



Arsenic in the Environment-Source, Human Health Effect and its Current Removal Technologies

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Abstract

Arsenic is a natural metalloid element. It is naturally present in the water as well as certain anthropogenic sources such as mining, smelting, thermal power plant and industrial processes. Arsenic is found in the environment in inorganic and organic forms as well as different oxidation states such as trivalent arsenite [As(III)] and pentavalent arsenate [As(V)] state. Its oxidation states change in sequential process from pentavalent to trivalent. Adverse health effects of arsenic are related to its chemical form, time and dose. Arsenite is more toxic species of arsenic than arsenate. Recent studies have reported the worldwide poisoning of arsenic in drinking water which is of primary concern to human health problems and remediation of arsenic from water. The present study deals with arsenic toxicity, its effect on the human beings and various conventional and physicochemical process of its remediation with particular emphasis on using nanoparticles for arsenic removal over another conventional process.

Introduction

Arsenic (As) is recognized as one of the world's greatest environmental hazards, threatening the lives of several hundred million people. Biogeochemical processes have resulted in release of naturally occurring As into groundwater and uncontrolled anthropogenic activities such as fossil fuel burning, smelting of metal ores, mining, pesticides, wood preservatives and additives of livestock feed may also release As directly to the environment (Mosaferi *et al.*, 2014). As is one of the natural constituents of the earth materials and ranks 20th most abundant element in the earth's crust, 14th in the seawater and the 12th most abundant element in the human body (Jomova *et al.*, 2011). In ground and surface water, the major chemical forms of As are the arsenite [As(III)] and

arsenate [As(V)] which are inorganic species of As. Organic As species are less prevalent in the environment but can be found in surface water and areas affected by industrial pollution. Inorganic As can interact with sulfur to form arsenic-sulfur species called thioarsenic. These thiolated species such as thiolated arsenites and thiolated arsenates can exist in iron rich or sulfide rich waters, particularly when arsenopyrite mineral undergo dissolution. Inorganic arsenic species are methylated to form monomethylarsonic acid [MMA(V)] or dimethylated as in dimethylarsinic acid [DMA(V)]. As(III) inhibits pyruvate dehydrogenase by binding to the sulfhydryl groups of dihydrolipoamide which reduces the transformation of pyruvate to acetyl coenzyme A (CoA), while both citric acid cycle activity and production of cellular ATP are decreased. It also inhibits numerous other cellular enzymes through binding of sulfhydryl group. It also inhibits the uptake of glucose into cells, gluconeogenesis, fatty acid oxidation and further production of acetyl CoA (Platanias, 2009). However, arsenobetaine and arsenocholine are the predominant occurring As species in the marine animals in contrast to the terrestrial animals. The structure of As species which require human monitoring is shown in figure 1.

