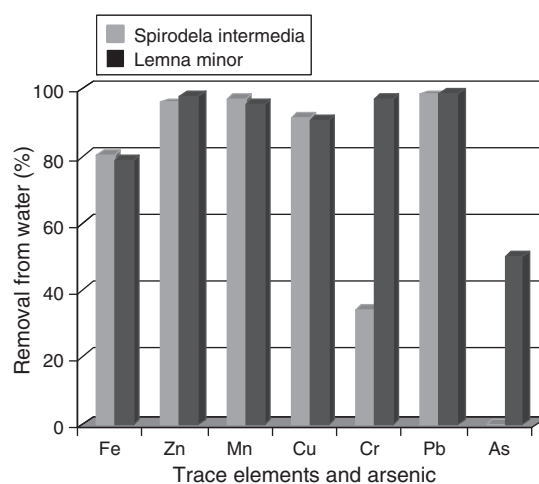

Fig. 4. Results of trace elements and arsenic removal experiences (Miretzky et al., 2004; Pérez Carrera and Fernández Cirelli, 2007; Pérez Carrera et al., 2010).

studies about genetic engineering, biological associations and agronomic practices.

Since 2005, several works have been done at the Soil Department of the Universidade Federal de Viçosa to evaluate the As-tolerance capacity of different herbaceous, shrubs and tree species (Dias et al., 2010; Mello et al., 2006; Melo et al., 2009, 2010). A strong difference of As-tolerance can be observed among the species (Table 1). Pigeon pea showed the lowest tolerance, whereas wand riverhemp and *Eucalyptus grandis* had the highest capacity to tolerate As, even though not filling the requirements to be considered As-hyperaccumulating species. Some of the evaluated species have the potential to be used in revegetation or phytostabilization programs of As-contaminated sites.

Both total contents and plant available content of As in soil are important for the evaluation and preparation of environment rehabilitation programs. The total contents are regulated by law worldwide, USEPA methodologies (3050, 3050B, 3051, 3051a and 3052) being commonly used (United States Environmental Protection Agency, 2008). However, the available contents can assist in actions in the short term, accelerating the recovery process. Nevertheless, quantification of available content is complex due to different soil characteristics and the variety of extractants used, generating large differences in the results. Recent studies by Assis (2010) show that

Table 2
Comparison among different experiments of arsenic removal from water with aquatic macrophytes at laboratory scale.

Species	$[As]_0$ in water ($\mu g\ L^{-1}$)	Time	$[As]_f$ in plants ($mg\ kg^{-1}$)	% removal	Reference
Lepidium sativum	<10 to 800	8 days	0.02 (control)–106 mg kg^{-1} (dry basis)	7.3	Robinson et al. (2003)
Eichhornia crassipes	150	21 days	1,8 mg kg^{-1} (wet basis)	–	Alvarado et al. (2008)
Lemna minor	150	21 days	2,5 Kg^{-1} (wet basis)	–	Alvarado et al. (2008)
Lemna minor	500; 1000 and 2000	13 days	ND	38–52	Pérez Carrera and Fernández Cirelli (2007)
Lemna gibba	10; 500; 100; 250; 500; 1000	21 days	ND	40.3	Mkandawire and Gert Dudel (2005)
Spirodela polyrhiza L.	300	6 days	26.5 (dry basis)	–	Rahman et al. (2007)
Azolla caroliniana	3700	10 days	284 (dry basis)	–	Zhang et al. 2008
Azolla filiculoides	3700	10 days	54 (dry basis)	–	Zhang et al. 2008
Ceratophyllum demersum	100; 300; 1000 and 3000	6 weeks, As solution replaced after 3 weeks	100–1800 (dry basis)	–	Robinson et al. (2005)
Rorripa nasturtium-aquaticum	100; 300; 1000 and 3000	6 weeks, As solution replaced after 3 weeks	100–300 (dry basis)	–	Robinson et al., 2005
Myriophyllum propinquum	100; 300; 1000 and 3000	6 weeks, As solution replaced after 3 weeks	100–400 (dry basis)	–	Robinson et al. (2005)
Spirodella intermedia	500; 1000 and 2000	13 days	ND	Arsenic concentration was the same during the exp.	Pérez Carrera and Fernández Cirelli (2007)

Ca(H$_2$PO$_4$)$_2$ in acetic acid, in contrast with Mehlich-3, Mehlich-1, DTPA and anion exchange resin (AR-103 QDP), had a higher potential to predict the content of As-available in Brazilian soils, which are, in general, highly weathered.

