Review

Possible treatments for arsenic removal in Latin American waters for human consumption

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Low-cost techniques should be urgently investigated to remove arsenic in drinking water in poor disperse rural and urban Latin American populations.

ABSTRACT

Considering the toxic effects of arsenic, the World Health Organization recommends a maximum concentration of 10 $\mu g\,L^{-1}$ of arsenic in drinking water. Latin American populations present severe health problems due to consumption of waters with high arsenic contents. The physicochemical properties of surface and groundwaters are different from those of other more studied regions of the planet, and the problem is still publicly unknown. Methods for arsenic removal suitable to be applied in Latin American waters are here summarized and commented. Conventional technologies (oxidation, coagulation–coprecipitation, adsorption, reverse osmosis, use of ion exchangers) are described, but emphasis is made in emergent decentralized economical methods as the use of inexpensive natural adsorbents, solar light technologies or biological treatments, as essential to palliate the situation in poor, isolated and dispersed populations of Latin American regions.

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1. The problem of arsenic in Latin America (LA)

As it has been largely reported, water pollution by arsenic is a worldwide problem with high impact mainly in the poorest regions of the Planet. Arsenic is classified as a Group I carcinogen (human carcinogen, IARC, 2004) and severe health effects have been observed in populations all over the world drinking arsenic-rich water over long periods (EPA, 2006). The permanent ingestion of waters with high arsenic concentrations provokes the appearance of arsenicosis, an illness with high incidence in Asia and LA; in LA, the disease is named Chronic Endemic Regional Hydro-arsenicism (in Spanish: HACRE). Symptoms of this illness are palm-plantar hyperkeratosis, damage to the central neural system, hepatic damage, hair loss, skin cancer and cancer of internal organs (lungs, liver, kidney and bladder). So far, there is no treatment for HACRE, and prevention is the only way to combat the illness, which involves reduction of As concentration in water or avoidance of people to As exposure (Das et al., 1996).

Arsenic is widely distributed in soils, water, air and biota (especially marine species). It is a component of more than 200 different minerals, as arsenates, sulfides, sulfosalts, arsenides, arsenites, oxides, silicates and elemental arsenic (Onishi, 1969; Yan Chu, 1994). Speciation of the element is a key factor in controlling mobility, availability and toxicity of arsenic in natural environments. Arsenic appears in inorganic as well as organic species, and the main oxidation states are +III and +V, depending on pH and redox properties of the media. Inorganic species (iAs) are much more toxic in general than organic arsenical compounds, and iAs(III) species are of great environmental concern in view of a combination of high mobility and toxicity in comparison with the pentavalent species, which can be more easily adsorbed and retained by different surfaces. Arsenic occurrence and mobilization takes place through a combination of natural processes (e.g., weathering reactions, biological activity, volcanic emissions, etc.). Although natural arsenic is the main source of pollution, anthropogenic activities account for a widespread As contamination, arising from a variety of industrial processes (mining, electrolytic processes, combustion of fossil fuels, wood preservation, urban wastes, medicinal use, sewage sludges, fertilizers, pigments,

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biocides, crop desiccants, glass, alloys, electronics, etc.) (Jacks and Bhattacharya, 1998; Juillot et al., 1999; Mulligan et al., 2001; Smedley and Kinniburgh, 2002).

Keeping in view the toxic effects of inorganic arsenic on humans and other living organisms, the World Health Organization (WHO) recommends a limit of 10 μg L^{-1} of arsenic in water for human consumption (World Health Organization, 2004). This guideline is based on a 6×10^{-4} excess skin cancer risk, which is 60 times higher than the factor typically used to protect public health (US Environmental Protection Agency, 1988). According to this value, the presence of arsenic in water for human consumption affects potentially around 140 million people (Ravenscroft et al., 2009). Four million of them were documented in LA according to the old regulation (50 μg L^{-1}, Bundschuh et al., 2009); however, in line with the new guidelines, much more people could be at risk.

In South America, especially in Argentina, Chile and Peru, the problem is known since several decades ago, affecting mainly urban and rural poor populations not connected to drinking water networks. Due to the ample extension of the territory and the amount of people living there, the problem of As in LA reaches the same order of magnitude as in other regions of the world, such as SE Asia. As we will see in Section 2, various studies have been undertaken by local researchers, leading to suitable treatment methods. However, technologies have been not yet commercialized due to a lack of interest of authorities, local industries and international agencies for financial and technical cooperation.

Excellent publications can be found on the (bio)geochemical origin of arsenic in the various occurrences and effects on health (among others: Bundschuh et al., 2008, 2009; Hopenhayn-Rich et al., 1996; Ravenscroft et al., 2009). In consequence, this topic will be not addressed in this review and only some brief details will be given. A special mention should be made to IBEROARSEN (IBEROARSEN network, http://www.cnea.gov.ar/xxi/ambiental/iberoarsen/default.asp), a thematic network sponsored by CYTED[1]) joining 46 groups of experts of 17 Iberoamerican countries, which handles the problem of arsenic in Iberoamerica from three points of view: 1) distribution of arsenic, 2) analytical methodologies, 3) removal technologies. It the web page recent information about the arsenic problem in the region, including a database on As occurrence can be consulted.

Ravenscroft et al. (2009) define four affected regions in South and Central America: the high volcanic mountains of the Andes, the arid Pacific coastal plains, the tropical river basins of Amazonia, and the semiarid Chaco-Pampean plain. Relevant data are also reported in publications of groups belonging to IBEROARSEN (Bundschuh et al., 2008, 2009). As an example, in Argentina, the regions with the highest arsenic contents in waters belong to the Chaco-Pampean plain ($\sim 1 \times 10^6$ km^2), Puna and Cuyo; other areas are still under investigation (Bhattacharya et al., 2006; Blanco et al., 2006; Bundschuh et al., 2008, 2009; Litter, 2002, 2006a,b; Litter and Jiménez González, 2004; Litter and Mansilla, 2003; Smedley et al., 2002). Arsenic content (mainly As(V)) varies largely, between 4 and 5300 μg L^{-1}; a value of 14 969 μg L^{-1} was reported recently in the Santiago del Estero province (Bhattacharya et al., 2006). In some places, 99% of groundwaters exceed the guideline of 10 μg As L^{-1}. In the north of Chile (Atacama Desert, from Arica to Antofagasta, approximately 250000 km^2), worrying levels of arsenic in drinking water are present, with concentrations 6–300 times higher than the limit recommended by the WHO (Bundschuh

et al., 2008, 2009; Litter, 2002, 2006a,b; Litter and Jiménez González, 2004; Litter and Mansilla, 2003). In Brazil, one of the most problematic zones is the large mining region belonging to the Minas Gerais state, known as the Iron Quadrangle, where values of As in waters up to 2980 μg L^{-1} have been measured; high concentrations have been also found in Ribera valley and in the Amazonic region (Borba et al., 2003; Bundschuh et al., 2008, 2009; Matschullat et al., 2000). Arsenic in Bolivia is present mostly in the occidental Andean region (in the As-Ag-Pb-Zn-Cu-Au belt), where values up to 4800 μg L^{-1} have been found in the Poopó lake (central Andes) and other places (Bundschuh et al., 2008, 2009; Matschullat et al., 2000). In México, the first cases of arsenicosis were identified in 1958 in the Comarca Lagunera (Durango and Coahuila states), where values higher than 700 μg As L^{-1} were found in waters (Bundschuh et al., 2008, 2009; Cebrián et al., 1994). In Zimapán (Hidalgo state), and in Chihuahua and Sonora states, concentrations up to ca. 6000 μg L^{-1} have been found, many of them related to mining (Carrillo-Chávez et al., 2000). In Nicaragua, the first cases of hydroarsenicism were identified in 1996 in El Zapote (Matagalpa), and many sources of drinking water in the zone present nowadays high concentrations, with a reported value of 1320 μg L^{-1} (Bundschuh et al., 2008). In El Salvador, arsenic pollution is amply distributed in surface, groundwaters and sediments, with high concentrations informed in the Olomega lake (4200 μg L^{-1}) and in other regions (Bundschuh et al., 2008). In Costa Rica, concentrations up to 30 000 μg L^{-1} were found in geothermal reservoirs and up to 2000 μg L^{-1} in hot spring waters (Bundschuh et al., 2008).

