

Occurrence and methods to remove arsenic and fluoride contamination in water

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Received: 26 April 2016 / Accepted: 4 October 2016 / Published online: 15 October 2016
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Abstract Groundwater contamination with arsenic and fluoride has affected over 300 million people worldwide. Ingestion of arsenic and fluoride for extended period of time or at high concentration causes severe health effects. Arsenic can cause thickening and discoloration of the skin, cardiovascular disorders, cancer and skin lesions. Excessive intake of fluoride leads to dental and skeletal fluorosis and bone deformities. This report reviews the distribution of arsenic and fluoride contamination, their sources, mobilization and associated health risks. Remediation technologies to remove arsenic and fluoride are presented.

Keywords Arsenic · Fluoride · Contamination · Health hazard · Treatment technologies

Introduction

Various natural and anthropogenic activities cause metal and non-metal contamination of different aquatic and terrestrial ecosystems. Agricultural and industrial based societies are being exposed daily to hazardous agricultural pesticides, herbicides, uncontrolled sewage discharge, toxic metals and other industrial chemicals (Husain et al. 2008; Bibi et al. 2008). Being toxic, arsenic and fluoride both are one of the biggest contributors to global water crisis and pose major health concerns resulting from drinking water (Bibi et al. 2015a). This issue has been aggravated due to increasing dependence on groundwater in many countries. High levels of arsenic and fluoride are simultaneously present in groundwater at numerous places throughout the world. The cause of co-occurrence of these contaminants in groundwater can be natural as well as anthropogenic. Drinking water containing high concentrations of fluoride and arsenic primarily from natural geogenic contamination is the major source of human exposure (Gebel 1999; Ruiz-Payan et al. 2005). Arsenic and fluoride release into environments by various natural process and human activities (Bibi et al. 2015a; Vithanage and Bhattacharya 2015; Rasool et al. 2016b). Arsenic occurs in numerous forms depending upon pH and redox potential of groundwater. On the other hand, high fluoride concentrations present in groundwater from calcium (Ca) poor aquifers and may also increase in groundwater where cation exchange of sodium (Na) for calcium takes place (Bibi et al. 2015b; Amna Ali et al. 2015; Ali et al. 2016).

There is knowledge on the toxicity of fluoride and arsenic individually; however, very limited literature is available about the effects of combined exposure to these toxicants. An in situ experiment showed that Swiss albino male mice exposure to arsenic and fluoride led to a

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substantial depletion of blood δ -aminolevulinic acid dehydratase (ALAD) activity and glutathione (GSH) level (Mittal and Flora 2006). In another laboratory experiment, it was found that combined exposure to arsenic and fluoride considerably reduced the levels of brain biogenic amines (Flora and Mittal 2009). Arsenic and fluoride act as an unseen exterminates in water as they do not impart any odour, colour or taste. The present study provides a comprehensive overview about arsenic and fluoride contamination, toxicity, sources and existing removal technologies that are being used for the treatment. Moreover, problem associated with current methods and future research directions for removal technologies have also been discussed. This study will be very helpful to implement suitable and cost-effective removal technology for safe water supply to rural population in developing countries.

Arsenic contamination

Arsenic has been reported worldwide including Argentina (Smedley and Kinniburgh 2002), Australia (Appleyard et al. 2006), Bolivia (Archer et al. 2005), Chile (Romero et al. 2003), Finland (Kurttio et al. 1999), Greece (Kouras et al. 2007), Germany (Mkandawire and Dudel 2005), Ghana (Asante et al. 2007), Hungary (Rowland et al. 2011), Italy (Angelone et al. 2009), Mexico (Camacho et al. 2011), Romania (Rowland et al. 2011), Spain (García-Sánchez et al. 2005), Thailand (Williams et al. 1996) and USA (Schreiber et al. 2000). However, the arsenic-contaminated water via natural sources in various regions of East and South Asia is of bigger apprehension and affects the health of millions of people (Berg et al. 2001; Polyá et al. 2005; Mukherjee et al. 2006). In East Asia it includes Cambodia (Luu et al. 2009), China (Xie et al. 2009), Mongolia (Guo et al. 2008), Taiwan (Liao et al. 2011) and Vietnam (Berg et al. 2001); and in South Asia Bangladesh (Chen et al. 2011), India (Kar et al. 2010), Nepal (Pokhrel et al. 2009) and Pakistan (Baig et al. 2012; Rasool et al. 2016a, b). Arsenic-contaminated groundwater affects the life of nearly 60 million people while arsenicosis has been reported among 700,000 people (Fewtrell et al. 2005). The predominant form of inorganic arsenic in aqueous oxic environments is arsenate [As(V) as H_3AsO_4 , $\text{H}_2\text{AsO}_4^{-1}$, HAsO_4^{-2} and AsO_4^{-3}], whereas arsenite [As(III) as H_3AsO_3 and $\text{H}_2\text{AsO}_3^{-}$] is more prevalent in anoxic environments (Oremland and Stolz 2003). Zero-valent (As^0) and tri-valent (As^{3-}) are rare in aquatic environments (Goldberg and Johnston 2001; Mandal and Suzuki 2002).

Sources of arsenic

The sources of arsenic contamination in water could be geogenic such as occurrence of arsenic in geological

formations in the form of arsenic-bearing rocks and minerals as well as human-induced, i.e. agricultural and industrial, activities. The details are discussed below.

Geogenic source

Arsenic is present as a chief constituent of more than 200 minerals, together with elemental arsenic, sulphides, arsenides, oxides, arsenites and arsenates (Smedley and Kinniburgh 2002). Rittle et al. (1995) reported authigenic arsenopyrite in sediments, and microbial precipitation is responsible for the formation of orpiment (Newman et al. 1998). Many sources of arsenic contamination that are present naturally include geothermal, biogeochemical and geohydrological disbanding of arsenic compounds from arsenopyrite (Mondal et al. 2006).

Anthropogenic sources

Anthropogenic sources of arsenic contamination include processing of contaminated ores (Leist et al. 2000), ingredient of pesticides (McArthur et al. 2001), feed additives (Mondal et al. 2006), hazardous waste site (seepage) (Mondal et al. 2006) and arsenic-contaminated coal burning (McNeill and Edwards 1997; Abdullah et al. 2015). Geothermal waste, mining and industrial discharge are the main source of river pollution which is a chief drinking water source for large number of people (Smedley and Kinniburgh 2002). In California, acid seepage from Richmond mine located at Iron Mountain reported $850,000 \text{ g}^{-1}$ of arsenic with highest concentration reported so far (Nordstrom and Alpers 1999).

Mobilization

Arsenic concentration in groundwater is dependent on several factors like depth of well, characteristic and nature of aquifers whether confined or unconfined. Sahoo and Kim (2013) proved the fact that soils contaminated with arsenate and higher levels of silicate or phosphate may increase the mobility of arsenic(V) in subsurface (Sahoo and Kim 2013). In aqueous phase, mobilization of arsenic has been affected by differential adsorptive affinities of arsenite and arsenate to various mineral surfaces (i.e. alumina, ferrihydrite; Mondal et al. 2006). Various chemical, physical and microbial processes initiate the subsurface arsenic mobilization (Islam et al. 2004). Theories proposed to justify this mobilization include: (1) oxidation of arsenic-containing pyrites (Bhattacharjee et al. 2005), (2) iron oxide reduction release of arsenic(V) (Oremland and Stolz 2003), (3) reduction of iron oxides by allochthonous organic matter (from dissolved organics in recharging waters; Santini et al. 2002), (4) exchange of adsorbed

arsenic(V) with fertilizer phosphates (Larios et al. 2013), (5) reduction by microbial oxidation of sedimentary carbon (Stüben et al. 2003), (6) displacement of arsenic by carbonate (Anawar et al. 2004).

Mobility of arsenic in the environment is a defining factor that determines the risk of exposure (Frankenberger and Arshad 2002). Arsenic exists most commonly in water as inorganic species arsenite (H_3AsO_3 , $\text{H}_2\text{AsO}_3^{1-}$, HAsO_3^{2-}) and arsenate (H_3AsO_4 , $\text{H}_2\text{AsO}_4^{1-}$, HAsO_4^{2-} , AsO_4^{3-}), which dominate in reducing environments and at oxidizing conditions (Zhang et al. 2008). Both arsenite and arsenate are anionic species, but at typical pH values of 6–9 arsenite remains predominantly neutral. The neutral oxidation state results in increased remedial difficulties (Mohan and Pittman 2007). Reduction process releases 10–13 times more arsenic than oxidation (Kanel et al. 2005). However, the competition with organic anions for sorption sites is the main process for solid-phase release of arsenic while redox reactions perform only negligible role.

