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Arsenic Mitigation Technologies from Ground Water: A Brief Review

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ABSTRACT

Contamination of drinking water due to the presence of as has become a global environmental and socio-economic threat. The appearance of high Arsenic (As) in drinking water causes a serious health issue around the world. Many countries in different parts of the world have reported high arsenic concentrations. Among all groundwater arsenic contamination affected countries, the position of Bangladesh is the worst. Therefore, it is very important to develop affordable and efficient techniques to remove As from drinking water to protect human health. The most used methods are oxidation, coagulation, adsorption, ion exchange, and membrane technologies. Oxidation is usually used as pretreatment for most of the methods. Coagulation is the most common arsenic mitigation technology in Bangladesh. This technique is effective from pH 6-8. Ion exchange resins can only remove arsenate. Activated alumina beds work best in slightly acidic waters and usually have much longer run times than ion exchange resins. A cost-effective method for mitigation of As from drinking water is the use of low-cost adsorbent. Membrane methods which are more costly than other arsenic mitigation techniques but very effective where very low arsenic levels are required. Providing a safe water source may not possible in some of the arsenic affected regions or sometimes this process becomes very expensive. Mitigation of As from drinking water may be more appropriate in these situations. This paper presents a review of the conventional methods used for mitigation of As from contaminated drinking water.

Keywords: Arsenic, Bangladesh, Contamination, Arsenic poisoning, Human health, and Mitigation technologies.

INTRODUCTION

Arsenic is found in the soils, rocks, atmosphere, natural water sources and organisms (Asere *et al*., 2019). It is 20th in abundance in the earth's crust (Hossain, 2006; Singh *et al*., 2015). Arsenic is a steel-grey brittle crystalline "metalloid" and a natural constituent with atomic mass 74.9; specific gravity 5.73, melting point 817^{0}C (at 28 atm), boiling point 613^{0}C and vapour pressure 1mm Hg at 372 ^{0}C (Verma *et al*., 2014). Arsenic exists in environment in four oxidation states: arsine (III), arsenite (+III), arsenic (0), and arsenate (V) in inorganic forms and the organic forms include monomethylarsonic acid [MMA; $CH_3AsO(OH)_2$], dimethylarsinic acid [DMA; (CH_3) AsOOH], trimethylarsine oxide [TMAO; $(CH_3)_3AsO$], arsenobetaine [AsB; $(CH_3)_3AsCH_2COOH$], arseno-choline (AsC), arsenosugars (AsS), arsenolipids etc. In the inorganic forms, both As(III) and As(V) are toxic, both As(III) and As(V) exist within the pH range of 6-9 (Pinheiro *et al*., 2017) but As (III) is more toxic than that of As(V) (Abedin *et al*., 2002). As(V) is thermo-dynamically more stable in toxic (aerobic) waters and

As(III) in anoxic waters, But they could coexist in both types of waters (Shankar *et al.,* 2014). On the other hand, in case of organic forms the dimethylarsinous acid and monomethylarsonous acid are more toxic than original compounds (Petrick *et al*., 2000). Four types of As are usually available in groundwater of Bangladesh, these arearsenite (H_2AsO_3) and arsenate ($H_2AsO_4^-$) (Shankar *et al.,* 2014) and monomethylarsonic acid [$CH_3AsO(OH)_2$] as well as dimethylarsinic acid [$(CH_3)_2As(OH)$] (Abedin *et al*., 2002). At early age when knowledge on the toxicity of arsenicals was very poor, arsenic sulphide (realgar) was occasionally used, both as a medicine and as a poison in Asian civilizations (Abedin *et al*., 2002). Several As compounds were used as wood preservative, pesticides in agriculture and has also been taken as medicine to treat many diseases like malaria, asthma, leukemia, eczema, chorea, African trypanosomiasis, skin and breast cancers though these uses have become outdated in recent years (Shah, 2012). Arsenic concentration in fresh water represents a health problem, since this element is toxic and carcinogenic in small quantities (Asere *et al*., 2019).

Arsenic in water is invisible enemy because it does not possess a particular color or taste (Bhowmick *et al*., 2018). Ingestion of large doses of As could lead to gastrointestinal symptoms, disturbances of cardiovascular and nervous system functions and finally death (Wang *et al*., 2009). However, intestinal problems are caused because of consuming in determined amounts; long time consumption develops a range of serious diseases such as high blood pressure, skin discoloration, blood vessel diseases, cancer of skin, kidney and lung and reproductive disorders (Smith *et al*., 2000). The European Union (EU), The United States (US) and The World Health Organization (WHO) have established a value of 10 µg/L as the maximum contaminant level for total As in potable water (WHO, 2011). Due to presence of As in drinking water many countries have faced significant health problems such as arsenicosis which is responsible for various diseases in bladder, skin, kidney and lung. According to WHO 10 µg/l is set as the standard value for arsenic in water (WHO, 2011). But the concentration of arsenic has been found very high in many countries such as Bangladesh (Islam *et al*., 2017), India, Vietnam (Berg *et al*., 2007), China (Yang *et al*., 2012), Mexico (Armienta and Segovia, 2008), Pakistan (Muhammad *et al.,* 2010), Japan (Ahn, 2011), Korea, New Zealand, Hungary, USA (Haque and Johannesson, 2006), Argentina, Chile, El Salvador, Peru and Nicaragua (Jain and Singh, 2012) WHO reported that at least million people of 50 countries are exposed to arsenic through arsenic-contaminated groundwater at levels above 10 µg/L (WHO, 2011) Bangladesh was detected as one of the worst arsenic-affected countries in the world (Muhammad *et al.,* 2010) in terms of population exposure to arsenic-contaminated water when 400 measurements were carried out in 6 (Smith *et al*., 2000). In almost half of the measurements, arsenic concentrations were above the maximum permissible limit of 0.05 mg/L which is safe limit for drinking water in Bangladesh (WHO, 2011). Here about 50 million people being at risk of As exposure. Department of Public Health Engineering (DPHE), Bangladesh, detected four tube wells in Chamagram, Nawabganj that yielded arsenic-contaminated ground water, and in the next year, eight arsenicosis patients were found in the same village (BGS and DPHE 2001). Soon after the identification of arsenic in tube well water, Bangladesh government initiated a number of programs with support from the national and international non-government organizations (NGOs) to determine the extent of arsenic contamination problem (NAISU, 2002). Survey showed that 27% of the shallow groundwater aquifers have arsenic concentrations greater than 50 µg/L in Bangladesh (Khan *et al*., 2010).