Investigations are underway or even pending in Uruguay, Paraguay, Perú, Ecuador, Cuba, Honduras, República Dominicana, Colombia and Venezuela, (Bundschuh et al., 2008, 2009; Castro de Esparza, 2003; Litter, 2006a; Matschullat et al., 2000; Smedley and Kinniburgh, 2002), where the presence of arsenic in groundwaters can be foreseen due to geological features. Recent data can be obtained from the IBEROARSEN web site (IBEROARSEN Network, http://www.cnea.gov.ar/xxi/ambiental/iberoarsen/default.asp; Morgada et al., 2008). This short overview only aims to point out that the degree of knowledge about arsenic concentrations in groundwater is still not complete in LA, and much more efforts are mandatory to complete the map of arsenic distribution, extending the study to soils and sediments.

2. Technologies for arsenic removal: which can be suitable for LA?

Arsenic removal from waters is not an easy task. Economical aspects are perhaps the most important factors for the selection of the technology, taking into account size of the population, incidence of chronic illnesses, lack of safe water, poverty conditions, and other socioeconomic variables. In most cases, sophisticated, expensive techniques cannot be applied in populations with low economical resources. In addition, arsenic treatment units require very sensitive monitoring and maintenance arrangements, which falls far beyond the economic scope of poor isolated communities. Moreover, a number of cultural and political factors play deciding roles in the implementation of new technologies (Kemper and Minnatullah, 2005).

It is necessary to remark that either to evaluate arsenic contents in waters and soils or to select a removal technology, it is essential to have suitable methodologies for quantitative measurement of low arsenic concentrations, mainly due to the low detection and levels that must be attained. Speciation and development of cheap in-field technologies are also needed. These issues are beyond the scope of this article, but the interested readers can consult an excellent comprehensive review (Francesconi and Kuehnelt, 2004). IBEROARSEN has also recently published in Spanish a volume

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[1] CYTED, Science and Technology for the Development, is an organization of 21 Iberoamerican countries, financed by the Spanish Government and the National Science and Technology Agencies of Portugal and Latin American nations.

devoted to this topic (Litter et al., 2009; IBEROARSEN Network, http://www.cnea.gov.ar/xxi/ambiental/iberoarsen/default.asp).

From a technical point of view, the physicochemical and microbiological characteristics of the waters and the available materials in the region will determine the most convenient technology for removal of arsenic in each site. The selection of the method depends greatly on arsenic speciation, chemical composition of the water, reduction potential, hardness, presence of silica, sulfate, phosphate, iron and other chemical species, volumes to be treated and degree of sophistication that may be applied. Kartinen and Martin (1995) stated that many of the existing processes are acceptable under the correct circumstances, but "the challenge is to determine which process goes with which set of circumstances". We should add that, sometimes, the removal technology is suitable, but its application is not possible due to the reluctance of people to accept the changes on the organoleptic properties of waters they have been drinking for years. Additionally, the volume, handling and final disposal of the generated wastes should be considered, but this aspect will be not treated here. Important references can be consulted (Hering et al., 1997; McNeill and Edwards, 1995; Meng et al., 2000; Sancha, 2003).

All technologies rely on a few basic chemical processes that can be applied alone, simultaneously or in sequence: oxidation/reduction, coagulation–filtration, precipitation, adsorption and ion exchange, solid/liquid separation, physical exclusion, membrane technologies, biological methods, etc. Most arsenic removal technologies are efficient when the element is in the pentavalent state, because it is present in the form of 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 arsenic remediation methods use, previously to other processes, an oxidation step. However, oxidation without help of other physical or chemical transformations does not remove arsenic from water. As it is obvious, boiling of water for purification does not remove arsenic and, on the contrary, this process increases As concentration by evaporation. This is a fact commonly ignored by the potentially affected people.

For previous reports on arsenic removal, see for example Feroze Ahmed, 2002; Newcombe and Möller, 2008; Pirnie, 2000. Comprehensive revisions, covering other complementary aspects, can be found in Ravenscroft et al. (2009) and in Sharma and Sohn (2009).

In what follows, conventional, well-established technologies for arsenic removal will be briefly mentioned. Emergent technologies will be addressed in more detail later, with emphasis on those methodologies that could be suitable for application in poor, isolated, decentralized rural and urban populations of LA, not connected to water network distribution.

2.1. Conventional technologies

The most common technologies include processes that can be used alone or in combination, such as oxidation, coprecipitation and adsorption onto coagulated flocs, lime treatment, adsorption onto suitable surfaces, use of ion exchange resins and membrane technologies (Newcombe and Möller, 2008; Ravenscroft et al., 2009; Sharma and Sohn, 2009). Most of them are confident and well understood technologies for arsenic removal in large and medium scale treatment plants for centralized services.

2.1.1. Oxidation and reduction

Oxidation is a previously required step to transform As(III) species in more easily removable As(V) species. Simple direct aeration is slow (Bissen and Frimmel, 2003), but a number of chemicals, including gaseous chlorine, hypochlorite, ozone, permanganate, hydrogen peroxide, manganese oxides and Fenton's reagent (H_2O_2/Fe^{2+}) can be employed to accelerate oxidation (see Feroze Ahmed, 2002; Pirnie, 2000). Chlorine is a rapid and effective oxidant, but it may react with organic matter, producing toxic and carcinogenic trihalomethanes as by-products. Potassium permanganate effectively oxidizes arsenite, and it may be a widely available inexpensive reagent suitable for developing countries. Hydrogen peroxide can be an effective oxidant if the raw water contains dissolved iron, which often occurs in conjunction with arsenic contamination, allowing the occurrence of Fenton reactions (see Section 2.2.6).

Ultraviolet radiation alone or with suitable light absorbers such as TiO_2 can be also convenient options for As(III) oxidation (see Section 2.2.6).

2.1.2. Precipitation

Methods taking advantage of the insolubility of certain arsenical inorganic compounds such as As(III) sulfide, calcium arsenate and ferric arsenate may be proposed to remove As from water. By adding calcium, magnesium, manganese (II) or iron (III) salts to As(V) solutions, As-containing solids are obtained that can be removed through sedimentation or filtration. However, the method is generally not suitable because of the instability of most of the solids, also inadequate for direct disposal, although it can be used to palliate the problem in mining sites where those salts are present naturally (Ladeira et al., 2002). The solubility of the different materials is very dependent on their nature, pH and other variables, and the aqueous solutions in equilibrium with the metal arsenates have extremely high arsenic concentrations, exceeding the guidelines for drinking waters and even for sewage effluents and wastes (Bothe and Brown, 1999; Magalhães, 2002; Ravenscroft et al., 2009; Vogels and Johnson, 1998).

2.1.3. Coagulation and filtration

The most common technology for arsenic removal is coagulation and filtration. Arsenic is removed in the pentavalent form, which adsorbs onto coagulated flocs and can be then removed by filtration. As(III) has to be previously oxidized, generally with chlorine (Kartinen and Martin, 1995). The most used coagulants are aluminum sulfate ($Al_2(SO_4)_3$), iron chloride ($FeCl_3$) and ferrous sulfate ($FeSO_4$), iron salts being generally better removal agents. $FeCl_3$ generates relatively large flocs, while smaller ones are formed with $FeSO_4$ (Edwards, 1994; Hering et al., 1997; Newcombe and Möller, 2008; Pirnie, 2000; Ravenscroft et al., 2009).