5.5. Studies in Argentina testing the potential of aquatic macrophytes for arsenic removal

At the Center of Transdisciplinary Studies on Water (CETA, Argentina), several studies were performed with some species of aquatic macrophytes to evaluate As removal from water. Floating macrophytes species tested, *L. minor and Spirodela intermedia*, were collected in Pampean ponds of the Buenos Aires province. Once in the laboratory, the plants were washed with distilled and deionized water to remove residual solid waste from the lagoon, placed in 2.5 L reactors with water from the lagoon (*ca.* 25 g in each reactor), and exposed for 8 h/d to fluorescent light. Working pH ranged 7–7.5. Arsenic concentrations tested for both species were 0.5, 1 and 2 mg L^{-1}, and experiments were performed in duplicate. For each species, blanks prepared with macrophytes in water without As were used. Controls with As at the three tested concentrations and no macrophytes were also prepared.

Water samples of each reactor were taken at 0, 2, 4, 6, 24 h and 3, 6 and 13 days. After this time, the experiment was stopped because no variations in As concentration were observed. The concentration of As in water was determined throughout the experiments to calculate the removal percentages for each studied species.

The best removals were observed with *L. minor* (38 to 52%), while *S. intermedia* gave practically no removal; the extent of removal depended on the initial As concentration (Table 2) (Pérez Carrera and Fernández Cirelli, 2007; Litter et al., 2008; Pérez Carrera et al., 2010).

The studied macrophytes are widely distributed and easily cultivated. Nevertheless, studies under the same experimental conditions showed that other trace elements (Cu^{2+}, Zn^{2+}, Pb^{2+}, Fe^{2+}, Mn^{2+} and Cr^{2+}) are more efficiently removed from water than As (Fig. 4, Miretzky et al., 2004). Further studies are needed to understand the sorption mechanism in different conditions to improve the use of macrophytes for As removal. It must be emphasized the convenience of these methodologies due to their low-cost and simple operation and maintenance.

6. Zerovalent iron and photochemical technologies

6.1. General

Zerovalent iron is an emergent material increasingly used for the treatment of several pollutants, particularly toxic metals, and one of the main components of permeable reactive barriers (PRBs). The method is useful to remove both As(V) and As(III) from water. In the case of As(III), removal takes place mainly by adsorption and coprecipitation onto iron hydroxides or oxides formed by Fe(0) corrosion in water and in the presence of oxygen (iron rusts, magnetite, lepidocrocite, maghemite, ferrous hydroxide, ferric hydroxide, etc., depending on redox conditions and pH) (Cortina et al., 2010; Litter et al., 2010a, 2010b, 2010c; Morgada et al., 2009). However, Karschunke and Jekel (2002), reporting results of lab-scale experiments for As removal from drinking water by iron hydroxides produced by enhanced corrosion of elemental iron (as fine iron wool), remark that a continuous and controlled rusting of iron in water is more difficult than expected as the corrosion rate decreases significantly in time.

Experiments performed in LA laboratories use cartridges filled with iron-coated sand, local iron materials, iron fillings, iron wool, packing wire, and iron nanoparticles (Litter, 2006a, 2010b; Litter and Jiménez González, 2004; Litter and Mansilla, 2003; Litter et al., 2008; Litter et al., 2010a; Morgada et al., 2008, 2009; Morgada de Boggio et

al., 2009; Morgada de Boggio et al., 2010). Some of them will be described in more detail in the following sections.

On the other hand, very cheap technologies combining the use of iron and light, abundant in many LA regions, can be adapted for As removal. Among them, the Solar Oxidation and Removal of Arsenic (SORAS), its modification by combination of iron and UV irradiation, and heterogeneous photocatalysis with TiO$_2$ combined with iron species are suitable methods to eliminate As from waters (Litter et al., 2010a, 2010b, 2010c; Morgada and Litter, 2010).

The SORAS technology is a very simple process, where contaminated water is put in transparent PET bottles together with some drops of lemon juice (citric acid), and irradiated under sunlight for several hours. If waters contain enough amount of iron, Fe(III)-citrate complexes are formed that under solar irradiation lead to photo-Fenton processes, where oxidizing species transform As(III) to As(V) together with the precipitation of iron oxyhydroxides, followed by As(V) adsorption and coprecipitation on the flocs. During the night, bottles are put in a vertical position to promote the settlement of flocs, and clear water is obtained by further decantation or filtration. Tests of this technology have been performed in LA in Mexico, Nicaragua, Peru, Chile and Argentina (Clido et al., 2003; Cornejo et al., 2004; Cornejo et al., 2008; García et al., 2003; García et al., 2004a, 2004b; Gelover et al., 2006; Lara et al., 2006; Litter, 2002, 2006a, 2006b; Litter and Jiménez González, 2004; Litter and Mansilla, 2003; Litter et al., 2010a, 2010b, 2010c).

However, groundwaters of several regions of LA do not have enough iron in solution to make efficient the SORAS technology. To increase As removal, iron has to be added externally, as drops of a Fe(III) water solution or as zerovalent iron. For this, some natural Fe-containing minerals, iron wool, packing wire or zerovalent iron nanoparticles can be used and were tested (Bundschuh et al., 2009; Litter et al., 2008, 2010a, 2010b, 2010c; Morgada de Boggio et al., 2009, 2010; Morgada et al., 2008, 2009; Morgada and Litter, 2010). Some of these results will be discussed later.