The percentage seems remarkably low but it is a matter of thought that in Bangladesh, more than 90% of the rural population gets drinking water from tube wells (Islam *et al*., 2017). The south and east parts of Bangladesh is the high risk region; here more than 60% of the tube-wells contain arsenic over 1 mg/L. DPHE randomly checked about 23,000 tube-wells and they identified that the southeast Dhaka was the worst affected area. Almost 20% of the shallow tube-wells contain As exceeding 50 µg/L, which is more than the Bangladesh standard. 80% of the As-contaminated hand tube wells showed 50µg/L (Chakraborti *et al*., 2015). In Bangladesh the As concentration in the groundwater has been found in wide range (<0.5-

>4600 µg/L) (Chowdhury *et al*., 2018). Different technologies have been implemented for the remediation of arsenic levels in drinking water in many countries (Radloff *et al*., 2017).

The most commonly used conventional technologies include lime treatment, co-precipitation, coagulation-flocculation, oxidation, adsorption, ion exchange resin, membrane techniques, cementation, biological process and emerging technologies (Ahmed *et al*., 2006; Banerji and Chaudhari, 2016; Gallegos-Garcia *et al*., 2012; Iervolino *et al*., 2016; Meher *et al*., 2015; Yavuz *et al*., 2010; and Yazdani *et al*., 2016). There are almost 14 technologies worldwide for the mitigation of arsenic with efficiency varying from 70 % to 99%. One of the most common technologies have been coagulation with metal salts, lime softening, and iron/manganese mitigation. Coagulation processes are sometimes unable to efficiently remove arsenic to permissible levels. Membrane processes have high mitigation efficiency but these methods are highly expensive. In case of Ion exchange process high levels of total dissolved solids, sulphate, fluoride, and nitrate affect the method's efficiency (Lin *et al*. 2017).

Among all methods adsorption can be considered as a low-cost, simple, and eco-friendly process for Arsenic mitigation. But treatments of natural adsorbents are required to enhance the mitigation efficiency which is very expensive. The aims of the present review is giving a scenario of possible sources of As contamination of groundwater, associated health risks, available technologies for mitigation of As pollution in groundwater, discussion about prospects and limitations of different treatment processes and delineate the areas of further improvement.

Sources of Arsenic

As released from both geogenic and anthropogenic sources (Alarcón-Herrera *et al*., 2013). The nature of the aquifers and the process responsible for the As in groundwater has been evaluated in several studies. As is commonly found in several minerals, among these oxides and hydroxides of metals (Mn, Al, and Fe), sulfides, arsenides, and arsenites are major minerals (Nriagu *et al*., 2007). Arsenic pyrite is the one of most important mineral source in the ore zones responsible for geogenic contamination of As. The presence of pyrite in the reduced alluvial aquifers sediments in Bangladesh causes release of As into the environment due to the desorption and dissolution of naturally occurring As bearing minerals (Das *et al*., 2018). Pyrite is found to be stable under reducing conditions, in presence of aerobic conditions it oxidizes and release As due to formation of iron oxides (Patel *et al*. 2019). Iron hydroxide acts as a sorbent to increase the amount of dissolved As in groundwater. A number of studies have reported that organic matter enriching fine-grained shale and clay are highly enriched with As (Bayatkashkoli *et al*., 2017; Reza and Jean, 2012). However, several studies have suggested that iron oxide coated sand and grains of mica of the sediment are the potential adsorbent of As (Freikowski *et al*., 2013). In the hydro geochemical environment, Arsenic is released under oxidizing as well as reducing conditions, the former is more common (Howladar, 2017; Nicolli *et al*. 2010; Su *et al*., 2016).

Under oxidizing conditions, the key mode of As is released by oxidation of As bearing sulfide minerals like arsenopyrite (FeAsS) (Kim *et al*., 2012; Yoshizuka *et al*., 2010) and in case of reducing aqui-fers, As is released by reductive hydrolysis of metal hydroxides (Berg *et al*., 2007). The principal cause of As release from aquifer sediments is the reductive dissolution of Fe oxides (Guo *et al*., 2011). The level of pH in ground water plays an important factor for As enrichment in groundwater (Wang *et al*., 2019). Positively charged minerals, namely, Fe or Al oxides can absorb As easily (Islam, 2004).