Filtration is a necessary step. Without filtration, arsenate removal is around 30%, but using a 0.1 or 1.0 μm filter, arsenate removal improves to more than 96% (Hering et al., 1996; Chwirka et al., 2000; Fields et al., 2000; Jekel and Seith, 2000; Madiec et al., 2000; Sancha, 1999, 2000).

The coagulation–filtration technology is simple, only common chemicals are used, installation costs are small and it can be easily applied to large water volumes. However, relatively large volumes of As-containing sludges are formed, typically disposed off in landfills, and being a potential source of contamination.

In Chile, some plants based on the coagulation technology were implemented in the 70's, constituting a great solution for the problem of arsenic for small and medium cities (Sancha and Ruiz, 1984; Sancha, 1999, 2000, 2003, 2006; Sancha and Fuentealba, 2009). The first plant, using direct filtration and $FeCl_3$ as coagulant was installed in 1970 in Antofagasta (El Salar del Carmen complex). Other As removal plants are those of Chuquicamata (Sancha, 2006) and Taltal (Sancha, 2006; Sancha and Fuentealba, 2009).

In Argentina, the Center of Sanitary Engineering of the National University of Rosario (Centro de Ingeniería Sanitaria de la

Universidad Nacional de Rosario) developed the ARCIS-UNR process, which uses a coagulation–adsorption method with poly-aluminum chloride (PAC) or ferric chloride, followed by a double filtration. The technology proved to achieve 80–90% of As removal (Ingallinella et al., 2003a,b; Ingallinella, 2006; Litter et al., 2008). Later, various small and medium plants have been successfully implemented in the country with this methodology.

2.1.4. Lime softening

In the presence of water and carbonic acid, lime forms calcium carbonate, and can be used to adsorb arsenic, the process ending with a coagulation step. The method is efficient to treat water with high hardness, especially at pH > 10.5. Addition of chlorine to oxidize As(III) is needed. The disadvantages are: 1) a very high pH in the resulting water (10–12), which implies a further acidification step; 2) a very high dose of coagulant is needed; 3) relatively low removal efficiencies (generally not less than 1 mg L^{-1}), secondary treatments being required (Fields et al., 2000; Kartinen and Martin, 1995; Newcombe and Möller, 2008; Pirnie, 2000; Ravenscroft et al., 2009).

2.1.5. Adsorption

Aluminum oxides (activated alumina), iron oxide/hydroxides, titanium dioxide, cerium oxide, or reduced metals can be used as adsorbents (Ravenscroft et al., 2009). Granular activated alumina ($Al_2O_3/Al(OH)_3$) is a commercially available porous oxide, success-fully applied at slightly acid pH (5–7), giving efficiencies higher than 95% for both As(V) and As(III) (Pirnie, 2000). The technology is very simple, does not require chemical addition and is useful at community or household levels. Granular iron hydroxide, GFH®, a synthetic akaganeite, proved to be a good material, able to retain As(V) and As(III) (Driehaus, 2002; Hering et al., 1997; Wang et al., 2000). Granular iron oxide (Bayoxide®, GFO) is another similar successful material, containing less than 70% of Fe_2O_3 (Severn Trent Services, 2007–2009). Commercial titanium dioxide (Bang et al., 2005b; Dow Chem, 2005), cerium oxide (Shimoto, 2007; Amimono, 2007) and manganese dioxide (Driehaus et al., 1995) proved to be also effective.

Microparticles with magnetic properties were developed to remove the adsorbent material after the treatment (Dahlke et al., 2003). In another design, iron hydroxide nanoparticles were intro-duced into a polymeric network of ionic exchange resin. The materials were tested with good results in arsenic-contaminated groundwater of a village bordering Bangladesh and India (Cumbal and SenGupta, 2009; DeMarco et al., 2003). Very cheap materials were developed using sand and quartz covered by metallic oxides and their use as emergent materials will be described in Section 2.2.4.

However, despite their simplicity, the adsorption methods usually fail in lowering arsenic concentration to acceptable levels, and are recommended to treat only water with low Fe/As content (Chaudhury et al., 2003; Driehaus et al., 1995). The alumina surface is saturated very rapidly at high As concentrations, and regenera-tion is necessary, usually with a caustic bath followed by an acid treatment (Kartinen and Martin, 1995). Efficiencies are higher with As(V) than with As(III).

2.1.6. Membrane processes

From the possible membrane processes, microfiltration (MF) or ultrafiltration (UF), which use low-pressure membranes (large nominal pore sizes, 10–30 psi) are not completely adequate because the arsenical species are very small and can traverse the membranes. In contrast, nanofiltration (NF) or reverse osmosis (RO), which use high-pressure membranes, 75–250 psi, or even higher (Clifford, 1999; Pirnie, 2000); iii) electric repulsion by membranes (Newcombe and Möller, 2008; Pirnie, 2000; Ravenscroft et al., 2009), have appropriate pore sizes. In RO, an external pressure is applied to reverse natural osmotic flow, and water flows from a more concentrated saline solution through the semipermeable membrane, which has a thin microporous surface that rejects impurities but allows water to pass through. The membrane rejects especially polyvalent ions, being suitable for arsenic oxyanions. The process is efficient over an extended pH range (3–11).

Because in NF and RO only a small amount of the raw water (10–15%) passes through the membrane, these processes are suitable for household or applications where only a small amount of treated water is required. For higher water volumes (e.g., municipal systems), multiple membrane units in series have to be used (Pirnie, 2000).

Operation and maintenance requirements for membranes are minimal: no chemicals are needed, and maintenance consists of only ensuring a reasonably constant pressure, and periodically cleaning of the membranes. The main disadvantages, especially for RO, are low water recovery rates (typically 10–20%), high electrical consumption, relatively high capital and operating costs (expensive membranes), and the risk of membrane fouling. High concentra-tions of suspended solids, organic matter, humic acids, hardness, sulfides, ammonium, nitrite, methane, etc. interfere. Only low levels of arsenic can be treated. Discharge of rejected water (20–25% of the influent) or brine is also a concern; therefore, the technology is not useful in areas where water is scarce. The method provides good As(V) but poor As(III) removal, and oxidation is difficult because residual oxidants can damage the membranes (EPA, 2006). In addition, RO eliminates not only arsenic but also ions, altering the chemical composition or organoleptic properties of drinking waters.

Several RO plants have been installed recently in Argentina, e.g. in the provinces of Santa Fe, Córdoba and La Pampa (D'Ambrosio, 2005).

In electrodialysis (ED), ions are transported from a lesser to a higher concentrated solution through ion permeable membranes under the influence of a direct electric current. The efficiency of the technique is similar to that of RO, mainly in treating water with high total dissolved solids (TDS). Electrodialysis with reversion of polarity of the electrodes (EDR) is an improvement of ED with minimization of scaling (Pirnie, 2000; Ravenscroft et al., 2009).

Garrido et al. (2008), developed capacitive deionization (CI), an advanced electrochemical method based on the deionization by flow through a capacitor like system with electrostatic load, configured as a low-cost filter of coal electrodes. This technology is recommended for water containing less than 3000 mg L^{-1} of total dissolved solids. The advantages of capacitive deionization over RO, NF and electrolysis are: 1) smaller amount of chemical reagents for the cleaning of cells or membranes; 2) both As(V) and As (III) can be removed; 3) the volume of rejected water is low (between 3 and 7% of the treated volume); 4) low operative and maintenance costs.

2.1.7. Ion exchange resins

Synthetic ionic exchange resins, generally of polymeric matrix (polystyrene cross-linked with divinylbenzene), linked to charged functional groups, can be applied for As removal; quaternary amine groups, $-N^+(CH_3)_3$, are the preferred groups. Arsenate removal is efficient, producing effluents with less than 1 µg L^{-1} of arsenic, while arsenite, being uncharged, is not removed, and a previous oxidation step is necessary (Pirnie, 2000; Ravenscroft et al., 2009).