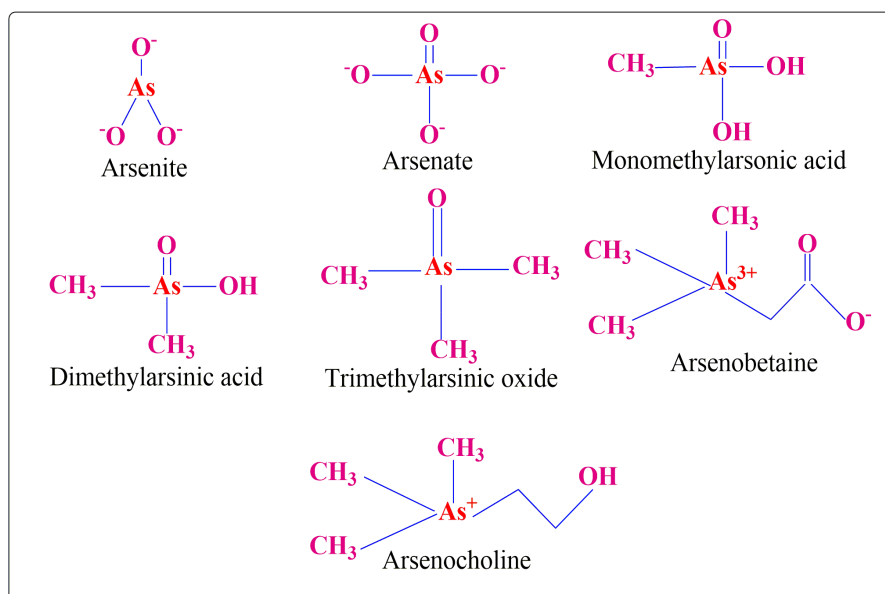


Figure 1. Structure of As species

The World Health Organization (WHO) has established 10 $\mu\text{g/L}$ as the maximum contaminant level for total As in potable water due to its significant toxicity. In the air, As concentration ranged from 0.02 to 4 ng m^{-3} in rural areas, 3–200 ng m^{-3} in urban areas to >1000 ng m^{-3} in industrial zones (WHO, 2011). Short and long term exposure to As can result in acute and chronic health effects. Acute health effects include gastrointestinal discomfort, vomiting, diarrhoea, bloody urine, anuria, shock, convulsions, coma, and death while chronic effects includes conjunctivitis, hyperkeratosis, hyper pigmentation, cardiovascular diseases, disturbance in the peripheral vascular and nervous systems, skin

lesions and black foot disease. Chronic exposure to As can result in carcinogenic effects such as cancer of the bladder, lungs, skin, kidney, nasal passages, liver and prostate gland. The International Agency for Research on Cancer (IARC) has classified As as a known human carcinogen (Group 1) based on epidemiological studies that showed an increase in skin cancer following As medical treatments (IARC, 2004). As species are converted among As(III), As(V) and As(-III) oxidation state by oxidation, reduction, methylation and demethylation in the various environment such as solid, aqueous and gaseous (Figure 2).

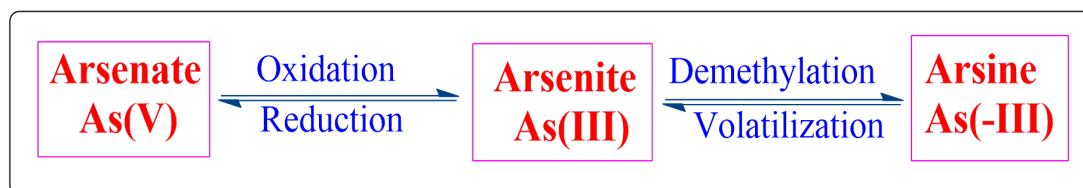


Figure 2. As transformation in the environment (Tamaki and Frankenberger, 1992)

In different countries of the world, As is found in groundwater in India, Bangladesh, China, Chile, Argentina, Mexico, Hungary, Taiwan, Vietnam, Japan, New Zealand, Germany and the United States due to naturally occurring As in the aquifer bed (Barringer and Reilly, 2013). In India, 12 million people in West Bengal are affected by As contamination. Further, seven of the nineteen districts of West Bengal have been reported to have ground water As concentrations above 0.05 mg/L. As in groundwater was also detected in parts of North-eastern states, Assam, Arunachal Pradesh, Manipur, Nagaland and Tripura (Haque *et al.*, 2003). Most As affected states in India are Assam, West Bengal, Bihar, Uttar Pradesh and Karnataka (Mishra *et al.*, 2016) as given in figure 3.

