

Arsenic Removal from Water: A Review

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Abstract: Arsenic contamination of underground water has been reported from many parts of the world. In most of the affected areas, substitution of drinking water source by a safe and easily available one may not be possible. Arsenic removal from these water sources is a more appropriate option. This article gives a brief overview over the possible arsenic removal options for safe drinking water supply in the arsenic affected areas. All these options were tested in the laboratory and effect of various parameters such as pH, initial arsenic concentration, arsenic speciation, dose of chemical used, interference by coions etc. was studied. Adsorption of arsenic on iron salts such as Granular Ferric Hydroxide (GFH) and silica ferric complex adsorbent (Sfca) was found to be most effective option for arsenic removal.

Key words: Arsenic, groundwater, drinking water, treatment, adsorption.

Introduction

Water is an essential natural resource for sustaining life and environment that we have always thought to be available in abundance and free gift of nature. However, chemical composition of surface or subsurface is one of the prime factors on which the suitability of water for domestic, industrial or agricultural purpose depends. Freshwater occurs as surface water and groundwater. Though groundwater contributes only 0.6% of the total water resources on earth, it is the major and the preferred source of drinking water in rural as well as urban areas, particularly in the developing countries like India because treatment of the same, including disinfection is often not required. It caters to 80% of the total drinking water requirement and 50% of the agricultural requirement in rural India. But in the era of economical growth, groundwater is getting polluted due to urbanization and industrialization.

In general, groundwater may contain three types of contaminants: biological, organic (from industrial and

agricultural waste leachates) and inorganic (mineralogical). Biological contamination can be avoided by creating awareness among masses for cleanliness and proper group sanitation. The leaching of industrial wastes can be avoided by banning its disposal directly in the surface water bodies or agricultural lands and making it mandatory to dispose it off in the municipal sewer system after due secondary treatment. Minimum use of chemical fertilizers and pesticides by promoting biofertilizers and biopesticides would also reduce the groundwater contamination from agricultural leachate. However, groundwater is particularly vulnerable to mineralogical contamination (e.g. fluoride, arsenic, iron, nitrate etc.) as a result of natural geochemical processes, which cannot be avoided. Unlike bacteriological pollution, the effect of these excess chemical constituents on human health is chronic in nature and manifest after consuming the water over a long period of time. Hence, the solution to this problem is to find out an alternate source of water and if it is not available, the only option left is to remove the unwanted minerals from the water before use, because various complex disease syndromes have been attributed to the regular consumption of water contaminated with

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critical levels of these toxic minerals. Presence of various hazardous contaminants like fluoride (Susheela, 1993), arsenic (Meng et al., 2001), nitrate (Nemade et al., 1997), sulfate (Subba Rao, 1996), other heavy metals (Gupta and Nathawat, 1991) etc. in underground water has been reported from different parts of India. In many cases, the water sources have been rendered unsafe not only for human consumption but also for other activities such as irrigation and industrial needs. Therefore, now there is a need to focus greater attention on the future impact of water resources planning and development taking into consideration all the related issues. In India, fluoride and arsenic are the major inorganic pollutants of natural origin found in groundwater. In this paper, detailed review on sources, ill effects and techniques available for arsenic removal is done.

Arsenic is a naturally occurring element found in soil, surface water and groundwater as a metalloid and in the form of organic and inorganic compounds (Matschullat, 2000). Drinking water has been identified as one of the major sources of arsenic ingestion by the general population (Saha et al., 2001). Its presence in fresh water may be attributed to geochemical reactions, release from subsurface minerals and sediments facilitated by various redox conditions, natural weathering, mining, waste discharges from metal, petroleum and pharmaceutical industries, pesticide manufacturing and agricultural use of arsenical pesticides, burning of fossil fuels, waste incineration etc. (Nickson et al., 1998; Chowdhary et al., 2000; Bagla et al., 1996). Arsenic does not occur in elemental state and is more common as sulfide and sulfate salts. It forms stable complexes with Fe (hydr-) oxides and aluminium (Dzombak and Morel, 1990; Halter and Pfeifer, 2001). The current Maximum Contaminant Level (MCL) for arsenic in drinking water as permitted by WHO standards is 10 ppb (Pontius et al., 1994). Arsenic levels above permissible limit of 10 ppb in groundwater have been reported from many parts of the world (Smith et al., 2002). Arsenic in groundwater was first detected in 1978 following reports of many people suffering from arsenical skin diseases. By 1995, the problem emerged as an universal issue. Arsenic toxicity is a serious health hazard in countries like Bangladesh, West Bengal region of India, inner Mangolia region of China, Taiwan, Japan, Thailand, Argentina, Ghana, Chile, Mexico, U.S.A., Hungary and Romania (Anawar et al., 2002; Chakravarty et al., 2002; Dhar et al., 1997; Davis et al., 1994; Meenakshi and Maheshwari, 2002).