Commonly, resins are pretreated with hydrochloric acid, to establish chloride ions at the surface, which are easily displaced by arsenic. Arsenate removal is relatively independent of pH and influent concentration. $HAsO_4^{2-}$ has adsorption ability higher than that of $H_2AsO_4^-$. Competing anions, especially sulfate, TDS, selenium, fluoride, and nitrate, interfere strongly and can affect run length.

Suspended solids, SS, and precipitated iron can cause clogging (Kartinen and Martin, 1995; Pirnie, 2000; Wang et al., 2000).

The most important producers of ion exchanger materials (Purolite, Bayer, Dow Chem and Rohm and Haas) have introduced new tailored anionic exchangers to attain values below 10 μg L^{-1}.

2.1.8. Comparison of conventional technologies for As removal

In Table 1 are compared the conventional technologies with their advantages and disadvantages.

2.2. Emergent technologies

In the last decades, a large amount of scientific and technological work has been devoted to develop new technologies for arsenic remediation that seek to minimize costs of investment, operation and maintenance (i.e., low-cost technologies) and technological development (i.e., low-tech systems). These technologies focus more on small scale or household treatments for isolated populations. However, the social acceptance, waste production and treatment and the corresponding required handling needs to be assessed before considering the implementation of each remediation option.

Some of these technologies are merely adaptation of conventional methods like coagulation–filtration, or adsorption, using very cheap materials (iron-coated sand, bricks, iron filings, activated alumina or carbon) to be employed at household or community scale (Kemper and Minnatullah, 2005; Ravenscroft et al., 2009).

2.2.1. In-situ remediation

In-situ based technologies have lower operation costs in comparison with on-site or off-site treatment as the classical "pump and treat" technologies. Different approaches have been applied by bioremediation, permeable reactive barriers, air dispersion, chemical oxidation, multiphase extraction, supervised natural attenuation, etc. Biological methods will be reviewed in Section 2.2.5.

The use of permeable reactive barriers (PRB) and reactive zones has been postulated as one of the most efficient technologies for in-situ removal of pollutants, particularly for As from groundwater. Fe (or Al) oxide-containing materials can be used as relatively cheap passive reactive barriers (Bhattacharya et al., 2002; Gavaskar et al., 1998; Gu et al., 1999; Lindberg et al., 1997).

In PRB technology, a reactive medium is interposed in the way of the contaminant plume as shown in Fig. 1. The appropriate reactive material is able to induce physicochemical and/or biological processes to remediate groundwater contamination. PRBs are particularly attractive for decontamination of groundwater because they are less expensive than conventional technologies and no costly equipments for operation are needed.

The involved main processes are sorption, precipitation, chemical reaction and/or biogenic reactions (Diels et al., 2003). For arsenic, PRBs should be built by materials that enable adsorption and/or coprecipitation of the anionic species, such as mixtures of iron oxides with silica and calcite (Lackovic et al., 2000). Important drawbacks of the technology are: 1) high impact of long-term microbiological and geochemical processes on the durability of the barrier, 2) degradation of the material by corrosion and 3) decrease of permeability by precipitation of sulfides, oxides, hydroxides and carbonates.

The technology has been directed recently to the use of zerovalent iron (ZVI), as a new sorption medium to remove both arsenate and arsenite; values always below 10 μg L^{-1} are achieved (Gibert et al., 2003a,b; Su and Puls, 2001, 2003). In form of columns, it can be applied directly for household applications (see Section 2.2.3).

2.2.2. Combined coagulation/flocculation and adsorption methods

Different technologies for single households were developed or adapted by scaling down and simplifying conventional methods

Table 1
Advantages and disadvantages of conventional technologies for arsenic removal.

| Technologies	Advantages	Disadvantages
Oxidation and reduction	Simple. Small installation costs. Easily applied to large water volumes. Arsenite can be directly oxidized by a number of chemicals and/or UV light.	Some oxidants produce toxic and carcinogenic by-products. Needs further removal treatment.
Precipitation	Solid obtained can be removed through sedimentation and filtration.	Solids rather unstable and inadequate for direct disposal as they will produce As-containing liquid residues.
Coagulation/filtration	Simple. Easily applied to large water volumes. Effective when As(V) is the only pollutant. Low capital and operative costs. Alum allows F removal.	Low removal efficiency. pH needs adjustment. Disposal of the arsenic-contaminated coagulation sludge may be a concern. Low removal efficiency. Filtration needed. As(III) must be previously oxidized.
Lime softening	pH > 10.5 provides efficient As removal. Efficient to treat water with high hardness.	Low efficiency. High coagulant dose. High pH in the effluent. May require secondary treatment.
Adsorption (activated alumina, iron oxides/hydroxides, TiO$_2$, cerium oxide, metals)	Simple. Not other chemicals required. Highly selective towards As(V). Effective with water with high TDS. Useful at community or household levels.	Moderate efficiency. Regeneration needed. Interferences: Se, F$^-$, Cl$^-$ and SO$_4^{2-}$. Application of point-of-use treatment devices needs regeneration and replacement.
Reverse osmosis and nanofiltration	Useful at community or household levels. Minimal membrane operation and maintenance. Highly effective towards As, effective in treating water with high TDS	Only low As levels can be treated. Poor As(III) removal. For high water volumes, multiple membrane units required. Low water recovery rates (10–20%). High electrical consumption. High capital and operation costs. Membrane fouling. Many interferences. 20–25% water rejection. Other ions can be removed.
Electrodialysis, electrodialysis with reversion of polarity of the electrodes	Efficiency similar to reverse osmosis, effective in treating water with high TDS. Minimize scaling by periodically reversing the flows of dilute and concentrate and polarity of the electrodes.	Very high costs.
Ion exchange	Effective removal. Not pH and influent concentration dependent.	As (III) is not removed. Sulfate, TDS, Se, F$^-$ and NO$_3^-$ interfere. SS and precipitated iron cause clogging. May require pretreatment.
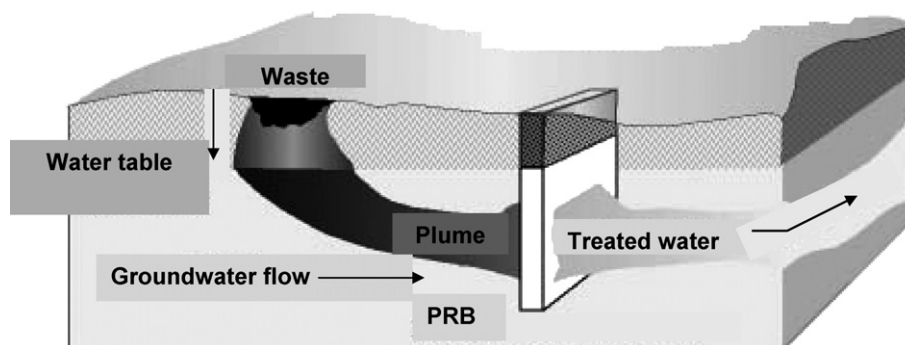
Fig. 1. Conceptual scheme of PRBs.

used in water treatment plants, which use the oxidation, adsorption and coagulation sequence for As removal from drinking water (Sastre et al., 1997; Castro de Esparza and Wong de Medina, 1998).

A household scale low-cost As removal methodology was developed in Peru (Castro de Esparza et al., 2005) using ALUFLOC, a 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 and allowed to remove up to 98% of the dissolved As (As concentration in raw water: 1 mg L^{-1}); at higher As concentrations, the removal efficiency decreased.

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 μg L^{-1} As) from different wells of the Tucumán province (Argentina) were tested, attaining final As concentrations below 10 μg L^{-1} in all cases.
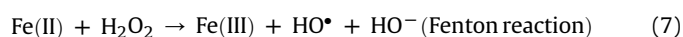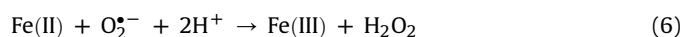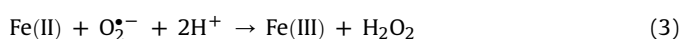
2.2.3. Zerovalent iron

Zerovalent iron is an emergent material increasingly used for the treatment of several pollutants, particularly toxic metals. As said before, ZVI is one of the main components of PRBs. In last times, arsenic removal with ZVI has been object of different studies (Kanel et al., 2005; Leupin et al., 2005; Leupin and Hug, 2005; Manning et al., 2002; Su and Puls, 2001).