At high pH values, the colloids as well as clay minerals carry the positive charges. The high level of As in groundwater is accompanied by high pH values ranges from 7.65 to 8.3. High occurrence of As in the soil might be occurs due to irrigation runoff (Polizzotto *et al*., 2013) as well as flooding may also be responsible for high As in groundwater (Yu *et al*. 2015). Furthermore, As also increases due to mining activities burning of fossil fuels, use of arsenical fungicides, herbicides and insecticides in agriculture and wood preservatives (Bose and De, 2013). Emission of As takes place in the environment because of volatilization of As_4O_6 by burning of coal, which condenses and ultimately transferred into water reservoirs.

Exposure Pathways and Toxic Effects of Arsenic to Human Health

Several studies have been carried out to document the toxicity of arsenic and its impacts on human health in various arsenic-contaminated regions around the globe (Engel and Smith, 2004). The main routes of exposure are arsenic contaminated water and food cooked with that water. Water soluble inorganic arsenicals are rapidly absorbed from the gastrointestinal tract (Khairul *et al*., 2017). Arsenic usually enters the body in the As (III) form through a simple diffusion mechanism. A small amount of As (V) could cross cell membranes through an energy-dependent transport system then it is reduced to As (III) to binds to DNA or protein molecules (Jomova *et al*., 2011). Arsenic poisoning is undetectable in primary stages and depending on the amount of As consumed, and immune system of the individual, it takes above 8 years to impact health. Recent studies have reported that human intakes of arsenic in a range of 0.05 mgL^{-1} leads to arsenicosis. Arsenic is associated with cerebrovascular disease, cardiac disease, leucomelanosis and hyperkeratosis, diabetes mellitus, pulmonary disease as well as diseases of the capillaries, arteries and arterioles (Fontcuberta *et al*., 2011).

Chronic arsenic ingestion from drinking water causes several disorders of the digestive system, respiratory system, cardiovascular system, hematopoietic system, endocrine system, renal system, neurological system, and reproductive system (Santra *et al*., 2013). These diseases ultimately increase the risk for bladder, kidney, liver, lung and lymphatic cancer, and diseases of the blood-vessels of the legs and feet, and possibly high blood pressure. In Bangladesh, several studies reported that about 25 million people of 2000 village areas of Bangladesh are at risk of As contamination and 3695 out of 17,896 people tested are suffering from arsenicosis. A large number of people in rural Bangladesh are becoming affected by the arsenic contaminated groundwater, which they collect from the tube-wells. The rural people are also getting affected due to the consumption of arsenic-contaminated foods, which they produce by using ground water containing arsenic (Huq *et al*., 2006). Several skin diseases for example, melanosis, hyperkeratosis, keratosis and leucomelanosis, etc., are the most common effects of drinking arsenic contaminated water. The other effects are liver enlargement and cirrhosis, peripheral neuropathy, hypertension, chromosomal abnormality, cardiac failure, diabetes mellitus, goiter, skin cancers and gangrene (Yunus *et al*., 2011).

The prolonged drinking of arsenic-contaminated water has effect on children's cognitive and psychological development (Asadullah and Chaudhury, 2011). Higher fetal loss and infant deaths have also been found in the regions where groundwater is highly arsenic contaminated (Sohel *et al*., 2010). Arsenic is also known to cause cytotoxicity, epidemiological toxicity and genotoxicity (Gentry *et al*., 2010, Suzuki *et al*., 2007). Epidemiological studies on the effects of arsenic consumption from drinking water on public health indicated a carcinogenic effect. Expert's indicated that a concentration level of 50 µg/L could lead to cancer in 1 in 100 individuals. During chronic poisoning, As causes strong pigmentation of hand and foot known as keratosis, and problems in other body system such as respiratory, neurological, high blood pressure, endocrine, cardiovascular and metabolic disorders (Ferlay *et al*., 2015).

Treatment Technologies for Mitigation of As from Ground Water

Many conventional and advanced treatment methods have been proposed for mitigating As from ground water under both laboratory and field conditions. Common methods for mitigation of As from contaminated water are use of chlorine, ozone, etc., physical method such as UV treatment, other filtration techniques such as reverse osmosis, membrane filtration, flocculation, adsorption, ion-exchange, etc. The following sections will demonstrate and evaluate some efficient and practical techniques for the purpose of arsenic mitigation. The performance and the deficiency of the existing techniques are further illustrated specifically.

Arsenic Mitigation by Oxidation

A common pretreatment step in most of the arsenic mitigation technologies is the oxidation of As (III) to As (V) because As (III) is the pre-dominant form of arsenic at neutral pH and adsorption of As (V) onto solid surfaces is easier than As (III) (Sharma *et al*.,

2007). Thus, oxidation followed by adsorption is thought to be efficient for the mitigation of As (Leupin and Hug, 2005).

$$H_3AsO_4 + 2H^+ + 2e^- \rightarrow H_3AsO_3 + H_2O$$

Arsenite could be oxidized by atmospheric oxygen, ozone or chemical oxidants. Chlorine, chlorine dioxide, hypochlorite, hydrogen peroxide, Fulton's reagent and potassium permanganate are usually applicable in this purpose, due to low cost and ease of availability (Lee *et al*., 2003).

