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## Review

## Remediation of inorganic arsenic in groundwater for safe water supply: A critical assessment of technological solutions

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## HIGHLIGHTS

- ▶ Various conventional treatment technologies for removal of As are critically reviewed.
- ▶ Hybrid or integrated systems may adjust the limitations of conventional processes.
- ▶ Reduction of As to metallic As by ZVI may prevent secondary environmental pollution.
- ▶ Directions for future research in mitigation of As are given.

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## ABSTRACT

Arsenic contaminations of groundwater in several parts of the world are the results of natural and/or anthropogenic sources, and have a large impact on human health. Millions of people from different countries rely on groundwater containing As for drinking purposes. This paper reviews removal technologies (oxidation, coagulation flocculation, adsorption, ion exchange and membrane processes) with attention for the drawbacks and limitations of these applied technologies. The technologies suggested and applied for treatment of As rich water have various problems, including the need for further treatment of As containing secondary waste generated from these water treatment processes. More efficient technologies, with a lower tendency to generate waste include the removal of As by membrane distillation or forward osmosis, instead of using pressure driven membrane processes and subsequently reducing soluble As to commercially valuable metallic As are surveyed. An integrated approach of two or more techniques is suggested to be more beneficial than a single process. Advanced technologies such as membrane distillation, forward osmosis as well as some hybrid integrated techniques and their potentials are also discussed in this review. Membrane processes combined with other process (especially iron based technologies) are thought to be most sustainable for the removal of arsenic and further research allowing scale up of these technologies is suggested.

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## 1. Introduction

As is a well known carcinogen, which is considered as one of the world's most hazardous chemicals (USEPA, 2001). Mass poisoning of the human population by As is a major environmental problem, since millions of people in several countries are exposed to high levels of As via intake of As rich ground water (Smedley and Kinniburgh, 2002). Considering the toxicity of As, the World Health Organization and national agencies have reduced the Maximum Contamination Level (MCL) in drinking water from 50 to 10  $\mu\text{g L}^{-1}$ . However, in some countries like India, Bangladesh, Taiwan, China and Vietnam, the MCL for As is still 50  $\mu\text{g L}^{-1}$  (Nriagu et al., 2007). Acute and chronic exposures to As are the cause of various type of human diseases, including non-pitting edema, respiratory diseases, gastro-intestinal, liver and cardiovascular problems, and ultimately cancer (Mandal and Suzuki, 2002; Kapaj et al., 2006).

Water is the principal route through which As enters into the human body (Chen et al., 2009). Thus treatment of As contaminated water with improved or completely new technologies and providing safe water to the community is an urgent issue at present. Several physico-chemical techniques for removal of As from water, both on-site and off-site, have been developed. The most commonly used technologies are oxidation, co-precipitation followed by adsorption onto coagulated flocks, lime treatment, ion exchange, adsorption onto various solid media and membrane filtration (Ng et al., 2004; Choong et al., 2007; Sharma and Sohn, 2009). However, integrated membrane systems and hybrid technologies coupled with membrane filtration have recently been proposed to reduce the concentration of As in the aquatic environment (Macedonio and Drioli, 2008; Yarlagadda et al., 2011; Jin et al., 2012).

Since exposure to low concentrations of As may be fatal to human health, treatment of contaminated water thus appears to be the only effective alternative. The previously established treatment technologies adopted for As removal from aqueous media have some drawbacks and their intrinsic by-products can be a further potential source for secondary As pollution. Therefore, sources of As in groundwater and different conventional and advance technologies that have been used for a long time for the removal of As from water are evaluated critically. Several sustainable and eco-friendly advanced and hybrid water treatment technologies have also been proposed in this review paper. Thus, this paper provides a concise overview of the source of As in water bodies, and the efforts to solve the problem by conventional as well as new technologies, including the options for new hybrid technologies so as to challenge the menace of As.