The method is useful for both As(V) and As(III). In the case of As(III), removal by ZVI takes place mainly by adsorption and coprecipitation onto iron hydroxides or oxides formed during ZVI oxidation (Bang et al., 2005a; Kanel et al., 2005; Lackovic et al., 2000). The mechanism involves preliminary Fe(0) corrosion: ZVI oxidation in the presence of water and oxygen produces Fe(II) and then different Fe(II)/(III) oxides or hydroxides such as iron rusts, magnetite, lepidocrocite, maghemite, ferrous hydroxide ferric hydroxide, etc., depending on redox conditions and pH:

$$2Fe(0) + 2H_2O + O_2 \rightarrow 2Fe(II) + 4OH^- \qquad (1)$$
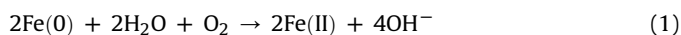
Once formed, Fe(II) reacts with dissolved O_2 generating Reactive Oxygen Species (ROS) like $HO^\bullet$, $O_2^{\bullet-}/HO_2^\bullet$, H_2O_2, and it is oxidized to Fe(III), according to the following simplified equations at neutral pH (Hug and Leupin, 2003; Joo et al., 2004; Leupin and Hug, 2005; Morgada et al., 2009; Voegelin and Hug, 2003).

$$Fe(II) + O_2 \rightarrow Fe(III) + O_2^{\bullet-} \qquad (2)$$

$$Fe(II) + O_2^{\bullet-} + 2H^+ \rightarrow Fe(III) + H_2O_2 \qquad (3)$$

$$2O_2^{\bullet-} + 2H^+ \rightarrow H_2O_2 + O_2 \qquad (4)$$

$$H_2O_2 + O_2^{\bullet-} \rightarrow HO^\bullet + O_2 + HO^- \qquad (5)$$

$$Fe(II) + O_2^{\bullet-} + 2H^+ \rightarrow Fe(III) + H_2O_2 \qquad (6)$$

$$Fe(II) + H_2O_2 \rightarrow Fe(III) + HO^\bullet + HO^- \, (Fenton\,reaction) \qquad (7)$$

Species of higher oxidation state, like Fe(IV), were also proposed to be formed, especially at neutral pH.

Recently, Ramos et al. (2009) studied the mechanism of As immobilization on nanoparticulate ZVI (NZVI) using High Resolution X Ray Photoelectronic Spectroscopy (HR-XPS) and informed clear evidence of As(0) formation together with As(III) and As(V) on the nanoparticle surface after reaction with As(III) or As(V) in solution. These results proved that both reductive and oxidative mechanisms take place during NZVI treatment. The dual function exhibited by NZVI is possible by the core-shell structure of NZVI, which contains a highly reducing metal core and a thin layer of amorphous iron oxyhydroxide, promoting coordination and oxidation of As(III).

According to Leupin and Hug (2005), the application of metallic iron to remove arsenic from waters is promising for several reasons:

i)   metallic iron is widely available and iron filings can be produced locally even in poor localities at low-cost,
ii) iron corrosion converts iron to strongly sorbing iron oxides,
iii) the reactive intermediates formed during corrosion of metallic iron in aerated waters oxidize As(III) to As(V),
iv) the combination of As(III) oxidation and sorption of As(V) makes unnecessary the use of oxidizing chemicals.

There are already results of laboratory experiments with zerovalent iron, which use cartridges filled with sand, local iron materials, iron wool and packing wire, as well as iron nanoparticles (NZVI) (Bundschuh et al., 2009; Leupin et al., 2005; Litter, 2006a,b; Litter et al., 2008; Litter and Jiménez González, 2004; Litter and Mansilla, 2003; Morgada et al., 2008, 2009; Morgada de Boggio et al., 2009, in press; Su and Puls, 2001). A recent work was performed recently in Argentina with commercial nanoparticles produced by a local industry. A rapid As(V) removal using very low iron concentrations (0.005–0.1 g L^{-1}) took place, attaining more than 90% after 150 min of contact time at the optimal NZVI concentration. The commercial NZVI presented an outstanding ability to remove As from waters from the Chaco-Pampean plain, due not only to a high surface area and low particle size but also to a high intrinsic activity. Irradiation strongly improved As removal in natural waters, and this aspect will be described in Section 2.2.6.

A method using electrochemical corrosion of a fixed bed of metallic iron was developed and applied in San Juan, Argentina (Cáceres, 2007; Litter et al., 2008). 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. Removals above 90% were obtained.

2.2.4. Geological materials as natural adsorbents and other low-cost materials

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 hematite (α-Fe_2O_3), goethite (α-$FeO(OH)$), gibbsite (γ-$Al(OH)_3$) and soils or sediments including these minerals (e.g., oxisols, laterite), indigenous limestone (Soyatal), iron-coated zeolites, clay minerals (montmorillonite, bentonite) were tested either with laboratory-prepared or natural waters, and identified as alternative adsorbents for small water volumes (Alvarez-Silva et al., 2009; Armienta et al., 2009; Bhattacharya et al., 2002; Castro de Esparza and Wong de Medina, 1998; Claesson and Fagerberg, 2003; Deschamps et al., 2003, 2005; Litter, 2006a,b; Litter and Mansilla, 2003; Litter and Jiménez González, 2004; Ladeira and Ciminelli, 2004; Mellano and Ramirez, 2004; Muñoz et al., 2005; Rivera and Piña, 2000; Simeonova, 2000; Storniolo et al., 2005; US Environmental Protection Agency, 2002; Weerasooriya et al., 2003). Materials based on iron/manganese oxides, like "greensand" and other various natural minerals, have been also investigated (Deschamps et al., 2005; Mohan and Pittman, 2007; Newcombe and Möller, 2008; Pirnie, 2000; Prasad, 1994; Shevade and Ford, 2004; Zeng, 2003).

Simple sand filters can be a very feasible option for As removal from groundwater exhibiting iron concentrations up to around 400 µg L^{-1}. The method has been proved efficient in Vietnam at household level (Luzi et al., 2004).

Interesting materials to remove As at small scale were iron oxide coated sand (IOCS) (Bhattacharya et al., 2002; Chen et al., 2004; Deschamps et al., 2005 Joshi and Chaudhury, 1996; Thirunavukkarasu et al., 2004), manganese dioxide coated sand (Bajpai and Chaudhury, 1999) and limestone particles covered by iron oxide (Banavali et al., 2008).

2.2.5. Biological methods

Relatively little is known about the use of biological removal of arsenic from water, although these methods show a great potential due to its environmental compatibility and possible cost-effectiveness. Microbial activity can remove, mobilize, and contain arsenic through sorption, biomethylation–demethylation, complexation, coprecipitation, and oxidation-reduction processes. Ex-situ bioleaching can effectively remove arsenic 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 (Katsoyiannis and Zouboulis, 2004a,b; Teixeira and Ciminelli, 2005; Wang and Zhao, 2009). Recent reviews are available (Lasat, 2002; Cherian and Oliveira, 2005; Dickinson et al., 2009).

In LA, different types of low-cost natural biological materials such as cellulose, milled bones, sedges, sorghum biomass, waste biomass, and others have been tested for their suitability to remove As from water. Muñiz et al. (2009) developed filters for As removal using a combination of cellulose and activated carbon from lignite as adsorbents. Another interesting approach was that of Teixeira and Ciminelli (2005), who tested activated waste biomass with high fibrous protein content (rich in keratin) obtained from chicken feathers. The method allowed selective adsorption of As(III) at low pH with sorption rates up to 270 µmol As(III) g^{-1} of biomass. Natural biogenic hydroxyapatite (HAPb) obtained from cow-charred bones resulted a good sorbent for As(V) in water under the conditions explored by the authors (1000 µg L^{-1}, 5 g L^{-1} adsorbent, circumneutral pH, 24 h contact time, Czerniczyniec et al., 2007).