Another possible photochemical process to oxidize As(III) in water is heterogeneous photocatalysis (HP), an Advanced Oxidation Technology that uses a particulate semiconductor, generally TiO$_2$ (Litter, 2009; Litter et al., 2010a, 2010b, 2010c; Morgada and Litter, 2010. In HP, after excitation of TiO$_2$ with UV light (energy equal to or higher than the TiO$_2$ bandgap), conduction band electrons (e$_{cb}^-$) and valence band holes (h$_{vb}^+$) are created, which react with species present in the system. Couples with redox potentials more positive than that of e$_{cb}^-$ can be photocatalytically reduced and those with redox potentials more negative than the redox potential of h$_{vb}^+$ can be oxidized. Hydroxyl radicals, HO$^\bullet$, are also formed by h$_{vb}^+$ to water. Application of HP to the case of As(V) implies complex mechanisms; among them, h$_{vb}^+$ or HO$^\bullet$ can promote oxidation of As(III) to As(V). If Fe is present in the water, flocs are formed where As is adsorbed and coprecipitated. Results of these studies will be presented in Section 6.6.

In what follows, some examples regarding application of the zerovalent iron technology and some photochemical technologies in LA are provided.

6.2. Low-cost continuous small-scale plant based on zerovalent iron for arsenic removal in water built and applied in San Juan and other Argentine provinces

The development of the procedure used to operate the plants is presented in Cáceres (2007), and it is based on thermodynamic studies performed by means of Pourbaix diagrams. During the kinetic study stage (with tests at laboratory and bench scale), two mathematical models were developed, one for the formation of the Fe$_2$O$_3$ sol and the other for the adsorption of arsenates into the sol and further coagulation. Combination of both models led to a global one that allowed predicting the amount of arsenic separated from the raw

water. Once the procedure was developed, results were validated in field.

Plants working with this technology were built in different provinces of northwestern Argentina (Cáceres, 2007; Litter et al., 2008; Cáceres, 2010). The procedure is ideal to be applied on a small scale (household to few hundred people) and consists of a first oxidative chlorination step, a second step using a bed filled with iron fillings or small pieces of iron, conditioning of the flocs and filtration. The small plants work with low-cost materials easy to obtain such as sodium hypochlorite and iron wool or iron filings; they were built with standard plastic materials (PVC and polypropylene pipes and fittings used in plumbing), do not use electric power, may be operated by people without technical knowledge, and the residues do not cause environmental problems. It can be calculated that 1 L of commercial bleaching ($45\ g\ L^{-1}$) is enough to treat approximately 5000 L of treated water and only 1 kg of iron is needed for approximately 100,000 L).

At the end of the process, As, hypochlorite and iron levels in water are in accordance to current standards. Replenishment of the metallic iron consumed should be made every year. Follow-up studies of the plants assembled in field show that, even when operated by non-experts, the percentage of As removed from water is higher than 90% for the cases presented below. Therefore, the plants were found appropriate to remove As from waters for human consumption at small scale in disperse populations.

A prototype of the plant was set up in "Dr. Juan Carlos Navarro" school in the district of El Encón, San Juan, in the western part of Argentina (32°12′00″S and 67°46′60″W). The reactor had a volume of 2 L and a production capacity of $6\ L\ h^{-1}$. Initially, the plant was made to operate with raw water from El Encón ($280\ \mu g\ As\ L^{-1}$, $2.4\ mg\ F\ L^{-1}$, $9.2\ mg\ SiO_2\ L^{-1}$, pH 8.3) and, in a second study, water coming from an aqueduct in Camarico (27 km from El Encón, containing $57\ \mu g\ As\ L^{-1}$, $1.1\ mg\ F\ L^{-1}$, $9.8\ mg\ SiO_2\ L^{-1}$, pH 8.1) was tested. Follow-up studies of the facility throughout a year allow reassuring that the percentage of As removed from water with this prototype is well above 90% for both raw waters studied.

A second prototype was set up in the school lodging house. The plant production capacity was approximately $60\ L\ h^{-1}$ (reactor volume: 18 L). The facility worked with raw water from Camarico and was operated by the lodging house staff. The percentage of As removed from water in this case was also above 90%.

Fig. 6. Time curve for As removal under optimized conditions (2.0 g of steel wool and 1 drop of lemon juice in one liter of water), using water of the Camarones river (pH≈8,0): (■) with citric acid ($4.5\ mg\ L^{-1}$) without sun exposure; (◊) in the absence of citric acid under sun exposure (6 h); and (●) with citric acid ($4.5\ mg\ L^{-1}$) under sun exposure (6 h).