The groundwater of vast areas in the Ganges Delta in West Bengal (India) and Bangladesh is highly contaminated by arsenic (Joshi et al., 2003; Rasul et al.,

2002). Of all the 18 districts of West Bengal together having 341 blocks, 10 districts with 69 blocks have already been identified as severe arsenic contaminated areas, which include many places around Kolkata, such as Barasat, Habra etc. in North 24-Parganas. The problem of arsenic contamination of groundwater is more serious in Bangladesh, where the groundwater in 59 of the 64 districts is contaminated with arsenic and about two-third of the population is exposed to 'the biggest mass-poisoning case' the world has ever known. Chronic toxicity of arsenic is manifested as (Mandal et al., 1996; Mazumdar et al., 1998):

- General weakness, loss of appetite, nausea, anaemia, inflammation of mucous membrane of the eyes, nose and larynx. Melanosis, keratosis and unusual pigmentation.
- Cancer of lung and liver, gastrointestinal injuries, kidney damage, circulatory collapse, respiratory failure, reproductive, neurological, genotoxic and cardiovascular disorders.

The nature and degree of arsenicosis varies from case to case depending on the arsenic level in drinking water and food, magnitude, frequency and duration of exposure, nutritional status of the subject, synergistic and antagonistic effect of other elements, genetic variant of human being etc. (Tondel et al., 1999). A diet rich in vitamin C, methionine, carbohydrate and protein contents protects against the toxicity of arsenic.

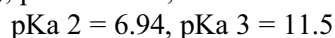
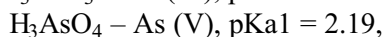
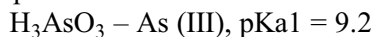
Although arsenicosis is not an infectious, contagious or hereditary disease, it creates social problems for the victims and their families. Skin lesions caused by arsenicosis are often confused with leprosy. Affected villages are treated much like isolated leper colonies. Sufferers are barred from social activities and often face rejection even by immediate family members. Thus, it is urgently needed to find alternate sources of safe drinking water or to develop some feasible technique for arsenic removal from contaminated water.

Arsenic Chemistry

The chemistry of arsenic in aquatic system is quite complicated as under different redox conditions, inorganic arsenic is stable in the four oxidation states (+5, +3, 0 and -3). However, in groundwater the arsenate (H_3AsO_4 , H_2AsO_4^- , HAsO_4^{2-} , AsO_4^{3-}) and arsenite (H_3AsO_3 , HAsO_3^{2-} and H_2AsO_3^-) species are more predominant (Ferguson and Anderson, 1974; Ning, 2002). Arsenate is the thermodynamically stable form of inorganic species and it generally predominates in

surface waters. Arsenite is favoured under reducing conditions, thus more likely to occur in anaerobic ground waters (Breslin, 1998). However actual dominance of arsenic species is a function of pH and redox potential (Christopher, 2004).

It is well established that the toxicity of arsenic depends on its chemical form. Arsenite, the trivalent form, is far more toxic in biological systems than arsenate, but the toxicity of organo-arsenicals is generally lower than that of inorganic species. Arsenate and arsenite have significant differences in the chemical behaviour. The equilibrium constants of dissolution of the two oxidation states of arsenic are quite different as reported below:



The stability and dominance of a specific species depends on the pH of the solution. In general, arsenate sorption is characterized by an apparent sorption maximum at a pH value of 4 (Pierce and Moore, 1992). In contrast, arsenite sorption is characterized by a sorption maximum occurring in the pH range of 8.5 to 11.0 (Manning and Goldberg, 1997). Removal of As (III) from aqueous solutions is usually poor as compared to that of As (V) by all the available treatment methods because in the pH range of 4 to 10, As (V) exists in water as monovalent or bivalent anions having negative charge, whereas As (III) exists predominantly as a neutral species, which cannot be removed by charged ion exchange resin or adsorbent. Therefore, a pre-oxidation step is required in order to remove arsenite effectively. In typical pH ranges of natural waters, the species H_2AsO_4^- and HAsO_4^{2-} are dominant. The oxidizing agents being used commonly are manganese oxide, ozone, iron, oxygen, chlorine, hypochlorite, hydrogen peroxide, Fenton's reagent ($\text{H}_2\text{O}_2/\text{Fe}^{2+}$), permanganate, UV light, ferric chloride, sonication etc. (Emett and Khoe, 2001; Myoung, 2000).

Techniques Available for Arsenic Removal

Various technologies are available for arsenic removal from water such as conventional co-precipitation with ferric chloride, lime softening, ion exchange resins, electrodialysis reversal, membrane filtration, adsorption etc. (Kartinen and Martin, 1995). As optimum arsenic removal is dependent on feed water characteristics and the existing operating conditions, various factors such as oxidation state of arsenic, pH, TDS, competing ions especially silicate, phosphate, fluoride and sulfate, point

of treatment cost etc. have to be considered while adopting any technique for arsenic removal. A brief description regarding each of these technologies is given here.

1. Coagulation/Precipitation

In this technique, addition of coagulant facilitates the conversion of soluble arsenic species into insoluble products by coprecipitation and adsorption. The most commonly used salts are calcium salts particularly lime (Nishimura and Robins, 1998), aluminium salts mainly alum (Gregor, 2001) and iron salts such as ferric chloride and ferrous sulfate (Hering et al., 1997). The calcium, aluminium and ferric salts undergo hydrolysis to give insoluble precipitate in the form of calcium arsenate, arsenic (III) sulfide and ferric arsenate respectively, which can be filtered off completely. Arsenate removal upto 99% can be achieved by this process under optimal conditions, but As (III) removal by calcium, aluminium and ferric salts is much lower than that of arsenate. Therefore, oxidation of As (III) to As (V) is necessary to achieve effective removal (Zouboulis and Katsoyiannis, 2002). Ferric sulfate has been found to be the most effective coagulant for arsenate removal. As compared to ferric salts, aluminium compounds (alum) are less effective (80-90%) in removing arsenic from water because they are less soluble in water than the ferric salts. When ferric coagulants are added, all the available iron gets precipitated as $\text{Fe}(\text{OH})_3$. However, not all the aluminium added as alum coagulant precipitates as $\text{Al}(\text{OH})_3$. Also FeCl_3 coagulation is not so pH specific whereas pH dependence for alum is much more pronounced.

The presence of co-occurring inorganic solutes such as sulfate, phosphate and silicates has a pronounced effect on the removal of arsenic by this process as they directly compete with arsenic for surface binding sites and may also influence the surface charge of ferric oxide (Meng et al., 2000). Silicates inhibit the removal of arsenic by precipitating out with FeCl_3 .

Limitations

- This process is inappropriate for small systems.
- It is effective for arsenic removal, but type of coagulant and dosage used affect its efficiency.
- It is a pH specific process. Deviation from the optimal pH significantly reduces its efficiency.
- Removal of As (III) is much less than that of As (V).
- Disposal of bulky sludge becomes a problem.
- Silicates, phosphates and alkalinity cause interference in arsenic removal.

2. Lime Softening

This process is similar to coagulation with metal salts. Two types of chemical additions are involved in this process. First, lime $\text{Ca}(\text{OH})_2$ is added in sufficient quantity to raise the pH to about 10 to precipitate out the carbonate hardness. Lime hydrolyzes and combines with carbonic acid to form calcium carbonate, which acts as the adsorbing agent for arsenic removal. Next, sodium carbonate is added to precipitate out the non-carbonate hardness. This process is typically used only with very hard waters and shifts the pH of treated water to markedly higher side in the range of 10-12. The pH and presence of chlorine significantly affect the arsenic removal efficiency of lime. The maximum arsenate removal is obtained above pH 11 and much poor removal occurs at pH 10 or less. With the use of chlorine, the arsenic removal rate is always more than that without chlorine (Kartinen and Martin, 1995).