## 2. Sources of arsenic in groundwater

In groundwater As commonly exists as an oxyanion of the inorganic form of arsenite [As(III)] and arsenate [As(V)]. However, the presence of As(0) is reported to be very rare (Oremland and Stolz, 2003). While As(V) is predominant in oxic groundwater environments and the major forms are  $\text{H}_3\text{AsO}_4$ ,  $\text{H}_2\text{AsO}_4^{1-}$ ,  $\text{HASO}_4^{2-}$  and  $\text{AsO}_4^{3-}$ , As(III) is more dominant in anoxic environments with  $\text{H}_3\text{AsO}_3$  and  $\text{H}_2\text{AsO}_3^-$  being the common species (Ng et al., 2004; Mohan and Pittman, 2007). Nevertheless, due to slow kinetics of

redox transformation, both the species of As co-exists in natural water and the predominance of the species depends on pH/ORP (Korte and Fernando, 1991). Organoarsenic are more common in water that is affected by industrial pollution (Smedley and Kinniburgh, 2002).

Several natural and anthropogenic sources are responsible for the contamination of As in groundwater. As occurs as a major constituent in more than 200 minerals (Bissen and Frimmel, 2003a) and the desorption and dissolution of naturally occurring As bearing minerals is generally considered as the principal source of As contamination in groundwater (Matschullat, 2000; Polizzotto et al., 2006). Arsenopyrite ( $\text{FeAsS}$ ) is the most abundant As containing mineral that commonly exists in anaerobic environments and can also be found in different concentrations in various rock forming minerals like sulfide, oxide, phosphate, carbonate and silicate (Smedley and Kinniburgh, 2002). It is present as a substitute of S in the crystal lattice of different sulfide minerals. Realgar ( $\text{As}_4\text{S}_4$ ) and orpiment ( $\text{As}_2\text{S}_3$ ) represent the two common reduced forms of As while in arsenolite ( $\text{As}_2\text{O}_3$ ), As exists in oxidized form (Nriagu et al., 2007). However, As comes into water naturally from oxide and hydrous metal oxides minerals due to desorption behavior of arsenate onto these oxides (Smedley and Kinniburgh, 2002).

As can also be found in sediments, in the concentration range from 3 to 10  $\text{mg kg}^{-1}$ , depending on the nature and texture of minerals (Ravenscroft et al., 2009). The mobility and adsorption of As in sediments, clays, and mineral soils depends on the form of As. The areas with high concentrations of Fe oxide or hydrous metal oxide or pyrites contain much higher levels of As in sediments than others. Higher concentrations are found in reducing sediments; the concentration increases gradually with the depth of the sediment (Charlet et al., 2007). Fe and Al oxides in the sediments play a major role for the contamination of As. Reductive dissolution of such metal oxide along with the activity of indigenous metal reducing bacteria is now considered as the dominant release mechanism of As, thereby directly effecting the mobility of As (Islam et al., 2004; Bhowmick et al., 2012).

Mining, burning of fossil fuels, use of arsenical fungicides, herbicides and insecticides in agriculture and wood preservatives are the main anthropogenic sources for contamination of groundwater with As (Nriagu et al., 2007). Burning of coal has an immense effect on contamination of As in environment. Emission of As occurs in the environment by volatilization of  $\text{As}_4\text{O}_6$  due to burning of coal, which condenses in the flue system and is later transferred into water bodies (Bissen and Frimmel, 2003a). Groundwater pollution by these anthropogenic sources is much less compared to the natural sources; however, their contribution cannot be neglected.

Elevated concentrations of As in groundwater have been found in various parts of the world. Because of different socio-political-economical contexts as well as the unavailability of different treatment technologies, the MCL of countries differs (Roy et al., 2008). The most affected countries of the world and their respective MCL are shown in Fig. 1 (Smedley and Kinniburgh, 2002; Choong et al., 2007). The affected countries have different characteristics of their contaminated aquifers and therefore, the mechanism of As mobilization in groundwater is often different. Extensive research has been done with the various As contaminated aquifers

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