Health risk of arsenic

Prolonged exposure to $0.04 \mu\text{g}^{-\text{kg}-\text{day}}$ or higher concentration of arsenic leads to gastrointestinal, haematological and other peripheral neuropathic effects (Brown and Ross 2002), while consumption of high concentration at once results in digestive system problems, i.e. stomach pain or vomiting, which lead to coma or death. Chronic poisoning with arsenic results in hypertension, cardiovascular and lung diseases, diabetes and different type of cancers (i.e. lungs, skin, liver, kidney, uterus Escobar et al. 2016). Drinking water contaminated with even low concentrations of $5\text{--}10 \mu\text{g L}^{-1}$ can cause depigmentation, skin lesion and hyperkeratosis (Duarte et al. 2011). Brown and Ross (2002) investigated that chronic arsenic dose of $0.04 \mu\text{g}^{-\text{kg}-\text{day}}$ or higher for 6 months to 3 years or slow dose of $0.01 \mu\text{g}^{-\text{kg}-\text{day}}$ or higher for 5–15 years can cause hyperpigmentation. In 2001, United State Environmental Protection Agency (USEPA) replaced the $50 \mu\text{g L}^{-1}$ old standard with $10 \mu\text{g L}^{-1}$, while Pakistan Environmental Protection Agency (PAKEPA) still uses the old standard of $50 \mu\text{g L}^{-1}$.

Removal of arsenic from water

Selection of removal technique depends on arsenic speciation, physio-chemical properties of water and concentration of SO_4^{2-} , PO_4^{3-} and Fe, etc. All methods are subjected on fundamental chemical processes that are applied disjointedly, concurrently or in series. All arsenic removal technologies require pH adjustment for optimized performance (Bissen and Frimmel 2003). Removal processes like sorption are predominantly sensitive to pH and function well at low pH (Wang et al. 2004). Arsenic removal is

more efficient when it is present in pentavalent state as oxianions, i.e. H_2AsO_4^- and HAsO_4^{2-} , at a pH range between 2 and 12 under oxidizing conditions, while the trivalent arsenic (H_3AsO_3) can be found at pH below 9.2, under reducing conditions (Wang et al. 2004). Due to this fact, many arsenic removal methods use oxidation step prior to other processes; however, this step is facilitated with other physical or chemical modifications for efficient arsenic removal. Diverse arsenic removal technologies are generally based on six principles: (1) oxidation and filtration, (2) co-precipitation: oxidation of arsenic(III) to arsenic(V) and then arsenic(V) removal by coagulation, sedimentation and filtration, (3) biological oxidation: microbial oxidation of arsenic(III) to arsenic(V) followed by removal through iron and manganese oxides, (4) adsorption, (5) ion exchange (by cation and anion exchange resins) and (6) membrane technology. For optimized performance, various arsenic treatment technologies require pH adjustment as it directly affects arsenic speciation in raw water (Bissen and Frimmel 2003). Removal processes like sorption are predominantly sensitive to pH and function well at low pH (Wang et al. 2004) because for most media types, a pH greater than 8.5 decreases the adsorption capacity of the media and consequently increases the replacement cost of single-use media.

Pre-treatment

A wide range of chemicals (chlorine, ozone bleaching powder, hydrogen peroxide or potassium permanganate) are used to oxidize arsenic(III) to arsenic(V) in different conditions. However, use of these chemicals can lead to undesirable by-products (Gallard and von Gunten 2002). Ultraviolet light was also not effective, because 7000 times the ultraviolet dose required for *Escherichia Coli* inactivation oxidized only 73 % arsenic(III) (Clifford and Ghurye 2002). Ultraviolet photooxidation shows promising conversion of arsenic(III) when water is spiked with sulphite (Clifford and Ghurye 2002). Dodd et al. (2006) studied arsenic(III) oxidation kinetics through chloramines, ozone and aqueous chlorine relation for drinking water treatment. In addition to chemical oxidation, arsenic oxidative process has also been catalysed by some species of bacteria that were applied in groundwater (Jain and Singh 2012). This technique can be applied generally under reducing aquifer conditions holding iron and manganese (Jain and Singh 2012). Iron- and manganese-oxidizing bacteria can transform arsenic(III) to arsenic(V) which is naturally present in groundwater. Arsenic can be removed by adsorption and co-precipitation. Prior to adsorption, soluble iron and arsenic(III) are oxidized and then arsenic(V) adsorbs onto iron hydroxide precipitates that are eventually filtered out of solution. Arsenic removal

efficiency is determined by iron and arsenic ratio and preliminary iron concentration. In some cases, ferric coagulant is added initially for efficient arsenic removal, and this process is independent of pH in the range of 5.5–8.5 (Violante and Pigna 2009). Furthermore, at pH 4.0 more than in neutral or alkaline systems the formation of aluminium arsenate precipitates seemed to be favoured (Violante and Ricciardella 2006). However, high levels of orthophosphates, silicates and organic matter weaken the process as it increases the competition for sorption sites (Fields et al. 2000a, b). Numerous studies also described arsenic sorption on manganese oxides and chemical oxidation (Tournassat et al. 2002). Hug et al. (2001) investigated solar oxidation and developed a simple procedure for arsenic removal at neutral pH through locally accessible resources, without chemicals addition and pH adjustment, in order to cater with arsenic problem in Bangladesh drinking water. Furthermore, the formation of iron arsenate precipitates onto the surfaces of iron oxides seems to be favoured at low pH values (Violante and Gaudio 2007).

Reduction in arsenic concentration has also been observed due to water oxidation all through collection and storage in houses (Ahmed 2001). Investigation carried out in Bangladesh demonstrates that arsenic removal and sedimentation depend upon precipitating iron (Ahmed 2001). Study showed that arsenic content reduced by more than half through sedimentation of 380–480 $\mu\text{g L}^{-1}$ arsenic contaminated well water with CaCO_3 and 8–12 $\mu\text{g L}^{-1}$ with iron but reduction up to desired level was not achieved (Ahmed 2001). Different studies reported 0–25 % reduction in arsenic concentration via sedimentation process. Generally, passive sedimentation has been suggested as abortive approach as it failed to achieve desired arsenic level (Kinniburgh and Kosmus 2002).

Biological removal

Microbial sorption, oxidation–reduction, complexation, co-precipitation and methylation–demethylation can reduce or mobilize arsenic contamination. Ex situ or in situ bioadsorption can be used; i.e. use of biofilm of either live or dead microbes (Kamran et al. 2014; bacteria, algae, aquatic macrophytes and biopolymers) for adsorption or co-precipitation of contaminants also ascertained to be useful (Hartley et al. 2009; Kamran et al. 2015). Iron bacteria facilitate both arsenic(III) and arsenic(V) adsorption and precipitation onto the biological flocks (Hartley et al. 2009; Wang and Zhao 2009). Various natural low-cost biological materials, i.e. sedges, waste biomass milled bones, sorghum and cellulose, have been investigated for remediation (Kamran et al. 2016a). Teixeira and Ciminelli (2005) investigated chicken feather biomass as adsorptive media due to high protein contents. Selected arsenic(III)

adsorption at the rate of 270 $\mu\text{mol arsenic(III)/g}$ of biomass took place at relatively low pH (Teixeira and Ciminelli 2005). Natural biogenic hydroxyapatite (HAPb) acquired from charred cow bones evidenced as favourable material for arsenic(V) removal from water in specific conditions (1000 $\mu\text{g L}^{-1}$, 5 g L^{-1} adsorbent, circumneutral pH, 24-h contact time) (Czerniczyniec et al. 2007). Phytoremediation is another emerging technique using plants to remove contaminants from water. Huang et al. (2004, 2015a, b) and Kamran et al. (2016b) studied arsenic uptake by two hydroponically cultured arsenic hyperaccumulating plants (*Pteris vittata* and *Pteris cretica* cv. *Mayii*) to remove arsenic (20–500 $\mu\text{g L}^{-1}$). Results show that *P. vittata* reduced arsenic concentration from 20 to 0.4 $\mu\text{g L}^{-1}$ and from 200 to 2.8 $\mu\text{g L}^{-1}$ in 24 h. Jasrotia et al. (2015) studied the potential of aquatic plant species for phytoremediation of arsenic. Water hyacinth (*Eichhornia crassipes*) and two algae (*Chlorodesmis* sp. and *Cladophora* sp.) *Cladophora* sp. was perceived to tolerate up to 6 mg L^{-1} of arsenic, whereas water hyacinth and *Chlorodesmis* sp. could persist under 2 and 4 mg L^{-1} arsenic, respectively. It was found that *Cladophora* sp. can reduce arsenic concentration from 6 to 0.1 mg L^{-1} , while water hyacinth reduced only 20 % of arsenic. Thus, *Cladophora* sp. was proposed to be the best option for coinciding treatment of both arsenic-enriched sewage and brine.

A phytoremediation study was conducted to evaluate the arsenic uptake potential of two Cyperaceae species, *Schoenoplectus americanus* and *Eleocharis macrostachya*, collected near the towns of Chihuahua State, Mexico (Bundschuh et al. 2010). It was found that both species are able to survive at high arsenic levels and can be used for rhizofiltration because 97 % of the plants tolerate the arsenic with no visible effect on plant growth. Bundschuh et al. (2007) investigated dried macroalgae (*Spyrogira* spp.) for arsenic removal from acid mine drainage, and 80–90 % removal was accomplished within 4 days. Aquatic plant species, such as species *Ranunculus trichophyllus*, *Ranunculus peltatus* spp. *saniculifolius*, *L. minor* and *Azolla caroliniana*, and the leaves of *Juncus effusus*, also had very high potential for arsenic phytoremediation when they were introduced into constructed treatment wetlands or natural water bodies (Parmar and Singh 2015).