Limitations

- The process operates at optimum pH of ≥ 10.5 .
- This process is unable to reduce the arsenic concentration to MCL. Therefore, some secondary treatment is required along with lime softening to meet the goal.
- Higher sulfate concentration in water interferes with arsenic removal.
- Large coagulant dose is required (800-1200 mg/l), producing large volumes of sludge, the disposal of which becomes a problem.
- Calcium arsenate formed in the process has to be disposed off in a landfill site where it may interact with CO_2 in air to form calcium carbonate releasing arsenic back into the environment.
- Operational care is required.

3. Ion Exchange

For arsenic removal, an anion exchange resin, usually loaded with chloride ions at the exchange sites is placed in vessels and the water to be treated is passed through them. Arsenic gets exchanged for the chloride ions (Korngold et al., 2001). The water exiting the vessel is lower in arsenic but higher in chloride as compared to the feed water. Eventually the resin becomes exhausted and has to be regenerated by passing saturated solution of sodium chloride through it.

As with the other processes, it is important that the arsenic should have (5+) oxidation state and that the pH of the feed water be in the range of 6.5 to 9.0 in order to achieve the best removal rates. Since the ion exchange is effective only in removing ions from the water, arsenic

that is in an ionic form only is exchanged for chloride in this process. These resins can effectively remove arsenate producing product water with less than 1 $\mu\text{g/l}$ of arsenic. Arsenite, being uncharged, is not removed (Datta et al., 1997). Therefore an oxidation step is necessary if arsenic is present in the reduced form.

Limitations

- Interference by sulfate, TDS, selenium, fluoride and nitrate is a major limitation of the process.
- As(III) cannot be removed without oxidation to As(V) and the ion exchange resins are, as a rule subject to degradation by oxidizing agents.
- The regenerant solution contains a high concentration of arsenic and therefore may pose a serious environmental problem. The ion-exchange regenerant wastes are most difficult to be treated for meeting in-stream standards.

4. Membrane Process

Arsenic removal efficiencies upto 98% have been documented by membrane process (Ning, 2002). Literature reveals that like all other techniques, RO is more effective in removing As(V) than As(III). Thus, to achieve best results, the feed water must be treated with an oxidizing agent to convert As(III) to As(V). However, the oxidizing agents including chlorine are, in general, harmful to RO membranes. Some membranes can withstand small concentrations of oxidizing agents on an infrequent basis. Therefore, the life of RO membranes gets substantially shortened if an oxidizing agent is injected into the feed water.

Limitations

- Less effective for As(III).
- The oxidizing agents are harmful to membranes.
- Waste disposal problem.

5. Adsorption

Adsorption removes arsenic by a physical/chemical separation process whereby arsenic molecules adhere to the surface of the adsorbent with which they come into contact, due to forces of attraction at the surface. Various adsorbents used for this purpose are activated carbon, activated alumina, flyash, ferric oxides, titanium oxide, natural manganese oxide, iron oxide/manganese oxide coated sands, zero valent iron, rare earth oxides, Fe(III) hydroxide loaded coral limestone (CaSiCo), bone charcoal, cellulose materials such as saw dust and newspaper pulp, biological materials such as living or non-living biomass etc. (Ouvrard et al., 2002; Raichur

and Penvekar, 2002). Also several derived products of biological origin, such as alginate, chitin, chitosan etc. have been found effective for arsenic removal. Activated alumina and ferric salts are the most commonly used adsorbents for arsenic removal.

(a) Activated Alumina

Activated alumina is a highly porous and adsorptive medium with excellent arsenic removal efficiency. The process is dependent on pH of feed water, influent arsenic concentration and speciation. Its arsenate removal capacity is best in the narrow pH range from 5.5–6.0, in which the alumina surfaces are protonated. Typically activated alumina has a point of zero charge (PZC) at pH 8.2, below which the surface is positively charged and above which the surface bears a negative charge. Arsenic removal capacity drops sharply as the PZC is approached and above pH 8.5, it is reduced to only 2–5% of its arsenic removal capacity at optimal pH. The arsenite form has to be oxidized to arsenate for optimal arsenic removal. The sorption sites on the activated alumina surface also attract a number of other anions like phosphates, sulfates, fluorides, bicarbonates etc. Phosphate ions offer significant interference in arsenic removal by activated alumina because of chemical and structural similarity of P (V) with As (V) and similar pK_a values resulting in a strong competition for binding sites (Chen et al., 1999). After continuous use, all the binding sites on adsorptive medium become loaded with arsenic and it has to be regenerated, which is accomplished by flushing it with a solution of 4% NaOH which displaces arsenic from the alumina surface. This is followed by flushing with acid to re-establish a positive charge on the alumina surfaces (Amy et al., 2000).