Phytofiltration, i.e., the use of plants to remove contaminants from water, is another emerging technology. Huang et al. (2004) investigated the potential of two hydroponically cultivated arsenic hyperaccumulating ferns (*Pteris vittata* and *Pteris cretica* cv. Mayii) to remove As (20–500 µg L^{-1}). As examples, *P. vittata* reduced As concentration from 200 to 2.8 µg L^{-1}, and from 20 µg L^{-1} to 0.4 µg L^{-1} in 24 h. Interestingly, the authors claim that the phytofiltration technique may provide the basis for a solar-powered hydroponic technique to treat arsenic-contaminated drinking water at small scale.

Alvarado et al. (2008) studied As removal (0.15 mg L^{-1}) by Water Hyacinth (*Eichhornia crassipes*) and Lesser Duckweed (*Lemna minor*). No significant differences were found between both species on bioaccumulation capability. The removal rate for Water Hyacinth was higher than that of Lesser Duckweed, this species representing a reliable alternative for arsenic bioremediation in waters.

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

Hansen et al. (2004) studied As(V) biosorption using dried algae (*Lessonia nigrescens*) collected in Valparaiso bay, Chile. The experiments were performed using laboratory solutions (200 mg As(V) L^{-1}, pH 2.5, 4.5 and 6.5). *Lessonia nigrescens* showed very good adsorption capacities and its use may be interesting for small-scale drinking water treatment, deserving further investigation.

Bundschuh et al. (2007) investigated the effectiveness and suitability of dried macro-algae (*Spyrogira* spp.) to remove As from acid mine drainage (AMD) and other waters from the Poopó lake basin (Bolivia, Andean highlands) finding higher efficiency than that of typical plants like *totora* (*Schoenoplectus californicus*) and *paja brava* (*Festuca orthophylla*): 80–90% of As removal was attained within 4 days.

2.2.6. Photochemical technologies

Very cheap technologies based on the use of solar light, abundant in many regions where, at the same time, the problem of arsenic is dramatic, can be adapted for arsenic removal. The use of solar or artificial light and dissolved iron has been object of several studies in the last decade, especially to facilitate oxidation of As(III) to As(V). Therefore, to effectively remove arsenic, a two-step process has to be designed: one for As(III) oxidation and the second one for elimination of the produced As(V). These two steps can be simultaneous or consecutive. Another possibility is the photochemical reduction of As(V) or As(III) to elemental As, a non-mobile, relatively stable As form, which can be, in this way, removed from the aqueous phase.
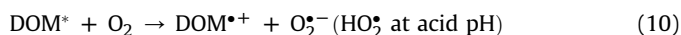
2.2.6.1. As oxidation by light and chemical reagents. As said in Section 2.1.1, As(III) oxidation by atmospheric oxygen is thermodynamically possible but rather slow. The process can be accelerated under UV light irradiation, as described by Bissen et al. (2001): although no oxidation of As(III) (1 mg L^{-1}) was observed after one week in dark oxygenated aqueous solutions, irradiation with a solar simulator produced 54% of As(III) oxidation in 45 min. The authors attribute the acceleration to i) emission of the lamp at wavelengths under 280 nm, where As(III) presents some absorption, ii) the presence of contaminants at trace levels on the walls of the reactor, iii) a very weak absorbance of As(III) ($\epsilon < 5$ L mol^{-1}) at wavelengths over 289 nm; or iv) increase of the temperature. However, the use of direct UV illumination to promote As(III) oxidation has low efficiency to guarantee its application in As removal.

Addition of H_2O_2 to the UV system triggers As(III) oxidation. As already said in Section 2.1.1, H_2O_2 is a good oxidant for As(III) in the dark, but a large excess of peroxide is needed to achieve complete oxidation. In contrast, Yang et al. (1999) found that UV light promoted As(III) oxidation in the presence of H_2O_2. Complete oxidation of 525 μM As(III) (air-saturated solution at pH 9) took place in less than 10 min with low H_2O_2/As(III) molar ratios (from 2:1 to 1:4). The authors attribute the enhancement of As(III) oxidation to the occurrence of H_2O_2 photolysis, which generates powerful oxidants, the HO• radicals:

$$H_2O_2 + h\nu \rightarrow HO^\bullet \tag{8}$$
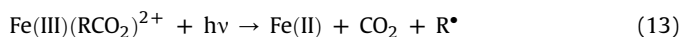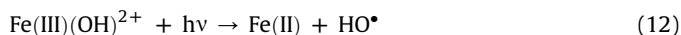
However, it is known that reaction (1) is effective only under irradiation with wavelengths lower than 300 nm, where H_2O_2 absorption is important; as a black lamp seems to be used in the reported experiments, it is probable that some impurity, causing Fenton-type reactions (Fe or Cu ions) is the responsible for the improvement of As removal (see Section 2.2.6).

If dissolved organic matter (DOM) is present in natural waters, H_2O_2 can be photochemically produced in-situ by the effect of solar light (Kocar and Inskeep, 2003):

$$DOM + h\nu \rightarrow DOM^* \tag{9}$$

$$DOM^* + O_2 \rightarrow DOM^{\bullet+} + O_2^{\bullet-} (HO_2^\bullet \text{ at acid pH}) \tag{10}$$

$$2HO_2^\bullet \rightarrow H_2O_2 + O_2 \tag{11}$$

Accordingly, Buschmann et al. (2005), using humic acid as representative of DOM, observed that the photooxidation rate of As(III) in air in the presence of UV light (366 nm) increased linearly with DOM concentration. This effect was much higher in the presence of Fe(III), due to a charge-transfer photochemical reaction forming HO• or R• from iron(III) hydroxo- or organocomplexes (Hug et al., 2001; Legrini et al., 1993; Litter, 2005; Zafiriou et al., 1984):
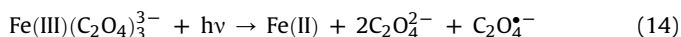
$$Fe(III)(OH)^{2+} + h\nu \rightarrow Fe(II) + HO^\bullet \tag{12}$$

$$Fe(III)(RCO_2)^{2+} + h\nu \rightarrow Fe(II) + CO_2 + R^\bullet \tag{13}$$

Once H_2O_2 and Fe(II) are formed, the known Fenton reaction(7) takes place, generating more HO•, and promoting a rapid As(III) oxidation (Litter, 2005).

Khoe et al. (1997) and Zaw and Emett (2002) used iron salts and UV or solar light for oxidation of As(III) present in AMD liquid residues. Iron acts in these processes as both oxidant and coagulant; stable solids are formed that can be safely disposed. In 1996, a demonstration was carried out in an abandoned mine in Montana (USA) using two types of photoreactors: (a) plastic containers around 85 L of capacity for a solar process and (b) a reactor with 24 low-pressure mercury lamps (65 W). Arsenic oxidation was completed in around 1–6 h under solar light (MSE Final Report, 1997). The process was later adapted for a field demonstration in Bangladesh (BGS Main report, 1999).

Kocar and Inskeep (2003) evaluated the use of ferrioxalate under UV radiation for As(III) oxidation in an ample pH range (3–7). Photolysis of iron oxalate leads to $CO_2^{\bullet-}$ that, under aerobic conditions, reacts very quickly with O_2, yielding superoxide radicals:

$$Fe(III)(C_2O_4)_3^{3-} + h\nu \rightarrow Fe(II) + 2C_2O_4^{2-} + C_2O_4^{\bullet-} \tag{14}$$

$$C_2O_4^{2-} \rightarrow CO_2^{\bullet-} + CO_2 \tag{15}$$

$$CO_2^{\bullet-} + O_2 \rightarrow O_2^{\bullet-} + CO_2 \tag{16}$$

Then, H_2O_2 and HO• are formed, according to the above equations (11)–(13) and (7).