$H_3AsO_3 + ½\ O_2 = H_2AsO^{4-} + 2H^+$
$H_3AsO_3 + HClO = HAsO_4^{2-} + Cl^- + 3H^+$
$3H_3AsO_3 + 2KMnO_4 = 3HAsO_4^{2--} + 2MnO_2 + 2K^+ + 4H^+ + H_2O$

54-57% of As (III) can be oxidized to As (V) in contaminated water with air and O_2 but complete oxidation (III) can be performed with ozone (Dodd *et al*., 2006). But this process is very expensive due to high energy input and mitigation of residual ozone and toxic. In general, oxidation with atmospheric air is a very slow process compared to chlorine and permanganate under wide range of conditions. However, chlorine dioxide is prohibited to use for surface water, American environmental agencies pay much attention to this factor. Chloroamine and hydrogen peroxide are slower than permanganate, chlorine and ozone. It is accepted that free Chlorine or hypochlorite is very effective for the oxidation of As (II), while the oxidation of As (III) is well achieved by ozone. However, chlorine dioxide is prohibited to use for surface water, American environmental agencies pay much attention to this factor. When manganese dioxide coated sand is combined with Fe containing compounds, this becomes efficient oxidizing agents because the treated products are easy to handle but in this process another treatment step is required for the mitigation of Mn from water. About the mitigation of arsenic, utilizing FeO_4^{2-} for purifying water is recommended HFO (hydrous ferric oxide) appears to be the most important phase responsible for removing the arsenic from drinking water around the world. It is well documented that High arsenic came into contact with iron oxides in the shallow aquifer, which could possibly lead to arsenic mitigation.

Thus, oxidation is very effective process for the mitigation of arsenic from water. Interfering particles present in water such as Fe (II), Mn (II), sulfide (HS^- and S^{2-}), total organic carbon (TOC) and dissolved organic carbon affect this process. Due to presence of S^{2-} and TOC, the oxidation rate of As (III) by ozone decreases significantly (Dodd *et al*., 2006). Thus, for the appropriate selection of oxidizing agents to accomplish high mitigation efficiency by oxidation it is very important to consider hydrophite chemistry and water composition.

Solar Oxidation Technique

In several studies photochemical and photocatalytic oxidation of As (III) has been investigated. UV irradiation increases the oxidation rate of As (III) with oxygen. This procedure can be catalyzed using sulfite, ferric iron or citrate. In case of solar light hydroxyl radicals generate by the photolysis of Fe (III) species. The oxidation rate is catalysed in presence of hydroxyl radicals. When As contaminated water in perchlorate and perchloric solution at pH 0.5-2.5was treated with Fe (III) followed by exposure to solar light the rate of mitigation of As increases. If lemon juice is applied after exposoure of solar light the mitigation rate of As from water becomes higher because the reaction of lemon juice (citrate) with strongly oxidizing radicals produces further radicals (Lara *et al*., 2006). Adsorption of As on TiO_2 after the oxidation of As (III) to As (V) by photocatalytic oxidation and TiO_2 was also investigated in several studies. This process can reduce arsenic levels to less than the standard limit given by WHO for drinking water (Miller *et al*., 2011). A TiO_2 impregnated chitosan bead (TICB) was synthesized and used for mitigation of As from aqueous solution. This study suggested that in the presence of UV light, a larger amount of As is adsorbed due to the increase of the surface area of the TICB and TiO_2 was able to photo-oxidize more As (III) to As (V) when comparing with the solution that was not exposed to UV light. In another study, nano-crystalline Al_2O_3 and TiO_2 impregnated chitosan was prepared for As mitigation (Yamani *et al*. 2012). Several factors such as the initial As concentration, pH, the presence of natural organic matter (NOM) and anions also influenced the rate of As (V) adsorption on TiO_2 (Bang *et al*., 2005, Miller *et al*., 2011). When a very low amount of TiO_2 is present the TiO_2/UV system has an inefficient As mitigation

due to incomplete oxidation (Guan *et al*., 2012). In addition, an acidic pH was more effective for adsorption of As (V) on the TiO_2 surface (Dutta *et al*., 2004). Presence of silicate, fluoride and phosphate bicarbonate affect the photocatalytic oxidation of As (III), and adsorption of As on the TiO_2 based adsorbent (Guan *et al*., 2012). Moreover, the treatment required for mitigation of arsenic residues is very complex. Due to several limitations, only oxidation is not considered as a highly effective procedure for mitigation of As.

In-Situ Oxidation

In the in situ oxidation process of arsenic and iron, the tube well water is allowed to oxygenate arsenite to arsenate by the oxygen which is present in the air and the ferrous iron in the aquifer is oxidized to ferric iron (Nicomel *et al*., 2016). This leads to a reduction in arsenic content in tube well water. The oxygenated water which containing As, and iron is flow back into the same tube well. When water is extracted again from the tube well, the concentration of arsenic will be low due to underground-precipitation and adsorption on ferric iron. The probable reactions of hydrous iron oxide with arsenate are shown below.

$$Fe(OH)_3\ (s) + H_3AsO_4 \rightarrow FeAsO_4.2H_2O + H_2O \ldots\ldots \quad (7)$$

$$FeOH^o + AsO_4^{3-} + 3\ H^+ \rightarrow FeH_2AsO_4 + H_2O \ldots\ldots\ldots \quad (8)$$

$$FeOH^o + AsO_4^{3-} + 2\ H^+ \rightarrow FeHAsO_4^- + H_2O \ldots\ldots\ldots \quad (9)$$

This process is suitable where the source of water is tube well or deep wells and others. To avoid contamination of the subsurface by introducing microbes from the surface care must also be taken. Some pore spaces may also become clogged with precipitates if dissolved iron levels are high in water. The potentiality of this process for the mitigation of arsenic is little; the results also indicate that arsenic concentrations can be minimized in the groundwater zone before water extraction (Sharma *et al.*, 2007).