A number of additional laboratory experiments were made with water from other sources (two places in Mendoza and one in Santiago del Estero, west and northwest of Argentina, respectively), starting from 190, 560 and $750\ \mu g\ L^{-1}$ initial total As in each case. The tests were carried out in a small plant with a reactor volume of 1.5 L, and operating at a flow rate of $6\ L\ h^{-1}$. The final As contents were <10, <10 and $20\ \mu g\ L^{-1}$, respectively, the first two in agreement with the requirements of the Argentine legislation ($10\ \mu g\ L^{-1}$, Código Alimentario Argentino, 2007).

Through an agreement with the Argentine Ministry of Health, work is being carried out to provide four plants with a reactor volume of 12 L to be operated at a flow rate of $40\ L\ h^{-1}$. The plants have been built in Lavalle, in the province of Mendoza.

Regarding residues, population has been trained for disposal of solid wastes after washing the filters. At household scale, the residues have to be deposited in the lowest part of a dunghill or in the latrine; at medium scale, as in small localities, the washings of the filter can be sent to the cesspit.
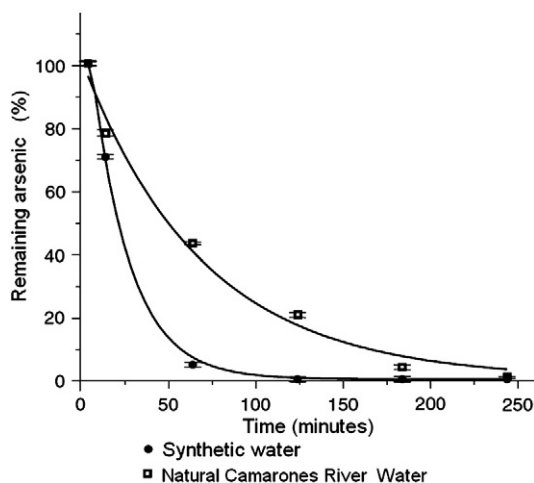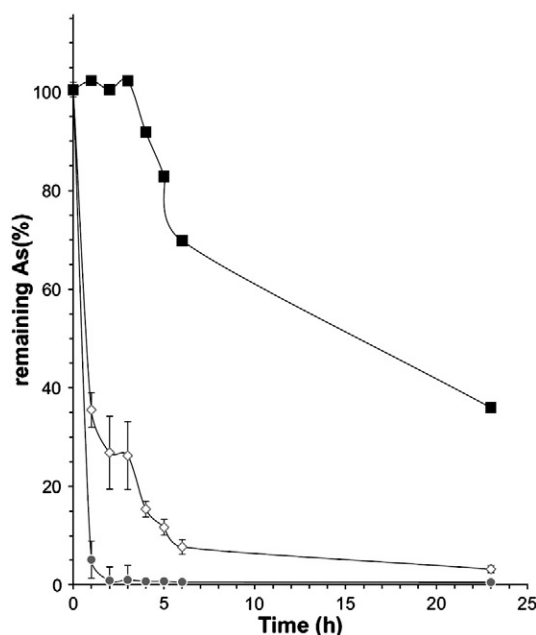

Fig. 5. Comparison of As removal in synthetic and natural water from the Camarones River applying solar radiation (Lara et al., 2006). Synthetic water: $[As(III)] = 500\ \mu g\ L^{-1}$, $[Fe(II)] = 9.5\ mg\ L^{-1}$, and $[citrate] = 2.5\ mg\ L^{-1}$. Natural water: $[As(V)] = 1000\ \mu g\ L^{-1}$, $Fe(II) = 19\ mg\ L^{-1}$, and $[citrate] = 4.5\ mg\ L^{-1}$ (as lemon juice). Irradiation performed at the city of Concepción (36.83 S, 73.03 W), average solar irradiation: $4.13\ kWh\ m^{-2}\ day^{-1}$.

Fig. 7. Removal unit used in the modified SORAS technology (Cornejo et al., 2010).

6.3. Modification of SORAS in northern Chile

In northern Chile, there are several rural communities without access to potable water systems that allow safe water consumption. Some of these communities are settled in the Camarones Valley (Atacama Desert, Arica and Parinacota regions), with a population of 1220 inhabitants. These communities use water from the Camarones River for agricultural activities, animal and human consumption. However, this water resource has high total As contents, between 1000 and 1300 mg L^{-1}, coming from the various substrates (water, soil, sediments and atmospheric materials) that compose the local environment. This causes that the villages settled in Camarones are exposed to high As levels; intake of water is one of the leading source of the entrance of inorganic As(V) to the body (Cornejo et al., 2006a; Mansilla and Cornejo, 2002; Mansilla et al., 2003; Yañez et al., 2005; Cornejo et al., 2008).