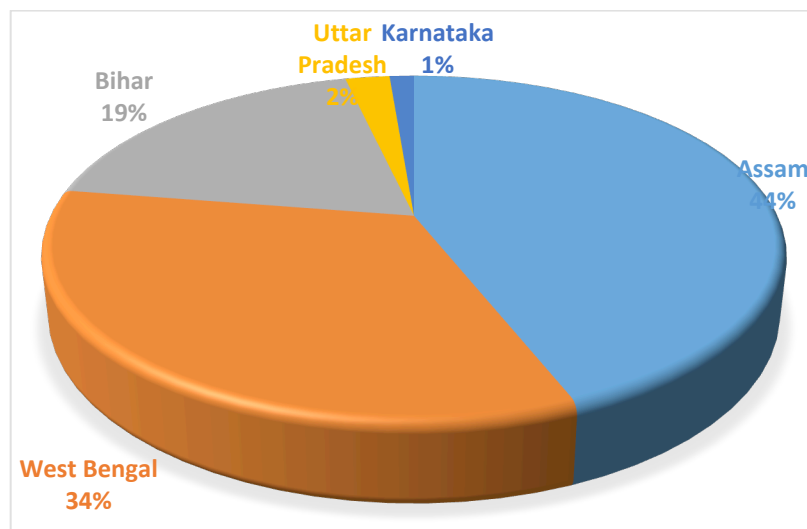


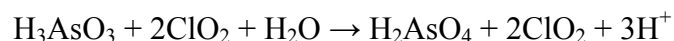
Figure 3. Most arsenic affected state in India (Bhattacharya, 2019)

Apart from these, Punjab state is also affected by the As (Hundal *et al.*, 2007). The ground water of the arid southwestern region of Punjab such as Bathinda, Mansa, Faridkot, Sangrur, Muktsar and Ferozepur was reported to contain 11 to 688 µg/L As in water which is exceeded more than 20 to 30 folds of the WHO safe limit (Virk, 2018). An epidemiological study by PPCB-PGIMER found that Bathinda surface water is contaminated with As, Cd, Cr, Se and Hg. Report of Central Ground Water Board (CGWB, 2011), Ministry of Water Resources and Government of India, also confirmed that As contamination in ground water in district Mansa, Punjab exceeded permissible limit. Punjab is known for agriculture and activities related to it. The soil characteristics in many villages in the cotton belt of Punjab changed due to excessive use of As based pesticides and irrigation of soil by As rich groundwater for decades. Application of deep irrigated water is one of the primary sources of As contamination in Punjab and uses of agricultural pesticides, herbicides, and fertilizers are probably reasons given for sources of As in Punjab and Bathinda region (Hundal *et al.*, 2013). So, in light of its availability in higher concentration and toxicity in human being, there is need to remediate the As from water by these physicochemical and conventional technologies.

Technologies for treatment of arsenic

The most conventional processes used for As removal alone or in combination are oxidation, coprecipitation and adsorption onto coagulated flocs, lime treatment, adsorption onto suitable surfaces, use of ion exchange resins and membrane technologies.

Oxidation and reduction: Oxidation is a first step required to transform As(III) species in more easily removable As(V) species. Simple direct aeration is slow, but many chemicals, including gaseous chlorine, hypochlorite, chlorine oxide ozone, permanganate, hydrogen peroxide, manganese oxides can be employed to accelerate oxidation.



The major disadvantage of this method is some oxidants produce toxic and carcinogenic by-products which need further treatment (Litter *et al.*, 2010).

Precipitation: This method is used for the removal of inorganic arsenical compounds such as As(III) sulfide, calcium arsenate and ferric arsenate from water. The addition of insoluble ferric hydroxide in the water can precipitate As by the iron co-precipitation process. The major disadvantage of this method is overdosing of iron with the addition of iron salt (Ravenscroft *et al.*, 2009).

Lime softening: In the presence of water and carbonic acid, lime forms calcium carbonate and adsorbs As, the process ending with a coagulation step. This method is efficient to treat water with high hardness, especially at $\text{pH} > 10.5$. The disadvantage of this process is a very high pH in the resulting water (10–12), which implies a further acidification, a very high dose of coagulant is needed with relatively low removal efficiencies (less than 1 mg/L) and also secondary treatments is required (Holl, 2010).