Arsenic Mitigation by Biological Oxidation

Several bacteria known as arsenate respiring bacteria (ARD) may couple anaerobic oxidation of organic substrates to the reduction of arsenates such as *Geospirillum arsenophilus*, *Geospirillum barnesi*, *Desulfutomaculum auripigmentum*, *Bacillus arsenic-selenatis*, and *Crysiogenes arsenatis* (Oremland *et al.*, 2009). Due to formation of undesirable byproducts, application of chemicals in drinking water treatment is often discouraged. Several species of bacteria have been applied to carry out biological oxidation of As. Bacterial activity acts as catalyst to remove the arsenic from water. Some microorganisms such as *Gallionella ferruginea* and *Leptothrix ochracea* accelerate biotic-oxidation of iron (Katsoyiannis *et al*., 2004; Ekhlas *et al*., 2014). Iron-oxides and micro-organisms were settled in a filter medium, which has a adjuvant environment for the adsorption of arsenic. These microorganisms oxidize As (III) to As (V), which adsorbed on Fe (III). This leads to up-to 95% mitigation of arsenic. Bacterial oxidation of As (III) followed by mitigation of As (V) by sorption onto biogenic manganese-oxides was also studied (Sharma *et al*., 2007).

Biological oxidation is a new technique of the oxidation of iron and manganese as a treatment technique for arsenic mitigation (Sanjrani *et al*., 2019). These biological treatment techniques are the natural biological processes, and it takes a couple of days for remediation of metals in soil and groundwater by certain plants and microorganisms. During treatment, the following sequences of reactions have taken place in the treatment system:

a) Fe (II) to Fe (III) and Mn (II) to Mn (IV) (oxid.).
b) As (III) to As (V) (oxidation).
c) MnO_2 (Precipitation).
d) Abiotic-oxidation of As (III) by MnO_2.
e) As (V) sorption via MnO_2

This natural process for treatment can lead to up-to 95% of the mitigation of arsenic (Pallier *et al*., 2010). Mitigation of As (III) and As(V) from groundwater by biological oxidation of dissolved Fe and Mn in a pipe reactor (PR), followed by microfiltration (MF) was also studied. The latest PR-MF process is very efficiently removes Fe, Mn, and As without application of toxic chemicals for oxidation purpose or pH adjustment and there is no need of regeneration or backwashing and follows the principles of green chemistry.

Arsenic Mitigation by Coagulation–Flocculation

Coagulation followed by flocculation is another widely used treatment for the mitigation of arsenic from ground water (Andrianisa *et al*., 2008; Baskan and Pala

et al., 2010; Lakshmanan *et al*., 2010; Lacasa *et al*, 2011). It is commonly used for larger capacity facilities and it requires the production of a floc used to mitigate As from groundwater. Among other various chemical coagulants, this process usually requires Fe and Al based coagulants i.e. ferric chloride, ferric sulfate, aluminium sulfate. These chemicals need to be added and dissolved in water under efficient stirring for 1-10 minutes. In this process, Cationic coagulants have to decrease the negative charge of colloids and aggregation of particles forms larger particles (Choong *et al*., 2007). More than 90% of As (V) and 77% of As (III) can be removed by this technique. Oxidation of As (III) to As (V) with the addition of hypochlorite or potassium permanganate is required for effective mitigation. Aluminium chloride and polyaluminium chloride are able to reduce the concentration of As below the MRL (Hu *et al*., 2012). When kaolinite and $FeCl_3$ are used as a coagulant/flocculent, mitigation efficiency is over 90%and 77% for As (V) and As (III), respectively (Pallier *et al*., 2010). Fe based coagulants have been found to be most efficient in water treatment than the Al based coagulants (Katsoyiannis *et al*., 2004).

For efficient mitigation of arsenic from water, the arsenic needs to be adsorbed on amorphous metal hydroxides formed from coagulant. The rate of As mitigation is dependent on the quality and pH of the water before coagulation. The presences of organic matter in groundwater also affect the mitigation efficiency of this technique. The optimum mitigation was observed at pH below 8.5. There is critical limitation in the process of coagulation/flocculation; it produces a large amount of sludge along with a big concentration of arsenic. The management of this sludge is required to prevent the impact of secondary pollution of the environment and the treatment procedure of sludge is expensive (Mondal *et al*., 2013). Moreover, in many cases it becomes difficult to lower the arsenic concentration to the acceptable level by this technique (Shakoor *et al*., 2017).