Another relatively emerging method is the biological oxidation of manganese and iron as a treatment method for arsenic removal. Groundwater contaminated with arsenic is mostly under reducing conditions and contains iron and manganese. Based on this fact the filters for removal of iron and manganese are populated with iron- and manganese-oxidizing bacteria, which can in turn oxidize arsenic(III) efficiently. Currently, the role of microbial processes in mineral oxide formation and removal of

arsenic during water treatment is unknown. The formation of manganese(III/IV) oxides contributes to abiotic arsenic(III) oxidation and immobilization of arsenic(V) by sorption to ferric(III) oxides (Nitzsche et al. 2015a, b). Biological iron and manganese oxidation has also been applied for groundwater removal of arsenic without any oxidizing agents (Zouboulis and Katsoyiannis 2005). Rate of bacterial oxidation of arsenic(III) to arsenic(V) is considerably higher than manganese oxide-added abiotic oxidation (Katsoyiannis et al. 2004). Therefore, bacteria play vital role in arsenic(III) oxidation and reactive manganese oxide surfaces generation for dissolved arsenic(III) and arsenic(V) removal.

Co-precipitation

Co-precipitation has been widely practised in various pilot-scale applications to treat arsenic-contaminated water. Three processes are normally used for chemical precipitation (Chwirka et al. 2000; Sancha 2006): (1) lime softening, (2) gravity coagulation–filtration and (3) microfiltration. Lime softening solely for arsenic removal has limitations as it is not economical or cost-effective. However, lime softening when accomplished for removing hardness, this process can be further enhanced to remove arsenic. Additional lime is added in order to increase the pH above 10.5 and to remove arsenic. In this pH range magnesium hydroxide precipitates and arsenic can be removed by co-precipitation. However, arsenic co-precipitation by calcium carbonate (i.e. pH <0.5) is not very efficient (removal efficiency <10 %; Fields et al. 2000b).

Coagulation

The coagulation–filtration process is rapidly used technique using iron(III) for arsenic(V) removal in water treatment process on pilot scale. For effective arsenic removal, the coexisting arsenic(III) is commonly converted to arsenic(V) before removal. Research studies show more efficient removal of arsenic(III) with higher initial concentrations; however, no significant removal efficiency was observed for initial arsenic(III) concentrations in range of 10–100 $\mu\text{g L}^{-1}$. The process using iron(III) can safely meet the permissible limit of 10 $\mu\text{g L}^{-1}$, only when the initial arsenic(III) concentration is less than 25 $\mu\text{g L}^{-1}$ and the iron(III) amount more than 5 mg L^{-1} . The efficient removal of arsenic(III) is limited due to the fact that arsenic(III) is usually present in uncharged H_3AsO_3 form under neutral pH conditions which is not adsorbed on iron oxy-hydroxides (FeOOH); the product of iron(III) hydrolysis (Ouzounis et al. 2015).

Coagulation–filtration includes the adsorption of arsenic to iron hydroxide. The process can trap arsenic(V) by

particle agglomeration. However, pre-oxidation is required due to arsenic(III) removal efficiency and neutral charge at natural pH conditions (Pokhrel et al. 2005). Generally, water characteristics determine the efficiency and suitability of the system, i.e. natural organic matter, dosage and type of coagulant, pH and mixing intensity (Pokhrel et al. 2005). In general, under optimized conditions, coagulation–filtration systems can achieve over 90 % arsenic(V) removal and produce water with less than 5 $\mu\text{g L}^{-1}$ of arsenic(V). Research shows that iron-based coagulants, i.e. ferric sulphate and ferric chloride, are more efficient in removing arsenic(V) as compared to aluminium-based coagulants (Pokhrel et al. 2005). Coagulation-based microfiltration uses the same coagulation process with modification of the granular media filtration step. However, the membrane needs to be backwashed periodically in order to remove solids and restore its hydraulic capacity (Thirunavukkarasu et al. 2003). New methods to remediate arsenic-contaminated water continue to be studied, particularly to fill the need for accessible methods that can significantly impact developing communities. A combination of cactus mucilage and ferric(Fe(III)) salt was investigated as a flocculation–coagulation system to remove arsenic(As) from water. Arsenic(V) solutions, ferric nitrate and mucilage suspensions were mixed and left to stand for various periods of time. At neutral pH, removal was dependent on iron(III) and mucilage concentration, and the age of the iron(III) solution. It was found that arsenic removal and settling rates were pH dependent; arsenic removal was between 52 (high pH) and 66 % (low pH) (Fox et al. 2016). Methylated arsenic is present everywhere in earth system (Hu et al. 2015a, b). So far, however, little information has been collected regarding their removal by coagulation. Methyl substitution during coagulation process has been used for monomethyl and dimethyl arsenate from drinking water (Hu et al. 2015a, b). By increasing the methyl group, negatively charged arsenic species decreased, which decreased the removal of methylated arsenic by coagulation. Hydroxide flocks adsorption was the main mechanism in coagulation. Additionally, traditional oxidants use and aid of coagulation revealed inadequate assistance for refining coagulation removal of dimethyl arsenate (Hu et al. 2015a, b).

Ion exchange

Ion exchange is physio-chemical method where exchange of ions took place between a solid resin and solution phase. The solid resin is typically a network of three-dimensional hydrocarbons holding many electrostatically bound ionizable groups. In treatment of drinking water, this technique is mostly applied for removing hardness and nitrates. This method can typically decrease arsenic concentrations from

50 to 10 $\mu\text{g L}^{-1}$ (Nguyen et al. 2009). However, its efficiency depends on the characteristics of untreated water and the contaminants. However, ion exchange is less common than precipitation–co-precipitation technology (Nguyen et al. 2009).

Zirconium oxide nanoparticles (HAIX–Zr) impregnated with hybrid anion exchange resins demonstrate high arsenic removal capacity. Non-hazardous and easy-to-transport, pre-calcined zirconium oxide has been used for the synthesis of hydrous zirconium oxide and tested for effective removal of arsenic(V) and arsenic(III). Over several cycles of exhaustion and regeneration, phosphate and silica competed for arsenic adsorption. Due to high regeneration ability of hydrous zirconium oxide nanoparticles (greater 90 %), hybrid anion exchange resins became more sustainable option for regeneration and reuse of hydrous zirconium oxide nanoparticles for effective reduction of arsenic concentration for numerous cycles. Dissimilar to other iron- or aluminium-based adsorbents, hydrous zirconium oxide nanoparticles are chemically stable at landfill conditions and could be disposed safely without leaching arsenic (Padungthon et al. 2015).

Membrane technology

Membrane technology eliminates an extensive variety of pollutants from water. But it also yields a higher volume of residues and is considered more costly as compared to other treatment technologies. Thus, adsorption precipitation–co-precipitation and ion exchange are more commonly used. Four chief membrane techniques include: reverse osmosis, microfiltration, ultrafiltration and nanofiltration. All the afore-mentioned processes are derived by pressure and depend on the size of particles to be passed through the membranes or by the pore size of membrane (Nguyen et al. 2009). The pore size determines the force required to drive fluid across the membrane; reverse osmosis and nanofiltration require higher pressure (50–150 psi), while ultrafiltration and microfiltration require relatively low pressure (5–100 psi) (Nguyen et al. 2009). Since arsenic species dissolved in water tend to have relatively low molecular weights, only nanofiltration and reverse osmosis membrane processes are likely to effectively treat dissolved arsenic (Nguyen et al. 2009). Reverse osmosis and nanofiltration primarily remove arsenic by categorization of size. A semipermeable membrane when exposed to a pressure gradient permits water to pass while retaining specific ions. Reverse osmosis membranes require higher driving pressures and are more selective than nanofiltration membranes. Arsenic rejection in reverse osmosis and nanofiltration is not sensitive to pH except that arsenic(III) is more efficiently rejected at pH greater 8 (Twidwell et al. 2005) because it is uncharged at low pH

but anionic at higher pH. Jain and Singh (2012) investigated removal efficiency of arsenic using nanofiltration membranes in China by varying initial arsenic concentration, pH, natural organic matter and other ionic compounds. Results show that nanofiltration point-of-use (POU) systems were most suitable to treat arsenic-enriched groundwater in areas of suburban China. Effective arsenic(III) removal by an oxalic acid complex is revealed by hybrid distillation system of forward osmosis membrane. It is very efficient process as it draws solute along with high water fluxes and insignificant reverse fluxes in forward osmosis to determine the factors involved in arsenic(III) removal. Comparatively high water fluxes (28 LMH) for forward osmosis mode and under the pressure lagging osmosis was achieved by using 1000 ppm arsenic(III) solution as the feed at 60 °C. Removal of arsenic(III) up to 21.6 % and water recovery (forward osmosis mode) 48.3 % (pressure restarted osmosis mode) were also achieved in 2 h (Ge et al. 2016).