A series of researches has been carried out to solve the problem of As in isolated villages of northern Chile by modification of the SORAS technology, taking into account that sunlight is a widely available resource in those regions.

In an early stage, laboratory studies have been conducted using artificial light sources (black light lamp, 360 nm), iron(II) and citric acid (Cornejo et al., 2004; Lara et al., 2006; Mansilla et al., 2003). The optimized conditions for As(III) removal were established using a factorial experimental design; the polynomial function obtained indicated that iron concentration is the most critical parameter for As removal. Further experiments with natural water (approximately 1000 µg L^{-1} As(V)) under sunlight and in the optimized conditions rendered approximately 95% removal after 1 h (Fig. 5).

In a second stage, a removal methodology was developed, based on the use of solar radiation, zerovalent iron in the form of steel wool and lemon juice, which was applied to natural waters from the Camarones river (Cornejo et al., 2006a, 2008). The analysis of a response surface method allowed to optimize the amount of steel wool and citrate concentration (lemon juice) to 1.3 g of steel wool and a drop of lemon juice (0.04 mL lemon juice) in one liter of water. Experiments under these conditions yielded removals higher than 99.5%, with a final As concentration in solution below 10 µg L^{-1} (Fig. 6).

It was concluded that this method for As removal is highly effective, easy to use and has low implementation costs. At present, campaigns are being developed in various communities of Arica and Parinacota to disseminate the technology, including training workshops and delivery of information regarding As removal units at household level. A scheme of the removal unit is shown in Fig. 7 (Cornejo et al., 2006b; Cornejo et al., 2010).
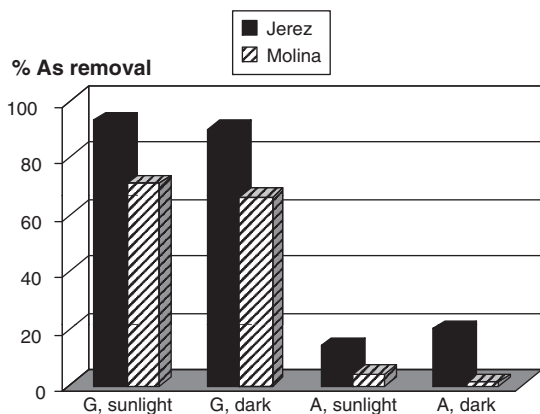
The amount of wastes generated in the solar zerovalent iron process is low and easy to be disposed off. Similarly, As fixed to the iron oxide presents very low desorption capacity; due to this low mobility, the environmental risk of As-containing residues is also very low in this technique.

6.4. Modification of SORAS to treat well waters of Tucumán, Argentina

Groundwaters of the east of the Tucumán Province (Salí river basin, Chacopampean plain, northeast of Argentina) are naturally contaminated with As. The area is occupied by isolated rural villages, and one of them, Los Pereyra (2500 inhabitants), was selected as study site (Blesa et al., 2002; de la Fuente et al., 2006).

The lack of drinking water network and the absence of surface bodies makes necessary to extract water from deep and shallow wells. The quality of this water is poor due to a high bacteriological content, nitrates, boron, fluoride and trace elements (e.g., Mn, As) (García et al., 2003). In particular, As fluctuates between 70 and 1000 µg L^{-1}, concentrations much higher than the 10 µg L^{-1} limit established by Argentine regulations (Código Alimentario Argentino, 2007). The low water quality, poverty and malnutrition cause a high incidence of hydric illnesses, in particular, of HACRE.

The efficiency of SORAS was evaluated with well waters of Los Pereyra. As these waters did not contain enough iron to make efficient the technology, in the first investigations (García et al., 2003, 2004a, 2004b) the efficiency was evaluated with laboratory waters of similar composition, adding different amounts of Fe(III) and lemon juice. The best conditions (As removal >90% for [As]$_0$ ranging between 80 and 1000 µg L^{-1}) were found with 3 mg L^{-1} of Fe(III) and 750 µL L^{-1} of lemon juice under more than 3 h of solar exposure. In natural well waters, however, the efficiency was much lower (close to 30%), suggesting that the influencing factors were the low iron concentration and the chemical matrix of the water. Addition of small amounts of citric acid (as lemon juice) was beneficial, but high concentrations were deleterious. Generally, the procedure was better under sunlight.