Adsorption

Adsorption is a simple process in which water is flown through a packed bed of solid adsorption media filled in a column (Shakoor *et al*., 2017). Solutes adsorb on the adsorbent surface and its concentration become reduces in the solvent (Dong *et al*., 2009; Ungureanu *et al*., 2015). For treating arsenic contaminated water, activated carbon, Iron-based adsorbents, and low-cost materials such as agricultural wastes and byproducts industrial waste and byproducts, mud, etc. have been used as adsorbents (Ranjan *et al*., 2009; Haque *et al*., 2007; Khosa *et al*., 2014; Chutia *et al*., 2009; Banerjee *et al*., 2008; Sasaki *et al*., 2009; Liu *et al*., 2012; Zong-liang *et al*., 2012). Nanoparticles and nanomaterial based absorbents have also been investigated for the mitigation of arsenic such as zero valent iron (ZVI) nanoparticles, cupric oxide nanoparticles, titanium oxide nanoparticles, iron oxide based nanoparticles, zirconium oxide nanoparticles, During the flow of water through adsorbent column, arsenic in water are adsorbed onto the surfaces. Due to its several advan-tages, adsorption is the most widely used technique for arsenic mitigation such as, relatively high arsenic mitigation efficiencies, easy operation, and handling, cost-effectiveness, and no sludge production (Anjum *et al*., 2011; Jang *et al*., 2008). The mitigation efficiency of this method depends on the surface area, particle size, pore characteristics, density, zeta potential, mineralogy, characteristics of the surface functional groups, etc. of the adsorbent and the such as temperature, pH, arsenic concentration and ionic strength of the solution (Giménez *et al*., 2010; Zhu *et al*., 2014). The rate of arsenic adsorption and capacity adsorbents further depend on the presence of other ions such as: silicate, phosphate, HCO_3^-, and Ca^{2+}.

Activated Carbon

Activated carbon is graphite with amorphous structure with a wide range of pore sizes. Activated carbon is used either in powdered or granular form for arsenic mitigation. Ancient Hindus in India used Charcoal for filtration of drinking water and carbonized wood was a purifying agent and medical adsorbent in Egypt by 1500 b.c. Bone-char was replaced by activated carbon in sugar refining in 1901. Activated carbon was first used in the US for treatment of water in 1930. Arsenic adsorption onto pure activated carbon is very poor and regeneration is also difficult, so it is not directly applied for water treatment (Daus *et al*., 2004).

Increase of arsenic absorption capacity by activated carbon could be achieved when it is treated with

various metal compounds. Impregnating iron compound onto activated carbon and treatment of activated carbon with Zr are two way in this purpose. The latter was not suitable for drinking water because of toxic nature. Active carbons are prepared from bones, coconut shells, bagasse, carbon cobs, cereals blood, coal, coffee beans, bark, fish, fertilizer waste slurry, wood, coal, lignite, coconut shell peat etc (Deng *et al*., 2016). Adsorption capacity depends on activated carbon properties, adsorbate chemical properties, temperature, pH, ionic strength, etc. Although availability of activated carbons, this process remains expensive and vast quantities of activated carbon is required (Ahmad *et al.*, 2018).

Activated Alumina

Activated alumina (AA) successfully removed arsenic from ground water (Xie *et al*., 2013). Activated alumina is a granulated form of aluminum oxide (Al_2O_3) having good sorptive surface. This is an effective medium for arsenic mitigation from water with high dissolved solid content (Golami *et al*., 2009). The mechanisms of arsenic mitigation are similar to ion exchange resin, and are often collectively referred to as 'adsorption'. Its efficiency is greater than 95% under acidic conditions when Alumina surface is protonated. This process is controlled primarily by pH and mitigation capacity is effective in the narrow pH range from 5.5 to 6.0 where the alumina surfaces are protonated. Above pH 8.5, Arsenic mitigation capacity is reduced to only 2-5% of capacity at optimal pH. Therefore, pH adjustment is required for efficient arsenic mitigation from neutral and basic waters. For arsenic mitigation, fine (28-48 mesh) particles of activated alumina are commonly used. Arsenic adsorbed on the alumina surface during the flow of water through surface. After that column becomes saturated first at upstream zone later at downstream zone and finally it is completely saturated. The advantages of activated alumina are that it requires no chemical addition, it can treat thousands of bed volumes before breakthrough, and filters could be operated for months before the media need to be regenerated.

Regeneration of saturated alumina is done by exposing the medium to 4% caustic soda, NaOH, resulting caustic waste water with high arsenic content. After washing out of the residual caustic soda, 2% solution of sulfuric acid is used to neutralize the medium. During regeneration, the efficiency of the regenerated medium is decreased by 30-40%. If aluminium based sludge is produced during rinsed with sulfuric acid, this sludge will contain a high amount of arsenic because of its arsenic adsorption characteristics. This sludge and the remaining liquid fraction of the solution will require disposal because both residuals contain arsenic. When the AA has reached the end of its useful life, the media itself will also become a solid residual that must be disposed (Tripathy and Raichur 2008).

Natural Wastes and Agriculture Wastes

The adsorption technique using agricultural waste has been investigated as a cost-effective method for the mitigation of arsenic in wastewater. Lignin and cellulose in agricultural material contained aldehydes, ketones, carboxylic, alcohols, ether and phenolic groups, which may bind heavy metal ions through complex formation (Ahluwalia *et al*., 2005). The technique has advantages over others due to simplicity, low cost, high efficiency, flexibility and recovery (Gueye *et al*., 2016). Different agro-wastes are used as adsorbents for heavy metal mitigation such as orange peel, sawdust (Memon *et al*., 2008), banana peels (Israt *et al*., 2008), potato peel, rice straw, seaweed (Basha *et al*., 2008), wood and bark, tea waste (Malkoc *et al*., 2007), maize corn cob, jatropha oil cake, sugarcane bagasse (Garg *et al*., 2007), tamarind hull (Verma *et al*., 2006), rice husk, saltbush (Sawalha *et al*., 2005), marine algal biomass, olive pomace, activated sludge, sugar beet pulp, wool, olive cake, sawdust, pine needles, almond shells, cactus leaves and charcoal, seafood processing waste sludge and pine bark (Charlet *et al*., 2007). The fresh agro wastes were washed with distilled water to remove all dirt, cut into smaller sizes and air dried to remove the free water before oven dry. Then dried in oven at 100°C for 24 hours and homogenized in a blender to utilize as an adsorbent.