Adsorption technology

Adsorption technology has been extensively used to treat arsenic-contaminated groundwater. The technology can minimize arsenic concentrations to $<10 \mu\text{g L}^{-1}$. Its efficiency is affected by a variety of untreated water characteristics and contaminants. The media used for adsorption is usually column packed. The contaminants get adsorbed as the contaminated water is passed through the column. When adsorption sites reach the threshold, the column must be regenerated or replaced with new media. Most commonly used adsorption media for arsenic removal includes activated alumina (AA), iron-coated sand, granular ferric hydroxide, indigenous cartridges and filters and other mixed adsorbents. The effectiveness of sportive media is dependent on the consumption of oxidizing agent that helps to adsorb arsenic.

Activated alumina

Activated alumina (AA) was the first adsorptive medium that was successfully used for arsenic removal from drinking water supplies (Ungureanu et al. 2015). It is a granular, porous material possessing good properties of sorption. Activated alumina grains have a high surface area for sorption and a usual diameter of 0.3–0.6 mm. It removes arsenic through microbial oxidation of arsenic(III) following a chemical adsorption step. A mixed culture of heterotrophic bacteria with high arsenic(III) oxidizing activity was obtained by acclimation to arsenic(III) from a soil sample that was free from contamination. The resultant mixed culture contained some genera of heterotrophic arsenic(III)-oxidizing and arsenic-tolerant bacteria:

Haemophilus, *Bacillus* and *Micrococcus*. The isotherms of arsenic(III) and arsenic(V) adsorption on activated alumina indicate that bacterial oxidation of arsenic(III) to arsenic(V) proves to be a significant pre-treatment process for arsenic removal (Baig et al. 2015; Ike et al. 2008).

Industrial based adsorbents (IBS)

Adsorption on industrial based adsorbents is a promising technology for arsenic removal. Zero-valent iron, granular ferric hydroxide, and modified iron, iron-coated sand and iron oxide are commercially available as adsorbents. Main driving force behind this process is chemisorption which is an irreversible process (Emdadul et al. 2016). Iron-based sorbents are considered a promising solution for arsenic removal as compared to the performance of activated alumina (Hussam and Munir 2007). Cornejo et al. (2008) have proposed a method based on zero-valent iron, solar radiation and lemon juice as adsorbent materials. By using 1.3 g L^{-1} of steel wool and one drop (ca. 0.04 mL) of lemon juice under solar radiation, arsenic removal attained up to 99.5 %. This method is very efficient and economical to treat arsenic-contaminated water. Ko et al. (2007) used sand coated with colloidal iron oxide for arsenic removal. Sylvester et al. (2007) used hybrid sorbent for arsenic adsorption by using nanoparticles of hydrous iron oxide while Chen et al. (2007) studied iron-impregnated activated carbons and found it very effective for arsenic removal. Iron oxide-coated *Aspergillus niger* biomass has also been used for arsenic removal (Pokhrel and Viraraghavan 2008). Thermodynamic study of the process demonstrated a spontaneous arsenic sorption on biomass by chemisorption process. Martin et al. (2007) synthesized iron(III) salt of mobilized ligands Octolig-21 which is available commercially, and it was found effective for arsenic removal from aqueous solutions.

Indigenous material

Various adsorbents are available for arsenic adsorption that have been developed from indigenous material. Iron ore, oxidizing iron, clay and cellulose-containing red soil have great capacity to adsorb arsenic, and some filters have been developed based on these materials, i.e. cellulose filters (Mandal 2015). A three-pitcher filter of brick chips and sand as filtering media was developed in Bangladesh, and baseline data collected after 1, 6 and 12 months demonstrate that due to inadequate maintenance this technology is only a short-term measure (Milton et al. 2007).

In recent years, various different adsorbents have been developed for arsenic removal. Hlavay and Polya (2005) characterized iron hydroxide-coated alumina for the treatment of arsenic-contaminated water. Hydrous iron oxide

nanoparticles (Sylvester et al. 2007), chitosan (Chen and Chung 2006), modified fungal biomass (Pokhrel and Viraraghavan 2006), iron oxide minerals (Öztel et al. 2015a, b), activated neutralized red mud (Tor et al. 2009), iron-containing mesoporous carbon (Baikousi et al. 2015), natural haematite, magnetite and goethite (Dai et al. 2016) and various other materials have been tested as promising materials for arsenic removal. Coating natural biopolymer, chitosan, on ceramic alumina using a dip coating process has also been developed as a novel biosorbent (Boddu et al. 2008).

Agricultural wastes

Agricultural wastes are by-products, typically underused or unused for animal feed. Agricultural by-products such as rice husks were used for removal of arsenic from water, and it was found that uptake of arsenic is directly proportional to temperature change (Malik et al. 2009). Different studies have used untreated rice husk for aqueous arsenic remediation (Amin et al. 2006 more references). Arsenic removal of both arsenic(III) and arsenic(V) using rice husk column was attained using initial concentration of arsenic 100 g L^{-1} with pH of 6.5 and 6.0, respectively. Blue Pine (*Pinus wallichiana*) wood shavings, walnut (*Juglans regia*) shell and chick pea testa were also used for arsenic remediation from aqueous solutions. Different conditions that affect the adsorption, such as pH, contact time, biosorbent dose, and temperature and adsorbate concentration, were studied. Blue Pine wood shavings proved a promising potential as a remediation material for arsenic removal from water samples. Walnut shell pieces also displayed good efficiency for biosorption (88 %; Saqib et al. 2013).

Geological materials

Geological materials are emergent remediation materials for house-level treatment in rural settlements, especially if the materials are locally available and can be collected by the population. Numerous natural iron- and aluminium-rich minerals such as haematite ($\alpha\text{-Fe}_2\text{O}_3$), goethite ($\alpha\text{-FeO(OH)}$), gibbsite (g-Al(OH)_3) and soils or sediments containing these minerals (e.g. oxisols, laterite), indigenous limestone (Soyatal), iron-coated zeolites, clay minerals (montmorillonite, bentonite) were tested in either laboratory prepared or natural waters, and recognized as substitute adsorbents for small water volumes (Alvarez-Silva et al. 2009; Armienta et al. 2009). Iron/manganese oxide-based materials, like “greensand” and other several natural minerals, have also been inspected (Mohan and Pittman 2007). Ordinary sand filters can also be a viable arsenic removal option from groundwater up to around $400 \text{ } \mu\text{g L}^{-1}$. This method has been ascertained very effective at household

level in Vietnam (Luzi et al. 2004). Recent technologies used for arsenic removal are given in Table 1.

Fluoride

Fluorine is the 13th most abundant element on earth. It cannot exist separately in environment without conjoining with other substances to become fluoride. The chief source of fluoride for human intake is contaminated groundwater by geological sources (maximum concentrations reaching 30–50 mg L⁻¹). Fluoride contamination level depends upon the nature of rocks and existence of fluoride-bearing minerals in groundwater. Fluoride concentrations in water are dependent on fluorite solubility in calcium-poor aquifers, and higher fluoride solubility should be expected in the groundwater. Fluoride is naturally present in soil, water, plants and animals in trace quantities. In groundwater, fluoride concentrations range from trace quantities to over 25 mg L⁻¹ naturally. When fluoride is ingested orally it is taken up by body tissues, with long-term deposition in teeth and bones. With the increasing industrialization, water bodies with excessive concentration of fluoride are becoming a matter of great concern.

Sources of fluoride in groundwater

Geogenic sources

Chief source of fluoride in the groundwater is fluoride-bearing rocks, and weathering or leaching from these rocks contaminates the groundwater reserves (Handa 1975; Hem 1985). Highly reactive fluorine is naturally found as calcium fluoride (CaF₂). It is an indispensable constituent in minerals like topaz, fluorite, fluorapatite, cryolite, phosphorite, theorapatite. Fluoride exhibits three forms, fluorospar or calcium fluoride (CaF₂), apatite or rock phosphate (Ca₃F (PO₄)₃) and cryolite (Na₃AlF₆). Fluoride concentration is five times more in granite than in basalt rocks. Likewise, higher concentration of fluoride is present in shale than sandstone and limestone. Higher concentration of fluoride is present in alkaline rocks (1200–8500 mg^{-kg}) (Dey et al. 2004). In groundwater low calcium and high bicarbonate alkalinity favours high fluoride concentration. Fluoride-contaminated water is soft with high pH and hold huge amount of silica. Depending upon different variables, groundwater fluoride concentration ranged from <1.0 mg L⁻¹ to more than 35.0 mg L⁻¹.

Anthropogenic sources

Anthropogenic sources of fluoride contamination in groundwater include: (1) agricultural field run-off and

infiltration due to use of fertilizers that contain high concentration of fluoride, (2) septic and sewage treatment system discharges, (3) seepage from industrial waste.