Coagulation and filtration: It is the most common technology for As removal. As is removed in the pentavalent form, which adsorbs onto coagulated flocs and then removed by filtration. Mostly used coagulants include aluminium sulphate $[\text{Al}_2(\text{SO}_4)_3]$, iron chloride (FeCl_3) and ferrous sulfate (FeSO_4). FeCl_3 generates relatively large flocs, while smaller ones are formed with FeSO_4 . Filtration is an inevitable step. Without filtration, arsenate removal is around 30%, however, if 0.1 or 1.0 mm filter is used, the removal percentage increases arsenate to more than 96%. The primary disadvantage of this process includes low removal efficiency, pH adjustment and proper disposal of the As contaminated coagulation sludge (Pal and Paknikar, 2012).

Membrane processes: In the membrane processes, microfiltration (MF) and ultrafiltration (UF), which use low-pressure membranes having large pore sizes, 10–30 psi are not entirely adequate because the arsenical species are microscopic and can traverse the membranes. In contrast, nanofiltration (NF) and reverse osmosis (RO), use high pressure membranes (75–250 psi). In RO, an external pressure is applied to reverse natural osmotic flow and water flow 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 As oxyanions. 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 (Choong *et al.*, 2007).

Adsorption: Adsorption is the most efficient process for As remediation due to its easy operation and low cost. Adsorbents are mainly divided into four classes according to their nature such as natural, biogenic, agriculture and chemical adsorbent **Fig. 4**. Aluminium oxides, iron oxide, titanium dioxide, cerium oxide, or reduced metals can be used as adsorbents. Granular activated alumina (Al_2O_3) is a commercially available porous oxide, successfully applied at slightly acid pH (5–7), giving efficiencies higher than 95% for both As(V) and As(III).

The technology is very simple, does not require chemical addition and is used for community or household levels. Granular iron hydroxide, a synthetic akaganeite, proved to be a suitable material, able to retain As(V) and As(III). Granular iron oxide (Bayoxide) is another similar advantageous material, containing less than 70% of Fe_2O_3 . Commercial titanium dioxide, cerium oxide and manganese dioxide also proved to be effective. The limitations are its moderate efficiency and regeneration, interferences of Se^- , F^- , Cl^- and

SO₄²⁻ ions and application of use treatment devices needs regeneration and replacement (German *et al.*, 2014).

Ion exchange resins: Synthetic ionic exchange resins such as polystyrene cross-linked with divinyl benzene linked to charge functional groups can be applied for the removal of As and quaternary amine groups. Arsenate removal is efficient in producing effluents with less than 1 mg/L of As. The disadvantage is that arsenite is not removed, and a previous oxidation step is necessary and precipitated iron cause clogging and require pre-treatment (Pal and Paknikar, 2012).