Ion Exchangers

Ion exchangers are also used for mitigation of As from water (Oehmen *et al*., 2006). Tetrahedron ion exchange resin filter tested under rapid assessment program in Bangladesh (BAMWSP *et al*., 2001) showed promising results in arsenic mitigation. In ion ex-

change, through dissolved ion the reversible displacement occurs of an ion that will adsorb on the surface of solid materials. For the treatment of As strong base anion exchange resins are commonly used where the oxyanionic species of arsenate are effectively exchanged with the anionic charged functional group of the resin (Chang *et al*., 2010). Thus produces effluents with low concentration of As (V) (Choong *et al*., 2007). Sulfate, phosphate and nitrate are mostly used for the mitigation of As via ion exchange. In case of arsenite oxidation of As (III) to As(V) is an important pretreatment step for ion exchange processes because arsenite usually exists as a neutral molecule. In this process at first the resin bed are usually flowed through HCl to implant labile Cl^- on the surface of resin, which is later exchanged with As. Thus, the effluent contains a large amount of Cl- and additional step is needed to enhance the quality. The arsenic exchange and regeneration equations with common salt solution are as follows:

Arsenic exchange: $2\ R\text{-}Cl + HAsO_4^{2-} = R_2HAsO_4 + 2Cl^-$

Regeneration: $R_2HAsO_4 + 2N^+ + 2Cl^- = 2R\text{-}Cl + HAsO_4^{2-} + 2Na^+$

Where R stands for ion exchange resin.

There are several disadvantages of this process such as. During the presence of sulfate in the treated water exchange of sulfate is more preferable than As for commercial resins. Additionally, impedence of the resin takes place due to iron precipitation and solid particles in aqueous solution. So the resin becomes exhausted, it needs to be regenerated which increases the cost of operation. Due to the generation of large quantities of toxic sludge and is considered as a more expensive treatment compared with other techniques (Ahmad *et al*., 2017). Moreover, during regeneration of resins, As rich brine solutions are produced which may again lead to environmental release of As (Cundy *et al*., 2008).

Membrane Filtration

Membrane filtration technique is also used for the mitigation of arsenic from groundwater. Membranes are synthetic materials having billions of pores serving as selective barriers which allow some dissolved compounds to pass through but retains contaminants. Pressure difference between the feed and the permeate sides is the driving force which is needed to transport the water through the membrane. There are two types of pressure-driven membrane filtrations: (i) low-pressure membrane processes, such as ultrafiltration (UF) and microfiltration (MF); and (ii) high-pressure membrane processes, such as nanofiltration (NF) and reverse osmosis (RO) (Shih, 2005).

These Separation processes mostly depend on the the membrane's pore size and increasing driving pressure increases the selectivity of the processes. Separation is accomplished via mechanical percolation for MF and UF membranes while, capillary flow or diffusion is responsible for the separation by NF and RO membranes (Shih, 2005). The size of the soluble As are small enough to pass through the MF membrane. Thus the efficiency of the process is highly dependent on the size distribution of As bearing particles in water (Shih, 2005) for improving the mitigation efficiency, such as coagulation and flocculation (Singh *et al*., 2015). The mitigation of As using flocculation followed by MF was better than flocculation sedimentation when $FeCl_3$ and $Fe_2(SO_4)_3$ are used as flocculants. But adsorption of As on the Fe (III) complex was interfered by the pH and other ions present in solution. UF is also a low pressure technique which is alone not effective for mitigation of As due to large membrane pores (Velizarov *et al*., 2004). The membrane's pores are not sufficient small to remove the dissolved As in water. As a result, surfactant-based separation processes such as micellar-enhanced ultrafiltration (MEUF) can be utilized to increase the mitigation of As (Iqbal *et al*., 2007). Micelles are formed after the adsorption of As onto the surface of the cationic surfactant, which are removed by UF. The study conducted by (Brandhuber and Amy 2001) found a moderate rejection of 65%and 53%for As (V) and As (III), respectively. Among different cationic surfacetants hexadecylpyridinium chloride (CPC) showed the highest mitigation efficiency, i.e., 96%. However, the mitigation of As was depended on the pH of the solution, the initial As concentration and the presence interfering particles (Mondal *et al*., 2013).

Reverse Osmosis

Reverse Osmosis is high pressure technique and able to remove dissolved As from water to an appreciable

level (Figoli *et al*., 2010). Both As (V) and As (III) can be effectively removed (up to 99%) from water RO membranes (BAMWSP *et al*., 2001). Here size exclusion regulates the separation and not the charge interaction. Charged membranes generally have a higher rejection for charged solutes than for non-charged solutes (Seidel *et al*., 2001). The size of pore of the membrane does not have a significant effect on As rejection, but rather charge exclusion is predominant over the size exclusion mechanism. RO membranes are non-porous so transport of the solvent occurs through the free volume between the segments of the polymer of which the membrane is composed. These membranes are used for water desalination and low molecular mass compounds can be highly rejected (Velizarov *et al*., 2004). It was shown that the mitigation efficiency for As (V) exceeded 85% while that of As (III) was far too low (Uddin *et al*., 2007). In case of RO membranes initial concentration of As had no effect on the removal rate but the removal of As was affected by the pH of the solution and operating pressure. The effect was much higher for As (III) than As (V). Yoon *et al*. (2009) concluded that mitigation of As (III) below pH 10 was low because of the existence of uncharged As (III) species in solution. When As (III) was the dominant As species, the mitigation efficiency of As from ground water became less than 50% (Walker *et al*., 2008). Therefore, RO is not effective for As contaminated water where As (III) is dominanted. Oxidation is required as a pretreatment step to mitigate total As at the desired concentration. As RO is very expensive, it is not favorable in developing countries (Uddin *et al*., 2007).