Health risk of fluoride

According to World Health Organisation report (WHO 2004) more than 200 million people around the globe rely on water containing fluoride concentrations that exceed the World Health Organisation guidelines of 1.5 mg L⁻¹. Depending upon the amount of fluoride in drinking water and extent of uptake, impact of fluoride uptake can be beneficial or detrimental to mankind. In drinking water fluoride has a narrow beneficial concentration range for human health. Small quantities usually have beneficial effect on rate and occurrence of dental caries, mainly among children (Mahramanlioglu et al. 2002). On the other hand, excess intake can lead to numerous diseases, i.e. osteoporosis, arthritis, brittle bones, cancer, infertility, brain damage, alzheimer syndrome and thyroid disorder (Harrison 2005). Fluorosis is a common symptom of high-fluoride ingestion revealed by teeth mottling in mild cases and bone deformities and neurological damage in severe cases (Fan et al. 2003). Some studies suggest that fluoride may hamper the deoxyribonucleic acid (DNA) synthesis (Zhou et al. 2004). High concentrations of fluoride can also affect with carbohydrates, lipids, proteins, vitamins and mineral metabolism (Islam and Patel 2011). Fluoride toxicity can take place by numerous ways. If ingested, fluoride firstly affects the intestinal mucosa, and later in stomach it can form hydrofluoric acid, followed by gastrointestinal irritation (Islam and Patel 2011). It can also affect numerous other enzymes disrupting oxidative phosphorylation, glycolysis, coagulation and neurotransmission (Islam and Patel 2011). Individuals with kidney ailment have a highest vulnerability to accumulative lethal effects of fluoride (Xiong et al. 2007). In addition, lethal dose of fluoride at once has been reported to disrupt kidney function over short-term exposures both in humans and in animals (Xiong et al. 2007). Fluoride can affect brain and pineal gland functions (Xiong et al. 2007). Fluoride mainly accumulates in pineal gland within the body, at a concentration higher than either teeth or bone (Xiong et al. 2007). Fluoride exposure can also cause bladder cancer, predominantly among the workers exposed to high concentration of fluoride in the workplace (Xiong et al. 2007).

Technologies for fluoride removal

The traditional fluoride removal technologies from drinking water include liming along with fluorite precipitation. The precipitation and coagulation processes with iron(III) (Tressaud 2006), activated alumina (Ghorai and Pant 2005)

Table 1 Recent advancements for arsenic removal from water

Adsorbent	Method/type of water	Uptake capacity (mg g ⁻¹)	Optimum temperature (°C)	Optimum pH	Sorbent dose (g L ⁻¹)	Contaminant concentration (mg L ⁻¹)	Contact time	References
Al-Fe (hydr)oxides	Water		25 °C	5.0–9.0	0.2 or 0.5 g L ⁻¹	1000 mg L ⁻¹	24 h	Qiao et al. (2014)
Thermostable enzyme ferritin from <i>Pyrococcus furiosus</i>	Aqueous solutions	12.3 ± 0.3	24	7.0	100 mM 3e	20 mL	24 h	Sevenco et al. (2015)
Water-soluble polymers	Aqueous solutions		22 ± 1	7	5–20 mg L ⁻¹	20 mL	24	Sánchez et al. (2015)
Iron oxide-coated pumice and sepiolite	Aqueous solution	0.579 and 0.309 mg g ⁻¹	22 ± 1 °C	7	2 g L ⁻¹	500 µg	84 h	Öztel et al. (2015a, b)
Continuous filter-press reactor	Groundwater/electrocoagulation	1.5 mg L ⁻¹	22 ± 1 °C	7.6		43 mg L ⁻¹	3 h	Guzmán et al. (2016a, b)
Nickel/nickel boride nanoparticles	Water	23.4 mg g ⁻¹	20 °C	3.3–11.5	10.0 g L ⁻¹	1000 mg L ⁻¹	24 h	Çiftçi and Henden (2015)
Polyaniline/Fe0 composite nanofibres	Aqueous solutions	232.6 mg g ⁻¹	20 °C	7.0	0.1 g L ⁻¹	1 mg L	24 h	Bhaumik et al. (2015)
Hydrated cement, marble powder (waste) and brick powder (waste)	Drinking water		25 ± 3 °C	8	10–40 g	100–1000 µg L ⁻¹	1.5 h	Bibi et al. (2015a, b)
Akaganeite decorated graphene oxide composite	Water	77.5 mg g ⁻¹	20 °C	3–10	3 mg	100 µg L ⁻¹	15 min	Chen et al. (2015)
Polyferric sulphate	Groundwater		20 °C	7.5–8.5	0.12 g L ⁻¹	500 µg L ⁻¹		Cui et al. (2015)
Chitosan/Cu(OH) ₂ and chitosan/CuO	Aqueous solution	39.0	20 ± 1 °C	1–12	50 mg	20 mL of 100 mg L ⁻¹	5 h	Elwakeel and Guibal (2015)
Nanoiron/oyster shell composites	Wastewater		20 °C	6.8	1.5 g L ⁻¹	1.8 mg L ⁻¹	24 h	Fan et al. (2015)
Goethite/parabuterite nanocomposites	Water	37.09 mg g ⁻¹	at 25 ± 2 °C	7	0.01 g	0.5–85 mg L ⁻¹	24 h	Fang et al. (2015)
Fe ₃ O ₄ -graphene macroscopic composites	Aqueous solutions		30 °C	4.0–10.0	10 mg	5 mL	24 h	Guo et al. (2015a, b)
Iron-impregnated biochar	Water	2.16 mg g ⁻¹	20 ± 2 °C	5.8 ± 0.2	0.1 g	0.1 and 55 mg L ⁻¹	24 h	Hu et al. (2015a, b)
Layered double hydroxides	Water	125.8 mg g ⁻¹	20 °C	7.0 ± 0.1	24 mg	30 mL	24 h	Huang et al. (2015a, b)
Layered double hydroxide compounds	Groundwater	14.6 mg g-LDH-1	20 °C	6.2 ± 0.3	25 mL	400 µg-As L ⁻¹	24 h	Jiang et al. (2015)
Ferrihydrite	Water	142.86 mg As(V)/g	25 °C	3.0–9.0	1.0 g	10–200 mg L ⁻¹	288 h	Jiang (2015)
Zero-valent iron (mZVI)	Groundwater		20 °C	5–9	0.15–2.5 g L ⁻¹	500 µg L ⁻¹	24 h	Katsoyiannis et al. (2015)

Table 1 continued

Adsorbent	Method/type of water	Uptake capacity (mg g ⁻¹)	Optimum temperature (°C)	Optimum pH	Sorbent dose (g L ⁻¹)	Contaminant concentration (mg L ⁻¹)	Contact time	References
Iron (Fe) ball and plate anodes	Groundwater		6.6–8.6			285 mg L ⁻¹	0–20 min	Kobya et al. (2015)
Acid mine drainage sludge (AMDS)	Aqueous solutions	18.25 and 4.97 mg g ⁻¹	25 °C	2–10	0.1 g	20 mL	168 h	Lee et al. (2015)
Titanium dioxide (TiO ₂) and granular ferric hydroxide (GFH)	Groundwater		21.9 °C	8.2	3 mg L ⁻¹	200 µg L ⁻¹		Lescano et al. (2015)
Aluminium hydroxide (Al(OH) ₃)	Underground waters	53.6 m mol ⁻¹	20 °C	4–9	1.77:1 mol mol ⁻¹	1.77:1 mol mol ⁻¹	24 h	Liu et al. (2015)
Denim fibre scraps	Aqueous solution	1.5 mg g ⁻¹	25 °C	7–9	10 g L ⁻¹	30 mg L ⁻¹	24 h	Mendoza-Castillo et al. (2015)
Chitosan-based electrospun nanofibre membrane (CS-ENM)	Aqueous solution		25 °C	3.3–11	0.5 g L ⁻¹	0.05–130 mg L ⁻¹	24 h	Min et al. (2015)
Iron oxide nanoparticles	Aqueous solutions		23 °C	7	150 mg g ⁻¹		1 h	Morillo et al. (2015)
Sand	Drinking water	115.1 ± 3.4 µg L ⁻¹	20 °C	6–8		115 µg L ⁻¹	312 h	Nitzsche et al. (2015a, b)
Fe(III) salts	Water		10–35 °C	7	20 mg L ⁻¹	25–100 µg L ⁻¹	2 h	Ouzounis et al. (2015)
Zr(IV) oxide	Groundwater		20 °C	7		1000 ppb	120 h	Padungthon et al. (2015)
Fe–Mn binary oxide-impregnated chitosan bead	Water		25 ± 1 °C	7.0 ± 0.1	1 g L ⁻¹	5–60 mg L ⁻¹	36 h	Qi et al. (2015)
Akhtenskite-coated waste goethite	Wastewater		20 °C	3.5	1e10 g	10 ppm		Shih et al. (2015)
Ceramic membrane materials	Water		110 °C			70 ppb	24 h	Sklari et al. (2015)
Fe(III) hydroxide	Water		25 °C	3.0–7.0	5 g L ⁻¹	100 mg L ⁻¹	5 h	Song et al. (2013)
Macroporous polymer impregnated with hydrous iron oxide	Water		25–45	2–12		0.078 mg dm ⁻³	1.5 h	Taleb et al. (2015)
Sands			20 °C		45 g	13–17 ppm	72 h	Trois and Cibati (2015)
Pine cone biochar	Aqueous solution		25 °C	2–12	10 g L ⁻¹ and 100 lg L ⁻¹	50–200 lg L ⁻¹		Van Vinh et al. (2015)
MIL-53(Fe)	Aqueous solutions	21.27 mg g ⁻¹	150 °C	6–10			16 h.	Vu et al. (2015)
Magnetic biochar from pinewood and natural haematite	Aqueous solutions		22 ± 0.5 °C	7	0.05 g	20 mg L ⁻¹	48 h	Wang et al. (2015b)