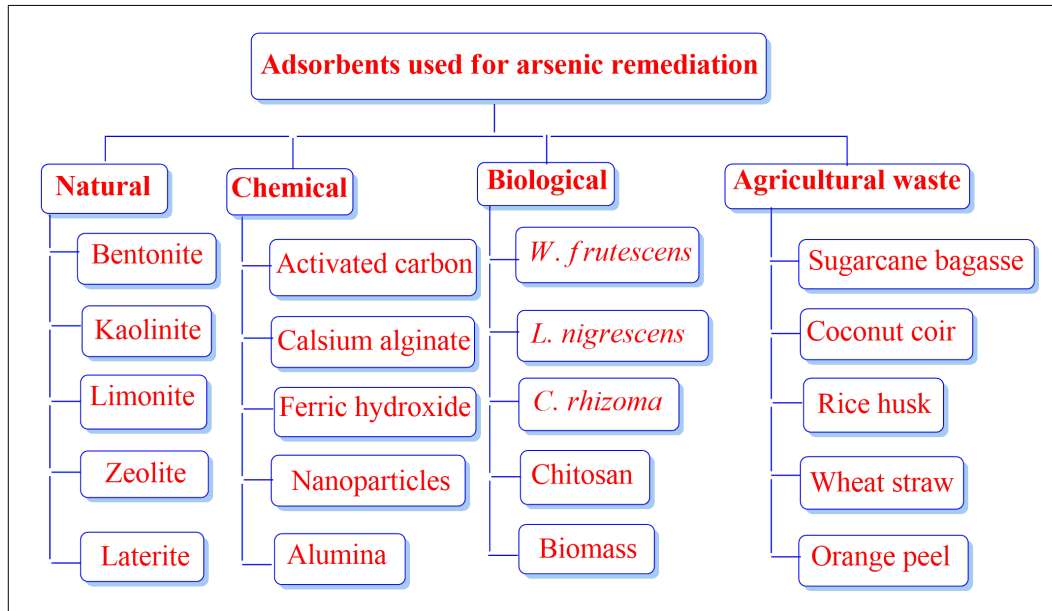


Figure 4. Different types of adsorbent used in arsenic removal

Nanoparticles and their relevance in arsenic removal: Various nanoparticles have been used for the remediation of As such as silver nanoparticles (Selvakumar *et al.*, 2011), iron oxide nanoparticles (Carabante *et al.*, 2009; Luther *et al.*, 2012), TiO₂ nanoparticles (Jegadeesan *et al.*, 2010), zirconium nanoparticles (Ma, 2011), calcium peroxide nanoparticles (Olyaie *et al.*, 2012), magnetite nanoparticles (Wang *et al.*, 2011) and copper oxide nanoparticles (Goswami *et al.*, 2012) as given in Table 1. Nanotechnology offers the potential use of nZVI for the treatment of surface water, groundwater, and wastewater contaminated by toxic metal ions, organic and inorganic solutes, and microorganisms. nZVI have particles with dimensions ranging from 1-100 nm.

The enormous surface area to volume ratio and small particle size of nanoparticles (a large number of reactive sites); account for their high reactivity and smooth delivery through small spaces in the subsurface to contaminated sites. It also has remarkable

properties to immobilize aqueous As species. nZVI in the form of powder is a strong reducer, easy to get and a magnet could readily recycle the spent iron particles. This nanomaterial is highly preferred for in-situ remediation because of time and cost effectiveness. *In-situ* remediation involves treatment occurring on site thereby eliminating the need to pump out groundwater for above ground treatment or the transportation of soil and water to any other places for contaminant clean up. These advantages sparked a great interest in nZVI for remediation (Selvarani *et al.*, 2012).

Table 1. Remediation of arsenic from water using nanoparticles

Nanoparticles	Adsorbate	Adsorption efficiency (mg/g)	Reference
Porous α -Fe ₂ O ₃	As(V)	5.31	Zhong <i>et al.</i> , 2006
Porous γ -Fe ₂ O ₃	As(V)	4.75	Zhong <i>et al.</i> , 2006
Follow-like porous Fe ₃ O ₄	As(V)	4.65	Zhong <i>et al.</i> , 2006
Chestnut like Fe ₃ O ₄	As(V)	6.07	Mou <i>et al.</i> , 2012
Commercial α -Fe ₂ O ₃	As(V)	0.46	Zhong <i>et al.</i> , 2006
Commercial Fe ₃ O ₄	As(III) and As(V)	1.56 and 1.08	Yean <i>et al.</i> , 2005
Fe ₃ O ₄	As(III) and As(V)	6.77 and 7.23	Wang <i>et al.</i> , 2013
Copper nanoparticle	Arsenic	1.08	Goswami <i>et al.</i> , 2012
Zirconium nanoparticle	As(V)	256.4	Ma <i>et al.</i> , 2011
nZVI	Arsenic	20	Sun <i>et al.</i> , 2006
nZVI	As(III) and As(V)	55 and 17	Giasuddin <i>et al.</i> , 2007
nZVI	As(III) and As(V)	14 and 12.2	Mosaferi <i>et al.</i> , 2014
nZVI	As(III) and As(V)	18.2 and 12	Zhu <i>et al.</i> , 2009
nZVI	As(V)	9.2	Bezbaruah <i>et al.</i> , 2014
nZVI	As(III)	49.5	Tandon <i>et al.</i> , 2013
nZVI	As(III) and As(V)	59.9 and 45.5	Bhowmick <i>et al.</i> , 2014