The Solar Oxidation and Removal of Arsenic method (SORAS) is a very simple process, used with partial success in poor, isolated populations in Bangladesh and India (Wegelin et al., 1994, 2000). In this method, contaminated water is put in transparent PET bottles of soft drinks or mineral water together with some drops of lemon juice (citric acid), and irradiated under sunlight for several hours. If there is enough iron in the waters, Fe(III)-citrate complexes are formed, and photo-Fenton processes take place, with the generation of oxidizing species. Then, As(III) is transformed to As(V) together with precipitation of iron(III) oxides/hydroxides, and As(V) adsorption and coprecipitation (Fig. 2, Hug et al., 1997; Hug and Leupin, 2003). During the night, bottles are put in a vertical position to promote settlement of the flocs and clear water is obtained by further decantation or filtration. Tests of this technology have been performed in Nicaragua, Bangladesh, Peru, Chile and Argentina (Cornejo et al., 2008; Emett and Khoe, 2001; García et al., 2004; Hug, 2000; Hug et al., 2001; Hug and Leupin, 2003; Lara et al., 2006; Litter, 2002, 2006; Litter and Jiménez González, 2004; Litter and Mansilla, 2003).

However, groundwaters of several regions of LA do not have enough iron to make efficient the SORAS technology. To increase As removal, iron has to be added externally, in form of some natural Fe-containing minerals, iron wool, packing wire or zerovalent iron nanoparticles (Bundschuh et al., 2009; Litter et al., 2008; Morgada et al., 2008, 2009; Morgada de Boggio et al., 2009, in press). Commercial zerovalent iron nanoparticles exhibited outstanding properties to treat As-polluted groundwater of the Chaco-Pampean plain of Argentina (Tucumán province): As (around 200 μg L^{-1}) in contact with these NZVI particles (0.025 g L^{-1}) was driven to levels in agreement with the regulations (<10 μg L^{-1}) after 3 h UV irradiation (Morgada et al., 2009; Morgada de Boggio et al., in press).

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, for water detoxification (Bahnemann et al., 1994; Hoffmann et al., 1995; Legrini et al., 1993; Linsebigler et al., 1995; Litter, 1999, 2009; Mills and Le Hunte, 1997; Rajeshwar, 1995; Serpone et al., 1988). In HP, after excitation of TiO_2 with UV light (energy equal to or higher than the TiO_2 bandgap, E_g) conduction band electrons (e_{cb}^-) and valence band holes (h_{vb}^+) are created, which can recombine or migrate to the particle surface where they react with donor (D) or acceptor (A) species (Fig. 3). 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. For Degussa P-25, the most popular
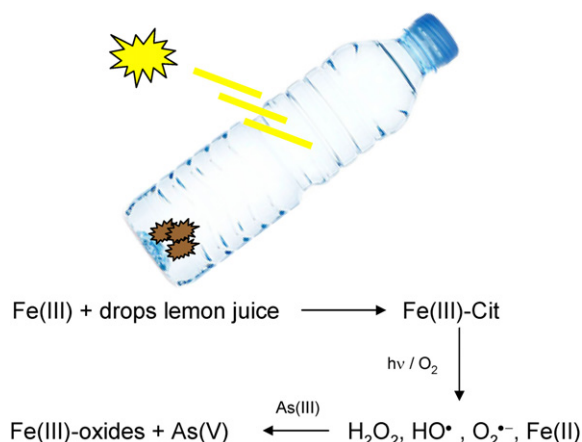
Fig. 2. Scheme of the SORAS process.

Fig. 3. Simplified diagram of heterogeneous photocatalysis with TiO_2 particles.

commercial form of photocatalytic TiO_2, these values have been calculated as -0.3 and $+2.9$ V vs. NHE at pH 0, respectively (Martin et al., 2004); therefore, h_{vb}^+ are strong oxidants that may attack D directly or form hydroxyl radicals (HO$^\bullet$) from water or surface hydroxide ions, while e_{cb}^- are mild reducing acceptors. It has been established that HP processes under regular illumination take place only through monoelectronic steps.

The above scheme can be described by the following simplified equations:

$$TiO_2 + h\nu \rightarrow e_{cb}^- + h_{vb}^+ \tag{17}$$

$$e_{cb}^- + A \rightarrow A^{\bullet -} \tag{18}$$

$$h_{vb}^+ + H_2O \rightarrow HO^\bullet + H^+ \tag{19}$$

$$h_{vb}^+ + D \rightarrow D^{\bullet +} \tag{20}$$

In particular, O_2 can play the role of acceptor and it is reduced by e_{cb}^- to $O_2^{\bullet-}$, from which other ROS (HO$_2^\bullet$, H_2O_2 and finally HO$^\bullet$) are formed, this being an alternative route of HO$^\bullet$ generation.

Application of HP to the case of As(V) implies complex mechanisms. Oxidation of As(III) can take place by h_{vb}^+ or HO$^\bullet$, going first to As(IV) and then to As(V):

$$As(III) + h_{vb}^+/HO^\bullet \, (or \, other \, ROS) \rightarrow As(IV) \rightarrow As(V) \tag{21}$$
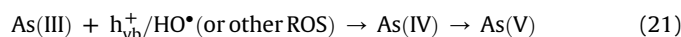
Several examples of photocatalytic oxidation of As(III) by UV/TiO_2 at laboratory level are reported in the literature (Bissen et al., 2001; Dutta et al., 2005; Ferguson et al., 2005; Jayaweera et al., 2003; Lee and Choi, 2002; Leng et al., 2007; Ryu and Choi, 2004; Xu et al., 2005; Yang et al., 1999; Yoon et al., 2009). In all cases, As(III) oxidation was very rapid, taking place in time scales of 10–100 min at various concentrations (micromolar to millimolar range). Methylated arsenical compounds like monomethylarsonic acid (MMA) and dimethylarsinic acid (DMA) were easily degraded by UV-TiO_2. Arsenate is the final product for both MMA and DMA oxidation, with the consequent mineralization of the organic moiety of the arsenical initial species (Xu et al., 2007).

A very interesting example is the use of an efficient, cost-effective and environment-friendly adsorbent formed by a mixture of TiO_2 and slag-iron oxide obtained from a municipal incinerator of solid wastes for photocatalytic oxidation of arsenite and simultaneous removal of the generated arsenate; the process was performed in a relatively short contact and illumination time (Zhang and Itoh, 2006). 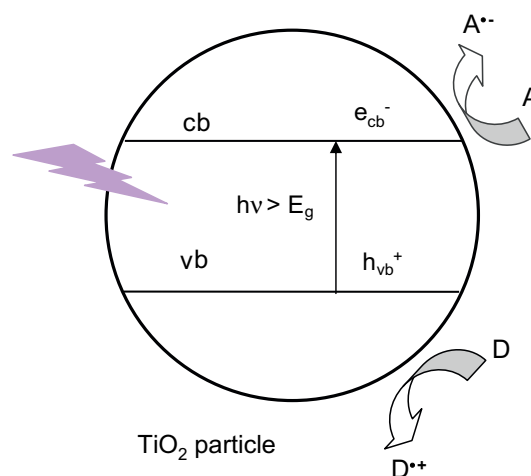Another example is the design of a fixed-bed, flow-through reactor for As(III) oxidation having TiO_2 immobilized on glass beads (Ferguson and Hering, 2006). Nakajima et al. (2005) proposed the combined use of TiO_2 as photocatalyst and activated alumina as adsorbent for the removal of inorganic As(III), MMA and DMA from aqueous media using Xe lamps or solar light.