Table 1 continued

Adsorbent	Method/type of water	Uptake capacity (mg g ⁻¹)	Optimum temperature (°C)	Optimum pH	Sorbent dose (g L ⁻¹)	Contaminant concentration (mg L ⁻¹)	Contact time	References
Iron manganese bimetal oxides	Aqueous solutions		25 °C	3.0–12.0	0.2 g L ⁻¹	100 mL	24 h	Wen et al. (2015)
Hydrous cerium oxide-modified graphene (GNP-HCO)	Aqueous solution	62.33 and 41.31 mg As g ⁻¹	25 °C	4–7	0.1 g L ⁻¹	10 mg L ⁻¹	12 h	Yu et al. (2015)
Montmorillonites	Aqueous solution		25 °C	3–12	0.5 g	1–2500 mg L ⁻¹	24 h	Zehhaf et al. (2015)
HBC-Fe ₃ O ₄ -MnO ₂	Aqueous solutions		25 °C	7.0 ± 0.1	0.01 g	50 mL of 100–800 µg L ⁻¹	16 h	Baig et al. (2014)

and calcium (Yin et al. 2015a, b) have been studied extensively.

Ion exchange and coagulation

Ion exchange (Meenakshi and Viswanathan 2007; Popat et al. 1994) and reverse osmosis (Viswanathan and Meenakshi 2009) have been mainly used for fluoride removal from drinking water. Nevertheless, there are certain limitations for the application of these methods including high operational and maintenance costs, complex treatment procedure and generation of secondary pollution.

Aluminium chloride (AlCl₃) and polymer aluminium chloride (PACl) coagulation behaviour towards fluoride had been investigated at different basicity. Coagulants attained optimum fluoride removal above pH 6–7, while polymer aluminium chloride revealed higher removal efficiency as compared to aluminium chloride (AlCl₃) above pH 4–9. Fluoride removal by these coagulants increased with fluoride concentrations from 0 to 20 mg L⁻¹. At high concentration of 80 mg L⁻¹, removal efficiency of aluminium chloride decreases to as low as 13.1 % due to the formation of soluble aluminium fluoride (Al–F) complexes. On the other hand, this effect had not been showed by polymer aluminium chloride due to the stronger stability of aluminium species (Al₁₃ or Al₆). Polymer aluminium chloride with diverse aluminium species showed higher removal efficiency towards fluoride than aluminium chloride, especially at high fluoride concentration or under acidic and alkaline pH conditions (He et al. 2016a, b).

Toxic dissolved fluoride and calcium fluoride (CaF₂) nanoparticle pollution is a serious environmental concern for the semiconductor industry. Coagulation combined with electroflotation technique is applied to simultaneous removal of suspended matter and fluoride. Under optimum conditions, the solid–liquid separation efficiency is about 97 % in terms of turbidity removal which corresponds to a residual turbidity of 4.4 nephelometric turbidity units (NTU) complying with the standard limit (5 NTU), while fluoride efficiency removal may reach 73 % corresponding to 10 mg L⁻¹, which is below the environmental recommendations (Guzmán et al. 2016a, b). In general, coagulation methods are effective in defluoridation, but they are not successful in bringing fluoride concentration to desired levels (He et al. 2016a, b).

Membrane technologies

Membranes have advantage that they do not require additives, but their key disadvantage is that they are relatively expensive to install and operate. Moreover, they are prone to fouling, scaling or membrane degradation (Aoudj et al.

2015). In recent years, reverse osmosis membrane method has developed as an ideal substitute to provide safe drinking water without posing the complications related to other conventional methods. Efficiency of the process depends upon various factors, i.e. characteristics of raw water, temperature and systematic monitoring and maintenance. This method is highly effective for fluoride removal. Membranes act as strong impediment to suspended solids, inorganic and organic pollutants, pesticides, micropollutants and microorganisms (Maheshwari 2006a, b). But the key shortcoming of this technique is that it removes all the ions present in water. The process is competitively expensive than other methods (Maheshwari 2006a, b).

Nalgonda technique

In some developing countries, Nalgonda technique is commonly used for water defluoridation (Waghmare and Arfin 2015). In this technique alum, lime and bleaching powder added has been added to raw water followed by rapid mixing, flocculation, sedimentation, filtration and disinfection. Insoluble aluminium hydroxide flocks sediment along with co-precipitation of fluoride, while bleaching powder acts as disinfectant. However, high residual aluminium concentration ($2\text{--}7\text{ mg L}^{-1}$) in the treated water than the set World Health Organisation standard (0.2 mg L^{-1} ; Ayoob et al. 2008b; Maheshwari 2006a, b) is the main disadvantage of this process.

Adsorption

Among several processes for fluoride removal from water, adsorption offers most acceptable results and seems to be a most striking method for fluoride removal in terms of design simplicity, cost and operation (Mohapatra et al. 2009). Activated alumina has been used for years among the researchers for removal of fluoride (Craig et al. 2015). Farrah et al. (1987) studied aluminium hydroxide ($\text{Al}(\text{OH})_3$), aluminium oxide (Al_2O_3) for fluoride adsorption for pH range between 3 and 8. Results indicate that at pH less than 6 nearly all amorphous gel gets dissolved and aluminium fluoride (AlF) complexes were formed. The quantity of substrate transformed into aluminium fluoride improved with weakening pH and augmenting preliminary fluoride concentration. Ku and Chiou (2002) deliberated the influence of pH on adsorption of fluoride, and highest fluoride removal ($16.3\text{ }\mu\text{g}^{-\text{g}}$) was established which ranged between pH 5 and 7. On the basis of low activation energy values, conclusion was made that fluoride removal occurred due to non-specific adsorption. Activated alumina surface was modified by researchers in order to increase its efficiency. La(III) and Y(III) were impregnated on aluminium in order to alter alumina (Tokunaga et al. 1997). Comparisons were made

between impregnated alumina and original alumina under a variety of conditions. The removal efficiency of modified alumina was found highest for fluoride than phosphate, arsenate and selenite. Broad research has been conducted for defluoridation of water using diverse calcium salts because calcium has great association with fluoride. Turner et al. (2005) carried out defluoridation studies using calcite, and batch studies were performed. Using atomic force microscopy (AFM) and X-ray photoelectron spectroscopy (XPS) and other potential measurements confirmed that combined action of surface adsorption and precipitation reactions was responsible for fluoride adsorption from water and degree of adsorption was based on calcite surface area (Islam and Patel 2007). These adsorbents have extensively studied for defluoridation as iron had strong attraction for fluoride. Mostly adsorbents used for defluoridation need pH because they are unstable at extreme pH values. Thus, wastewater from polishing industries contains high fluoride and stands as a major environmental problem. Due to its magnetic properties and stability at lower pH, schwertmannite is being used for the defluoridation of polluted wastewater (Eskandarpour et al. 2008).

Nanotechnology

Nanotechnology is an emerging and promising technology in a variety of fields. Use of nanoparticles as adsorbents for the treatment of water has also gained broad consideration in past few years. Due to their smaller sizes, huge surface area, elevated mechanical strength and incredible electrical conductivities carbon nanotubes (CNTs) have gained enormous interest and became potential adsorbents. Li et al. (2001) applied carbon nanotubes as supportive media to deposit aluminium oxide (Al_2O_3) and discovered the prospect of carbon nanotubes for defluoridation of drinking water. Isotherms data indicated that best defluoridation results were achieved in 5–9 pH range. Defluoridation of water was performed using activated carbon nanotubes which were developed by xylene decomposition in the presence of ferrocene catalyst (Li et al. 2003). Wang et al. (2009) carried out defluoridation by using aluminium hydroxide (nanoscale in comparison with traditional methods makes these nanoparticles more suitable media). Zhao et al. (2010) combined the advantages of aluminium hydroxide ($\text{Al}(\text{OH})_3$) and magnetic nanoparticles to fabricate nanosized adsorbents with high surface area, high affinity towards fluoride and good magnetic separability, to develop a new kind of magnetic fluoride adsorbent.

Natural materials

Natural materials which are easily accessible in huge quantity are being investigated for F-contaminated water

(Biswas et al. 2010). Studies have also been carried out on fluoride removal with the help of low-quality coal (Borah and Dey 2009). Acid-modified raw laterite has also been evaluated for defluoridation of water (Maiti et al. 2011). Alteration of bentonite clay using lanthanum, magnesium and manganese was also carried out for its adsorption and defluoridation activity (Kamble et al. 2009). In order to achieve effective fluoride removal, low-cost bentonite clay was modified chemically by MgCl_2 (Thakre et al. 2010).