Nano-Filtration

NF is suitable for the mitigation from water of dissolved compounds with a molecular weight above 300 g/mol (Seidel *et al*., 2001). In neutral and alkaline solution NF membranes are generally possesses negatively charged. Here separation of As is occurred due to electrostatic repulsion between the anionic As species and the charge of membrane (Velizarov *et al*., 2004). NF is more sensitive than RO with respect to pH and the solution's ionic strength. The rejection of As (V) was better than As (III) and As (III) could not be reduced to MCL (Uddin *et al*., 2007). Thus oxidation of As (III) to As (V) is required pretreatment step. Saitua *et al*. (2005) found that As rejection was independent of transmembrane pressure, cross-flow velocity as well as temperature. But a recent study demonestrated that the efficiency of As (V) mitigation increased with increase of pH, decrease of operating temperature and the initial As concentration (Figoli *et al*., 2010). Although NF gives desired results in mitigation of As (V) from solution, the problem of this technique is the mitigation of the uncharged As (III) species in the near neutral pH range. Thus, without oxidative pretreatment, NF is not enough to remove total As (Uddin *et al*., 2007).

Comparison between Different Techniques

Many factors can affect arsenic mitigation efficiency including arsenic concentration, speciation, pH and co-occurring solutes. Therefore, any method should be tested using the actual water to be treated, before implementation of arsenic mitigation systems at the field scale.

Table 1: summarizes the comparison of some effective available techniques on the basis of expense, suitability and percentage of arsenic mitigation.

Techniques		Removal efficiency of As (III)	Removal efficiency of As (V)	Relative Cost	Operating skill	Sludge disposal	Pretreatment required
Oxidation		Less than 30%	60% to 90%	low	Low	Yes	Yes
Coagulation flocculation	Iron	Less than 30%	Greater than 90%	high	High	produces toxic sludges	Yes
	Alum	60% to 90%	Greater Than 90%				
Ion exchange adsorption		Less than 30%	Greater than 90%	high	High	Solid sludge disposal problem	No
Membrane		60% to 90%	60% to 90%	high	Medium	No sludge disposal	Yes

Techniques					problem	
Activated carbon	Less than 30%	Greater than 90%	low	Low	No sludge disposal problem	
Activated alumina	60% to 90%	Greater than 90%	Medium	Low	Toxic solid waste	Yes
Natural waste	-	22.8% to 82%	Low	Low	No sludge disposal problem	no
In situ oxidation	60% to 90%	Greater than 90%	Medium	Medium	No arsenic-rich wastes	
Precipitation	Less than 30%	Greater than 90%	Low	Low	No sludge disposal problem	Yes
Reverse Osmosis	60% to 90%	60% to 90%	High	High	No solid waste. Removes other contamination too.	Yes
Nano-filtration	60% to 90%	60% to 90%	High	High	No sludge disposal problem	Yes

CONCLUSION

The accumulation of arsenic in drinking water from different sources plays an important role in water pollution and this is a major worldwide problem. Arsenic has created health problems around the world; arsenic has been documented in the Americas, Africa, Asia, Europe and Pacific countries. Bangladesh, India, Nepal and North American countries are having severe condition of arsenic contamination. Many Governments and WHO had made several efforts to improve the condition of contaminated groundwater and its severe effect on human health. But as Bangladesh is highly mass infected, this problem is not in control here and it is going bad day by day. Among different types of sources anthropogenic sources are getting more effective day by day in modern ways of development throughout the world. Arsenic enters in the human body by direct consumption of drinking water contaminated by arsenic and the indirect intake through foods and crops cultivated using arsenic-contaminated water. For arranging safe drinking water, it is required to mitigate of arsenic from the water source. The conventional arsenic mitigation technologies include oxidation (biological and chemical), adsorption, ion exchange resin, membrane processes, co-precipitation and bacterial treatment. Most of these technologies for mitigation of arsenic involve the direct mitigation of As(V) or converting As(III) to As(V) followed by mitigation of As(V). Many water purifier manufacturing companies are using reverse osmosis due to its high efficiency. But in case of mass infected people, it seems that it is unable to fulfill the requirement. However, more research is required to evaluate the practicability and feasibility of these treatment processes as well as to lower the expenses of treatment. The government should monitor and document industrial and agricultural activities as they brought the arsenic pollution issue to the bodies of water in the first place. There should analysis of the discharge from industrial plants, which aim to supply safe drinking water to people in rural areas. The government should take steps and put restrictions about the handling of industrial waste. Investment should be employed on the great engineering system for water transportation and documentation of water quality.

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CONFLICTS OF INTEREST

The Authors declares that there is no potential conflict of interest.

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