A low-cost HP procedure was proposed to remove As in well water samples of the Chaco-Pampean plain (Las Hermanas, Santiago del Estero province, Argentina). Walls of PET plastic bottles were internally impregnated with TiO_2 by a very simple technique (Meichtry et al., 2007). When As-contaminated water samples ($500-1800$ μg L^{-1}), poured into these bottles, were exposed to artificial UV light in the presence of Fe(III) salts, more than 94% As removal took place (Bundschuh et al., 2009; Litter, 2006a,b; Litter et al., 2008; Mateu, 2007; Morgada et al., 2008; Morgada de Boggio et al., 2009, in press). Fostier et al. (2008) obtained similar results with laboratory samples under solar irradiation.

According to the redox potential of TiO_2 e_{cb}^-, direct photocatalytic reduction of As(V) or As(III) is not thermodynamically possible, as proved by Yang et al. (1999) and confirmed by experiments with the stopped-flow technique using irradiated TiO_2 nanoparticles (Litter et al., unpublished results). Nevertheless, TiO_2 heterogeneous photocatalytic reduction of As(III)/(V) is indeed possible at acid pH by an indirect route in the presence of organic donors. These donors are oxidized by h_{vb}^+ or HO$^\bullet$ in irreversible reactions, and produce strongly reducing radicals R$^\bullet$, which in turn reduce arsenical species to elemental As:

$$HR + h_{vb}^+/HO^\bullet \rightarrow R^\bullet \tag{22}$$

$$R^\bullet + As(V)/(III) \rightarrow As(0) \tag{23}$$

The most common donors are methanol, ethanol or 2-propanol, which lead to 1-hydroxyalkyl radicals, while carboxylic acids generate strong reducing radicals such as $CO_2^{\bullet-}$ in the cases of formic or oxalic acids. This can be, then, a method to immobilize dissolved As in the form of the solid As(0) form.

Combining photocatalytic oxidative and reductive pathways, Yang et al. (1999) proposed a two-step pollution abatement scheme: 1) initial oxidation of As(III) to As(V) either via UV/TiO_2 or UV/H_2O_2, followed by 2) immobilization of As(V) as the elemental species on TiO_2 in a second reductive stage, after adjustment of the effluent to acid pH.

Other light-driven oxidative technologies have been tested. A UV light assisted process, which worked at neutral or alkaline pH,

Table 2
Advantages and disadvantages of emergent technologies for arsenic removal.

Technologies	Advantages	Disadvantages
In-situ remediation (PRBs)	Low operational costs. Low-cost local materials can be used.	High impact of microbiological and geochemical processes at long term. Corrosion of materials. Permeability diminished by precipitation of sulfides, oxides, hydroxides and carbonates.
Zerovalent iron	Widely available local iron materials at low-cost. As(III) and As(V) can be treated.	Produces toxic solid wastes.
Zerovalent iron nanoparticles	Higher contact surface results in a lower amount of iron. As(III) and As(V) can be treated.	Complicate synthesis of material.
Geological materials as natural adsorbents	Feasible process in developing countries.	Possible growth of microorganisms. Becomes clogged, if excessive iron.
Biological methods: bioadsorption, ex-situ bioleaching, phytofiltration, phytoremediation	Environmental compatibility and possible cost-effectiveness.	Much research still needed.
Photochemical oxidative technologies: Fe salts/solar light, SORAS, TiO_2 Heterogeneous Photocatalysis, ZVI, NZVI	Friendly and non-expensive technologies for poor and isolated populations. Based on the use of solar light and low-cost materials. Simultaneous oxidation of As and removal of natural organic pollutants, toxic metals and microbiological contamination can be achieved in most of the cases.	External addition of iron to the waters before or after treatment is needed in LA.
Reductive TiO_2 Heterogeneous Photocatalysis	Provides immobilized As(0) on TiO_2.	Addition of organic donors and acid pH is required. Much research is still needed.

proposed the use of sulfite or another oxidizable sulfur form as photoabsorber (Khoe et al., 1998; Zaw and Emett, 2002). Photochemical As(III) oxidation by combined vacuum-UV and short UV irradiation (185 and 254 nm) was recently investigated (Yoon et al., 2008). The method proved to be better than other photooxidation procedures (e.g., UV-C/H_2O_2, UV-A/Fe(III)/H_2O_2 and UV-A/TiO_2), transforming 100 mM As(III) almost completely in 10 min. The reaction occurs through $HO^\bullet$ produced by 185 nm photosplitting of water ($H_2O \rightarrow H^\bullet + HO^\bullet$), and it is enhanced by the presence of Fe(III) and H_2O_2. The method was also effective to oxidize As(III) in a real natural water. Adsorption on activated alumina or coagulation/precipitation with $FeCl_3$ was used to remove the produced As(V).

Neppolian et al. (2008) described a very rapid photochemical As(III) oxidation using low amounts of potassium peroxydisulfate ($K_2S_2O_8$). The oxidant was proposed to be the sulfate radical ($SO_4^{\bullet-}$).

Recently, Yeo and Choi (2009) studied the reaction of arsenite under 254 nm irradiation mediated by iodide, finding quantitative oxidation to As(V). The authors suggest that the process could be applied in treatment plants for acid waters polluted with As in the range 1–1000 μg L^{-1} and higher, employing directly the already existing germicide lamps in these plants.

2.2.7. Comparison of emergent technologies

In Table 2 are listed the main advantages and disadvantages of emergent technologies for arsenic removal cited in this work.

3. Conclusions

Arsenic occurs in natural waters predominantly in inorganic forms, such as arsenite (As(III)) and arsenate (As(V)). The presence of arsenic in water for human consumption causes the appearance of HACRE, an endemic illness, affecting a large number of people in LA. As(V) is more effectively removed from source waters than As(III) by iron coagulants, by precipitation of natural iron, and by adsorptive media. To remove As(III), a previous oxidation to As(V) is needed. Conventional technologies (coagulation–coprecipitation, adsorption, reverse osmosis, etc.) can be applied at medium or large scale. However, for domestic or community purposes as those frequently found in rural or urban isolated population in LA, low-

cost technologies using non-expensive materials, sunlight or biological methods should be developed. However, further research is needed to find additional methods and to assess the effectiveness of the proposed technologies. In each case, the validation of the method with real waters of the site before application is mandatory.

Arsenic removal from water for human consumption seems to be a very difficult task. Not a universal method exists and the election is very dependent on the composition of waters to be treated. Socioeconomic features should be carefully taken into account to select the technology.

The problem of As in LA is of the same order of magnitude as in other world regions, such as SE Asia, but it is often not described in English. Although various studies have been undertaken by numerous local researchers, and proven treatment methods for the specific water conditions have been encountered, technologies have been not yet commercialized due to a lack of interest of authorities, local industries and international agencies for financial and technical cooperation.

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Abbreviations

A: acceptor
ALUFLOC: a household scale low-cost As removal methodology
AMD: acid mine drainage
ARCIS-UNR: process developed by the Center of Sanitary Engineering of the National University of Rosario
CI: capacitive deionization
CYTED: Science and Technology for the Development
D: donor
DMA: dimethylarsinic acid
DOM: dissolved organic matter
ED: electrodialysis
EDR: electrodialysis with reversion of polarity of the electrodes
EPA: Environmental Protection Agency
GFH: granular iron hydroxide
GFO: granular iron oxide
HACRE: Chronic Endemic Regional Hydroarsenicism
HApb: Natural biogenic hydroxyapatite
HP: heterogeneous photocatalysis
HR-XPS: High Resolution X-Ray Photoelectronic Spectroscopy
IARC: International Agency for Research on Cancer
iAs: inorganic arsenic
IBEROARSEN: Iberoamerican CYTED Network
LA: Latin America
MF: microfiltration
MMA: monomethylarsonic acid
NF: nanofiltration
NZVI: nanoparticulate ZVI
PAC: polyaluminum chloride
PRB: permeable reactive barriers
RO: reverse osmosis
SORAS: Solar Oxidation and Removal of Arsenic
SS: suspended solids
TDS: total dissolved solids
UF: ultrafiltration
WHO: World Health Organization
ZVI: zerovalent iron