Mechanism of arsenic remediation by nZVI: nZVI consist of a polycrystalline Fe⁰ core encapsulated by a thin layer of the shell of amorphous iron oxide which is approximately 3 nm thick. This fine scale oxide film may enhance the rate of electron transfer or mass diffusion across the oxide layer. Thus, the nanoparticle may exhibit redox behavior on a small scale (Ramos *et al.*, 2009). As is found in multiple valence states including As(V), As(III) and As(0). These valence states are distributed in a layered structure with As(V) existing at the oxide surface, As(III) distributed across the oxide shell and As(0) residing in a subsurface region close to the Fe⁰ core. nZVI have dual redox capability i.e. As(III) oxidation occurred at oxide shell and reduction on the Fe⁰ core.

The gradual transition from a mixed Fe(II)-Fe(III) oxide with Fe(0) to ferric oxyhydroxide at the outer periphery can generate oxygen vacancies and lattice disorder which lead to elevated charge transfer and ionic mobility (Yan *et al.*, 2012). The aqueous arsenite is adsorbed at the surface of the iron oxide layer of nZVI and reduced arsenite complex forms the As-O bond at the surface. Translocation of As across the oxide shell breaks As-O bond and is diffused into the Fe⁰ core. As accumulates at the surface of Fe⁰ forming a thin layer of Fe-As compound (Figure 5).

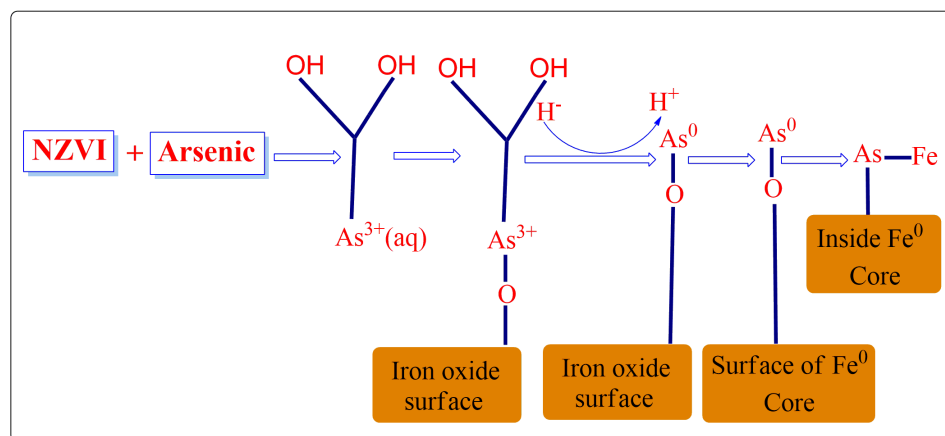


Figure 5: Arsenic remediation mechanism by nZVI

Conclusion

As occurs in natural waters predominantly in inorganic forms, such as arsenite and arsenate. The presence of As in water for human consumption causes endemic diseases. A wide range of methods has been developed for As remediation from the water. Among these conventional processes, adsorption is the most efficient process due to the low-cost adsorbent and is easy to handle. However, nanoparticles have been proposed for both *in-situ* and *ex-situ* application for removal of As from drinking water by adsorption or co-precipitation. For the remediation of As, a growing body of theoretical and empirical evidence has proven nZVI as both highly efficient and versatile.

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