Later on, since it had been demonstrated that external addition of iron was needed for a good As removal, the studies focused on the search of low-cost iron materials to be applied in the technology. Typical regional rocks and sediments (pelites and sandstones from Lules and Potrero de las Tablas, 60 km east of Los Pereyra), non-galvanized packing iron wire (used in agricultural activities in the zone) and common iron nails were tested, which were compared with synthetic goethite (γ-FeOOH) and with the addition of FeCl$_3$ solution. Results of experiments with laboratory waters showed that the efficiency was better with sunlight (4 h) and in the presence of lemon juice (García et al., 2004a, 2004b). Neither sandstone nor iron nails were found to be good materials for As removal. The low efficiency

Fig. 8. SORAS results in well waters of Los Pereyra (Jerez and Molina families) in field conditions. G: goethite (1 g L^{-1}), A: sandstone (5 g L^{-1}). Solar irradiation time: 4 h; 750 µL L^{-1} of lemon juice.
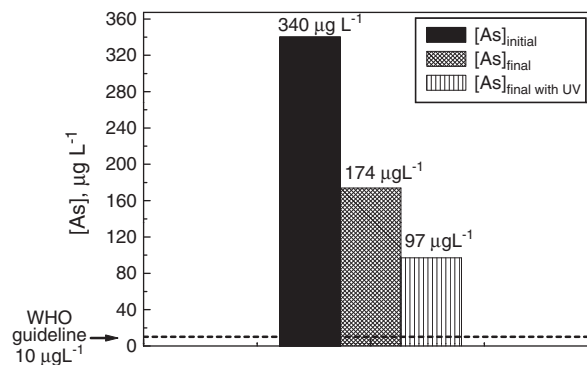

Fig. 9. As removal efficiency with packing wire (6.0 g L^{-1}) in waters of Los Pereyra wells. Conditions: [As$_{total}$]$_0$ = 340 µg L^{-1}; pH 7.8; 6 h sunlight (1600–2000 µW cm^{-2}) or in the dark; 600 mL PET bottles, V$_{solution}$ = 250 mL. No further settlement in the dark.

found with $FeCl_3$, added at 0.3 mg L^{-1}, a concentration regularly found in natural well waters, was attributed to an amount of iron much lower than the optimal for the SORAS technology. In addition, a yellow color was observed at the end of the experiment. Removal in the presence of pelites was reasonably good; however, a color in the final water was also observed. Synthetic goethite yielded an excellent removal, but the precipitation of suspended matter took some days, with a high remaining turbidity and color; moreover, final Fe in solution exceeded amply the limits for drinking water (<0.3 mg L^{-1}, Código Alimentario Argentino, 2007). In contrast, although As removal with packing wire was lower than in other cases, the resulting water did not present turbidity or high iron content in solution.

After the experiments in laboratory waters, tests with sandstone (5 g L^{-1}) and goethite (1 g L^{-1}) were performed in well waters of Los Pereyra families (Jerez and Molina) using 750 μL L^{-1} lemon juice. Fig. 8 shows the results, which indicate again that sandstone is not an adequate material, and that goethite combined with solar irradiation and low amounts of citric acid yields a very good removal. However, an orange color together with a very fine suspended particulate was observed, and neither good filtration nor decoloration was possible. In contrast with the results in laboratory waters, the final Fe concentration in solution (average <0.2 mg L^{-1}) did not exceed the allowed limits. In waters of the Jerez family, As removal was better than in those of the Molina family, probably due to a lower initial amount of As in the first sample (96 vs. 709 μg L^{-1}).

Further experiments were performed with iron packing wire and no other reagent since it had been proved that lemon juice was not strictly necessary for As removal. Packing wire was considered one of the most available, cheapest and easily to be handled iron material in the Chacopampean plain. Iron wool for household uses, in the form of commercial Virulana®, was tested as another low-cost material.

Laboratory waters (250 mL, $[As(V)]_0 = 1000$ μg L^{-1}, pH 7.8) were poured in 600 mL PET bottles. Packing wire or iron wool was added at 0.6 g L^{-1}, the optimal concentration found in previous SORAS tests in Los Pereyra (d′Hiriart et al., 2009). Bottles were exposed to UV light (366 nm; 800 μW cm^{-2}) for 8 h, followed by 24 h settlement in the dark; similar experiments were performed without sunlight exposure. It was found that As removal was considerably faster with iron wool, reaching complete removal in 2 h while with packing wire only 30% was attained in the same time. After further 24 h settlement in the dark (where the adsorption process on the iron flocs was completed), the amount of removed As was 83% and 100% with packing wire and iron wool, respectively. However, iron wool was difficult to be removed because it disintegrated in very fine particles. No significant effect of UV light was observed in these experiments. However, when Los Pereyra well waters were tested similarly, a significant higher removal efficiency was observed under sunlight
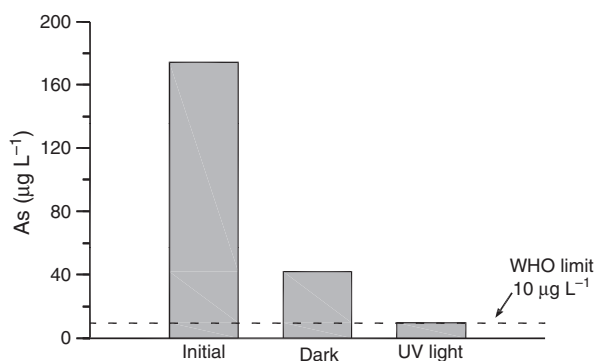

Fig. 10. Results with waters from Los Pereyra using zerovalent iron nanoparticles. Conditions: $[As_{total}]_0 = 174$ μg L^{-1}; $[NanoFe®] = 0.025$ g L^{-1}, irradiation time: 3 h, artificial UV light (5000 μW cm^{-2}).

irradiation for 6 h (Buenos Aires City in Spring, $34°38'S$, $58°28'W$, 1600–2000 μW cm^{-2}) compared with the same experiment in the dark (Fig. 9). This can be attributed to the operation of a photo-Fenton reaction caused by the presence of natural organic matter (NOM) in the well water.