Biosorption

Various biosorbents have been developed for fluoride removal. Among a variety of biosorbents available for defluoridation, wide attention has been given to derivatives of chitin and chitosan because of their cost-effectiveness and higher concentration of $-\text{NH}-$ and OH functional groups they showed considerable adsorption capacity for different of water pollutants. These show significant adsorption potential for the removal of various aquatic pollutants. A comparison of adsorption efficiency for defluoridation was made between chitin, chitosan and 20 % La-chitosan adsorbents (Kamble et al. 2007). Fluoride uptake comparison was made between distilled water and contaminated actual water, and results indicate that application was high in distilled water than field water due to competing ions effect and high pH. Yao et al. (2009) have tested modified chitosan for defluoridation of water. Optimum pH for defluoridation was 7.0, and under acidic conditions chitosan was found unstable. Several plant-based biosorbents such as dried orange juice residue, tamarind (*Tamarindus indica*) fruit shell carbon, *Moringa indica*-based activated carbon have been studied very recently by various investigators and reported for fluoride removal from water with different degrees of success. Neem leaf powder (NLP) developed from the mature leaves of neem (*Azadirachta indica*) trees has also been shown to be an active biosorbent for removal of fluoride from water (Bharali and Bhattacharyya 2015a, b).

Waste materials

Industrialization produces enormous quantity of waste in the form of by-products. To transform these materials into cheap adsorbents will be the most appropriate use of these waste products in order to treat wastewater. A variety of treated and untreated industrial by-products are evaluated for defluoridation of water. The ability of fly ash (a thermal power plant waste) to remove fluoride from water and wastewaters was studied (Husain et al. 2015). The residue of industrial waste which is produced during process of alum in thermal power plants was evaluated by Chaturvedi et al. for defluoridation of water (Chaturvedi et al. 1990).

High fluoride removal efficiency was observed at high temperature and low fluoride concentration under acidic conditions. The highest removal efficiency was observed by using Langmuir isotherm of $20.3 \text{ mg}^{-\text{g}}$ (Chaturvedi et al. 1990). Marble powder, brick powder and hydrated cement had been studied for the removal of fluoride from drinking water (Bibi et al. 2015a, b).

Only few researchers have evaluated construction materials as adsorbents for the treatment of fluoride-contaminated water. Yadav et al. (2006) evaluated and compared brick powder for fluoride removal from water and compared with analytical grade activated charcoal. Removal % of brick powder was found 56.8 % for a pH range of 6.0–8.0. While for activated charcoal, removal efficiency decreased by increasing pH. Under optimum pH interaction between metal and fluoride ions takes place. Other competing ions did not interfere with the defluoridation process. By increasing the contact time from 15 to 20 min, the defluoridation efficiency increased up to 54.4 per cent by brick powder and up to 80 per cent with activated carbon. Comparison between two adsorbents revealed that brick powder had better adsorption efficiency and it is cost-effective. Concrete (gas), a material used for construction had also proved very effective adsorbent for defluoridation (Oguz 2005). Hydrated cement had also been tested as a potential adsorbent for defluoridation. The potential of hydrated cement (HC) for the removal of excess fluoride from aqueous solution has also been tested (Kagne et al. 2008). The kinetics of defluoridation efficiency from water by alumina cement granules (ALC) has also been examined (Ayoob et al. 2008a). A comparison of fluoride removal efficiency of ALC from synthetic water and natural ground water was also studied by Ayoob and Gupta (2009).

Agricultural waste

The use of agricultural waste products for treatment of water is very cost-effective option because of their easy availability and cost-effectiveness. Parmar et al. (2006) used aluminium- and calcium chloride (Al/CaCl_3)-treated corn cobs powder for defluoridation of water. Al/Ca -treated cobs showed excellent results. Mohan et al. (2008) used different agricultural materials including coconut shell, shell fibres and rice husk in order to remove pollutants, i.e. fluoride. Carbon of coconut shell proved best adsorbent. Sivabalan et al. (2003) studied fluoride adsorption by using palm seed coat charcoal. Adsorption process depends upon pH, and adsorption efficiencies were found higher in range of 4–8. Zirconium-treated coconut shell carbon was examined to treat water contaminated by fluoride (Sathish et al. 2007). Researchers have also investigated the potential of zirconium-impregnated cashew nut

Table 2 Recent advancements for fluoride removal from water

Adsorbent	Method/type of water	Uptake capacity (mg g ⁻¹)	Optimum temperature (°C)	PH	Sorption dose (g L ⁻¹)	Particle diameter	Contaminant concentration (mg L ⁻¹)	Contact time	References
Lanthanum-carbon	Drinking water	9.96	25	7 ± 0.2	5 to 30	<50 nm	1.0–80.0	1 h	Vences-Alvarez et al. (2015)
Lanthanum-loaded magnetic cationic hydrogel	Drinking water	136.78	25	7.0 ± 0.2	0.3	1 µm	29.3	2 h	Dong and Wang (2016)
Iron nanoimpregnated adsorbent	Water		20	7	2.5	40–50 nm	4	25.0 min	Ali et al. (2015)
Al(III)–Zr(IV) binary oxide adsorbent	Aqueous solutions/co-precipitation combined with calcination	114.54	45	2			50–150	4 h	Zhu et al. (2015)
Aluminium fumarate metal organic framework	Groundwater	600	20	7	0.75	17 Å (pore diameter)	30		Dechnik et al. (2016)
Mg–Fe–La trimetal composite	Adsorption/complexation and ion exchange interaction	112.17	25	7	0.5	31.16 nm	10–150	180 min	Wang et al. (2015a)
Sulphate-doped hydroxyapatite	Aqueous solutions	28.3	25	7	0.5	3–4 µm	2–100	24 h	Chen et al. (2016)
Bone char	Ionic exchange	5.92	30	7	2	2–50 nm	5–80	24 h	Rojas-Mayorga et al. (2015)
Neem (<i>Azadirachta indica</i>) leaf powder	Aqueous solutions/electrostatic interactions	4.7	30	5.0–7.0	1.0–5.0	10 µm	3.0–15.0	60 min	Bharali and Bhattacharyya (2015a, b)
Natural calcium-rich attapulgite	Contaminated water	140	25	8	0.5	0.2–0.5 mm	5–2000	24 h	Yin et al. (2015a, b)
Regenerated bone char	Drinking water	0.88				0.63–2 mm			Kanyora et al. (2015)
Pyrolysed delonix regia pod	Particle diffusion	107.15	60	2	1.5	32–36.5 µm	10 Feb	300 min	Ajisha and Rajagopal (2015)
Hydroxyapatite	Industrial wastewater	12.4	20	12.5	0.4–4		8.69 ± 1.84	4 h	Melidis (2015)
Regenerated aluminium oxide-coated media	Chemisorption	34.24	28	7 ± 0.05	12	0.8–1.2 mm	40–60	5 days	Buamah et al. (2016)
Tea waste loaded with Al/Fe oxides	Drinking water	18.52	25 ± 2	4.0–8.0	0.4–8.0	5 µm	5–200	120 min	Cai et al. (2015)
Zirconium-impregnated fibrous protein	Drinking water	12.6	25	5				6 h	Deng and Yu (2015)
Sorghum and	Aqueous solution		30	5	2–14		10–100	10–180 min	Zazouli et al. (2015)
Canola			30	5	2–14		10–100	10–180 min	Zazouli et al. (2015)

Table 2 continued

Adsorbent	Method/type of water	Uptake capacity (mg g ⁻¹)	Optimum temperature (°C)	PH	Sorption dose (g L ⁻¹)	Particle diameter	Contaminant concentration (mg L ⁻¹)	Contact time	References
Lamb and chicken bones	Chemical precipitation and ion exchange			6–7	15, 20	<0.075 mm	10	120 min	Ismail and AbdelKareem (2015)
Chemically activated cotton nut shell carbon	Drinking water		25	7	1.75	1 µm	2.0–10	180 min	Mariappan et al. (2015)
Alumina-based composite	Drinking water	17.57	25 ± 2	7.43	0.4	10 µm	5.3	18 h	Waghmare et al. (2015)
Magnetite-chitosan composite	Drinking water	9.43	50	7	0.25–2	3.4 nm	1.0–10	60 min	Mohseni-Bandpi et al. (2015)
Jajarm bauxite	Aqueous solution	0.59		7	15	<250 µm	2.3–2.7	90 min	Malakootian et al. (2015)
Cellulose nanofibres from waste pulp	Aqueous solution	10.6		6.4	16.7	10–100 nm			Sehaqui et al. (2016)
Ultrasonically prepared aluminium hydroxide	Aqueous solution	3.8	25		4	2.25 mm	>20	5 h	Gai et al. (2015)
Hydroxyapatite (HAP) nanowires	Anion exchange and electrostatic interactions	40.65	25	7	0.5	150 µm	200	12 h	He et al. (2015a, b)
MgO microspheres	Hydroxyl and carbonate coexchange mechanism	115.5	25	7.00	1		>20	12 h	Jin et al. (2015b)
Alumina-modified expanded graphite composite	Aqueous solution	1.18	30	4.00	0.2	10–30 nm	5	120 min	Jin et al. (2015a)
Mg–Al oxide	Aqueous solution		30					120 min	Kameda et al. (2015)
3D hierarchical amorphous aluminium oxide microspheres	Adsorption and diffusion	126.9		7		10 µm	5–150	600 min	Kang et al. (2015)
Mg-/Fe-layered double hydroxides	Aqueous solution	50.9		7			5–75		Kang et al. (2013)
Aluminium alginate beads	Aqueous solution	75.2	25	2	1	200 µm	1.9–190	4 h	Kayguzuz et al. (2015)
Chitosan beads	Complexation followed by an electrostatic interaction	17.47		7	1		>20	80 min	Prabhu and Meenakshi (2015)
Triethylamine-modified maize tassels	Aqueous solution	0.19		4	0.03		10–100	20 min	Mwangi et al. (2016)
Ce–Fe bimetal oxide adsorbent	Aqueous solution	60.97	20	2.9–10.1	0.3–1.5	1.68–9.54 nm	10		Tang and Zhang (2016)
Chitosan/montmorillonite/ZrO ₂ nanocomposite	Aqueous solution	23	30	4	0.1		20	60 min	Teimouri et al. (2015)