Limitations

- This process is not effective for arsenite removal, because arsenite remains uncharged until it reaches its first pK (pH 9.3) above which it becomes negatively charged. Therefore, arsenite is not possible to be removed by alumina, which has positive charge only upto pH 8.2.
- It operates optimally at a relatively narrow pH range.
- Arsenic removal is interfered by phosphate and other ions.
- A significantly longer empty bed contact time (5–8 minutes) is required.
- Alumina loses about 30–40% of its capacity with each regeneration, so it may not be efficient after 3–4 regenerations and must be replaced.
- Disposal of concentrated regenerant is a problem.

(b) Ferric Salts

Development of iron-based adsorbents has been found to be one of the most promising solutions to the problem. Several Fe (III) oxides such as granular ferric hydroxide (GFH), poorly crystalline hydrous ferric oxide (ferrihydrite), goethite (α -FeOOH) and zero valent iron etc. are promising adsorbents for removing both As (V) and As (III) from aqueous solutions (Wilkie and Hering, 1996; Sun and Doner, 1998). Both arsenate and arsenite have strong affinities for iron complexes; however they behave oppositely w.r.t. pH. In general, in the pH range 5–10, adsorption of arsenate decreases with increasing pH, while that of arsenite increases (Jain and Loeppert, 2000). Arsenic adsorption by iron complexes occurs by ligand exchange of arsenic for OH₂ or OH[−] in the coordination spheres of surface structures of Fe atoms.

GFH is a poorly crystallized beta-FeOOH. It is a relatively new product developed in Germany for selective removal of arsenic from natural water. It has a specific surface of 250–300 m²/g and a porosity of 75–80%. The bulk density of GFH saturated with water is 1.32 g/cm³. The grain size ranges from 0.2 to 2.0 mm. As (III) and As (V) bind on the surface of ferric hydroxide building inner-sphere complexes. Studies have shown that it is 5–6 times more efficient than activated alumina, whereas residual mass-produced is only 1/10th of that of activated alumina (Pierce and Moore, 1992). It employs packed beds of GFH and gives longer run than AA. It needs shorter EBCT (Empty Bed Contact Time) and the adsorbent is less sensitive to pH which can be used in the pH range 5.5 to 9.0. However, the arsenate adsorption decreases slightly with pH, typical for anion adsorption.

At present 16 drinking water treatment plants in Germany are using this technique for arsenic removal (Deiehaus, 2002). Of all the known arsenic removal systems, the adsorption of arsenic by GFH in a fixed bed reactor is the most simple, safe and effective method for elimination of arsenic from contaminated groundwater. The plants are easy-to-install, compact and virtually maintenance free. The exhausted GFH medium is not considered regenerable and is simply thrown away. The spent GFH is a non-toxic solid waste and its volume being small (5–25 g/m³ of treated water), disposal is less problematic. Under normal environmental conditions, no leaching of arsenic takes place out of the spent GFH (Pal, 2001).

Limitations

- Potential head loss of adsorbent occurs during regeneration; therefore it is only suitable for one-time use.
- Poor physical integrity demands longer settling time.
- It imparts colour to the product water due to significant disintegration after contacting with water, which is its major drawback.
- It is not easily available in India.

To circumvent all these problems, a new ferric silica complex adsorbent (Sfca) has been developed in the laboratory of CRDT, IIT, Delhi, which has greater physical integrity and comparable arsenic removal capacity. It does not impart colour to water, needs shorter settling time and the loss of adsorbent is also very less. Experiments have shown that it can remove both arsenate and arsenite upto 90% under wide pH range and due to its high adsorption capacity, residual mass produced is very small reducing the disposal problem.

Conclusion

The new adsorbent developed at CRDT, IIT, Delhi might provide a cost effective, user friendly and technically viable solution to arsenic problem in groundwater in years to come.

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