6.5. Use of zerovalent iron nanoparticles in Argentina

Commercial zerovalent iron nanoparticles exhibited outstanding properties to treat As-polluted water (Morgada et al., 2009; Morgada de Boggio et al., 2010). The nanoparticles (NanoFe®, $S_{BET} = 55$–63 m^2 g^{-1} and 5–15 nm particle size) were prepared by a local company according to a novel technology.

Experiments of As removal were performed with laboratory waters in 250 mL glass Erlenmeyer flasks in the dark with different masses of NanoFe® (0.005–0.1 g L^{-1}, $[As(V)]_0 = 1000$ μg L^{-1}, pH 7.8). Results indicate a very rapid removal, which increased with $[NanoFe®]$. For $[NanoFe®] = 0.05$ and 0.1 g L^{-1}, removal was higher than 90% after 150 min of contact time. The needed iron mass was two orders of magnitude lower than that used with packing wire or iron wool (see Section 6.4), explained not only by a higher surface area but also by a high intrinsic activity of the material.

Similar experiments under UV irradiation ($\lambda = 365$ nm, 5000 μW cm^{-2}) indicated that light enhanced only slightly the As removal efficiency. However, removal was much improved after addition of humic acids, indicating the operation of a photo-Fenton process induced by NOM. Results with Los Pereyra water samples (Fig. 10) showed a very rapid As removal in the dark after 3 h of contact time, but under UV light irradiation, removal was better and the required limit for drinking water was attained.

Additional advantages of the use of nanoparticles are a significantly low mass of wastes formed at the end of the process and the possibility of removing magnetically the flocs.

6.6. Tests with heterogeneous photocatalysis combined with zerovalent iron in Argentina and in Brazil

To perform HP experiments, walls of PET plastic bottles (600 mL of capacity) were impregnated with TiO_2 by a very simple technique (Meichtry et al., 2007). As(III) solutions (250 mL, $[As(III)]_{initial} = 1000$ μg L^{-1}, pH 7.8) were placed in these bottles and irradiated during 6 h by UV light in a horizontal position. (366 nm, 800 μW cm^{-2}). After irradiation, 1.5 g packing wire was added. After 24 h settlement in the dark, removals ranging 80–86% were obtained. Similar results were obtained when packing wire was added at the beginning of the irradiation. The same bottle could be reused at least three times without loss of efficiency (Litter et al., 2010a, 2010b; Morgada de Boggio et al., 2006; Morgada et al., 2008; Morgada de Boggio et al., 2009).

The HP procedure was proposed to remove As (500–1800 μg L^{-1}), in well water samples of Las Hermanas, a locality close to Santiago del Estero City, capital of the province of the same name ($27°46'60''S$ $64°16'00''W$, Argentina). When water samples were poured into the bottles and exposed to sunlight (Buenos Aires City in spring, 1600–2000 μW cm^{-2}), followed by $FeCl_3$ addition (in ca. 100–200 Fe:As molar ratio) at the end of the irradiation, more than 94% As removal took place. It was concluded that natural waters yielded better results than laboratory waters and, again, the higher efficiency could be explained by the presence of NOM giving rise to photo-Fenton processes (Litter, 2006a, 2006b; Litter et al., 2008, 2010a, 2010b; Mateu, 2007; Morgada de Boggio et al., 2006; Morgada et al., 2008; Morgada de Boggio et al., 2009, 2010).

Fostier et al. (2008), obtained similar results with laboratory samples under solar irradiation. They applied the HP system to natural waters collected at Ouro Preto and Mariana (Minas Gerais, Brazil, $[As]_0$ ranging 40–63 μg L^{-1}), adding Fe(II) (7.0 mg L^{-1}) and

exposing the TiO_2-coated bottles to sunlight for 2 h. The authors also analyzed the influence of anions present in the waters such as F^-, Cl^-, NO_2^-, NO_3^-, PO_4^{3-} and SO_4^{2-}. After the treatment, As removal was higher than 99% in all cases, showing that the investigated water matrix did not interfere in the oxidation process.

Although somewhat more complex than the previous solar methodologies, HP technology allows the simultaneous elimination of As together with other pollutants such as pesticides and even microbiological contamination.