Table 2 continued

Adsorbent	Method/type of water	Uptake capacity (mg g ⁻¹)	Optimum temperature (°C)	PH	Sorption dose (g L ⁻¹)	Particle diameter	Contaminant concentration (mg L ⁻¹)	Contact time	References
Lanthanum-impregnated green sand	Aqueous solution	3.74		6 to 9	6	100 µm	15	240 min	Vivek Vardhan and Srimurali (2016)
Laterite and bone char									
Aluminium-impregnated coconut fibre	Synthetic fluoride solution and natural water	3.19	40	5	0.05	20 µm		40 min	Mondal et al. (2015)
Hydrothermally modified limestone powder	Aqueous solution	6.45		7.10–7.50	3.3	1 to 10 µm	>10	3 h	Gogoi and Dutta (2016)
Marble waste powder	Aqueous solution	1.2	30	7	8	572.2 nm	10	3 h	Mehta et al. (2016)
Al ₂ O ₃ /TiO ₂ nanocomposite	Aqueous solution	1.9			0.002	50 ± 6 nm and	239 ± 33 nm	2	30 min
Suriyaraj et al. (2015)									
Cerium-loaded mesoporous zirconium phosphate	Aqueous solution	20.5		6	0.01	3.7–6.2 nm	>50	1 h	Dash et al. (2015)
Fe ₃ O ₄ superparamagnetic nanoparticles with zirconium oxide	Aqueous solution	145.2	25	2.5	1	10 nm	20	1 h	Riahi et al. (2015)
Commercial activated carbons using fluoride ion selective electrode	Aqueous solution		45		10		40	360 min	Ravančić and Habuda-Stanić (2015)
Aquatic plant species Ipomoea aquatica	Industrial wastewater						5–20	10 days	Singh and Majumder (2015)
Industrial based adsorbent (cement)	Groundwater	1.72	25 ± 3	7	30	10 µm	30	60 min	Bibi et al. (2015a, b)

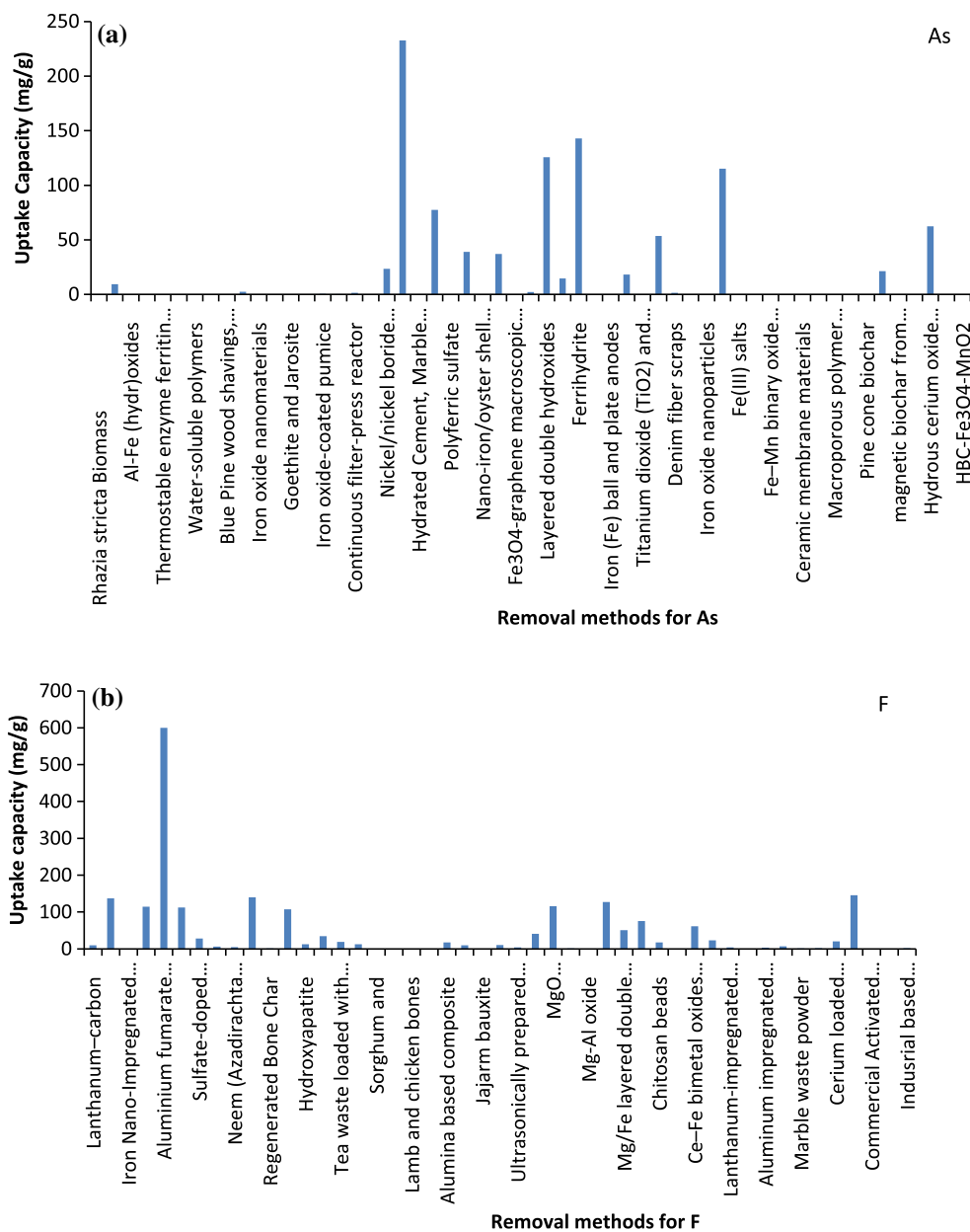


Fig. 1 Comparison of uptake capacities of different materials used during 2015–2016: **a** for As, **b** for F

shell carbon and compared its performance with that of cashew nut shell carbon for fluoride removal from aqueous solutions (Alagumuthu and Rajan 2010).

Six principles (biological oxidation, oxidation–filtration, co-precipitation, adsorption, ion exchange and membrane technology) are the basis of different technologies available for the removal of arsenic and fluoride from contaminated water. Every technique has its own merits and demerits. Typical treatment efficiencies for processes operated under normal conditions are provided in Tables 1 and 2. Figure 1 provides the comparison for different materials that have been used recently for arsenic and

fluoride removal, and their adsorption capacities have been compared. In case of arsenic adsorption capacities of nickel- and iron-based materials are much higher than other materials, while for fluoride, aluminium-based materials showed tremendous adsorption capacity towards fluoride.

Conclusion

Drinking water contamination by arsenic and fluoride is a problem just about anywhere in the world, predominantly in developing countries of Asia. These contaminants are

generally present in geological formations, so remediation technologies are the only option to diminish the effect. Traditional treatment processes such as coagulation–filtration, lime softening, iron/manganese oxidation and membrane filtration have been used in water treatment plants for the removal of these contaminants. Ion exchange, filtration and adsorption have been employed at domestic level. Advanced technologies, i.e. biological treatment, reactive membrane, phytoremediation, are also used for water treatment of arsenic contamination. Yet, various techniques are still at experimental stage and some have not been established at full scale. It is suggested that combination of ion exchange, filtration and adsorption as a low-cost chemical treatment with bioremediation could be advantageous for the decontamination of drinking water. Arsenic and fluoride alleviation approach should be adopted according to the specific geographic and morphological characteristics and socio-economic conditions of the area. All removal technologies discussed above have advantages and disadvantages, so suitable technology for specific situation should be opted. Technologies should be modified on pilot-scale implementation to effectively remove these contaminants by reducing operational and maintenance cost in a user-friendly manner. In many affected areas, arsenic and fluoride removal method could be the only alternative for safe water supply. So it is strongly recommended that efficient technologies that are found effective and safe should be indorsed for arsenic and fluoride removal in affected areas to evade ingestion of these toxins via drinking water.

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