7. Discussion

Relevant examples of emerging, low-cost technologies proposed to mitigate the problem of As in drinking water in isolated rural and periurban areas lacking centralized water supplies in LA have been described in this article. These technologies were designed to solve problems for small towns, communities, settlement or isolated houses with less than 1000 inhabitants and not connected to a centralized water distribution network.

The methods are based on existing technologies, previously used at large or medium scale, either conventional or novel, such as the combination oxidation–adsorption–coagulation/flocculation, use of natural or synthetic low-cost adsorbents, non-expensive electrochemical technologies, biological methods, especially phytoremediation, use of zerovalent iron and solar technologies.

The most relevant aspects of this type of technologies are: 1) they are affordable for populations with low economic income and resources, 2) they rely on local materials or available free energy resources like sunlight, 3) they work with small-scale equipment and treat small volumes of water to supply small communities or individual houses, 4) their design is simple and the installation and maintenance can be easily handled by the local population, and 5) the technology is environmentally friendly, i.e., they are accepted by the population. Some of the methods include addition of chlorine for As(III) oxidation, which assures simultaneous disinfection, being an important advantage in many rural and remote areas of LA where hydric diseases are chronic.

The main disadvantage of the presented methods is the risk induced by the possible inadequate handling and disposal of the generated wastes. Therefore, the efficient communication of the involved risk during the technological transference of the processes of As removal is mandatory. The infrastructure for disposal should be also clearly available.

Coagulation/filtration with iron and aluminum salts, is a well-established, efficient method for large-scale water treatment. In the described applications of this article, the method has been scaled-down for household applications. Simple operation and basically no maintenance needs make this option attractive to remote, domestic applications, if the reagents (e.g. iron and aluminum salts) can be provided at low cost.

Capacitive deionization resulted in an efficient and potentially economic technology for poor populations, economically competitive with reverse osmosis. The low energy consumption in the treatment for water containing high As concentration provides a low-cost technology, with easy operation and maintenance.

The use of plants, including phytoremediation using constructed wetlands constitutes another promising innovative method for rural communities. Plants can be also used for revegetation or phytostabilization programs of As-contaminated sites. The systems based on use of plants do not require maintenance by highly trained personnel. It can be also designed to work by gravity, with minimal energy consumption. **Zerovalent iron** is a very promising material combining the advantages of natural, low-cost materials, with the simple operation of sorption processes (no need of electricity and consumables). The natural oxidation of iron to colloidal oxyhydroxides provides an *in-situ* production of a highly efficient material for As

removal. When associated with **photocatalytic oxidation** (i.e. using sunlight), the method also allow simultaneous As(III) oxidation and water disinfection. Fixed bed, ZVI operation combined with chlorination is one of the emerging technologies deserving special attention, when the supply of water on a community-scale basis is considered. The use of zerovalent nanoparticles results in an outstanding increased removal rate, but more research is needed.

TiO_2 heterogeneous photocatalysis is also a promising, emergent technology that allows the simultaneous As(III) oxidation and removal of organic pollutants, toxic metals and microbiological contamination. A low-cost iron material has to be added to retain As on a solid surface. This technology should be still explored in field tests.

8. Conclusions

Although the problem of As in LA drinking waters was rather successfully solved in urban areas and others with access to centralized water supplies, most of these technologies are rather expensive and sophisticated to be applied in isolated rural and periurban areas, where the population still depends on untreated water containing As in toxic levels. Generally, these localities are inhabited by low-income people, comprising the periphery of large cities, small cities, towns, villages and disperse settlements of few families.

A lot of effort has been made by several local scientists and technologists in LA countries affected by the As problem to develop low-cost technologies to palliate this dramatic situation. However, most of the proposed emergent technologies are still at the laboratory stage and have been neither tested nor implemented in field. Some of these examples have been reported in this review with the intention of calling the attention of the existence of these solutions. The cited literature comprises not only refereed international journals but also meeting proceedings, web sites, master thesis; many articles are written in Spanish or Portuguese, and do not allow an easy comprehension of the achievements. The proposed methods are environmentally friendly, and they rely on the use of locally available low-cost materials or sunlight, abundant in the LA region. Without any doubt, if once applied, they will have the proper social acceptance by the population, leading to minimal waste production. As some technologies have been tested only at the laboratory level, enough information to estimate the treatment costs including waste generation is not available. However, it should be emphasized that as a general rule for the reliability of a technology, treatability tests in laboratory are mandatory before application.

Concerning the generated wastes, a careful analysis of their amount after the operation of the presented technologies, as well as the lifetime of the materials, should be made for every technology to determine the amount of As present in the residues, its environmental availability, the associated risk in the public health and the environmental impact. It is also important to provide the alternatives to warrant the proper handling and disposal of the wastes together with their associated costs.

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