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Arsenic Removal Technologies and the Effect of Source Water Quality on Performance

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Arsenic Removal Technologies and the Effect of Source Water Quality on Performance

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Abstract

Arsenic removal technologies that are effective at the tens of ppb level include coagulation, followed by settling/microfiltration, ion exchange by mineral surfaces, and pressure-driven membrane processes (reverse osmosis, nanofiltration and ultrafiltration). This report describes the fundamental mechanisms of operation of the arsenic removal systems and addresses the critical issues of arsenic speciation, source water quality on the performance of the arsenic removal systems and costs associated with the different treatment technology categories.

TABLE OF CONTENTS

List of Figures

List of Tables

1.0 Introduction

Aquatic Chemistry of Arsenic

Arsenic is a metalloid found in group V of the periodic table, along with nitrogen, phosphorus, antimony, and bismuth. Arsenic can occur in water in four oxidation states: +V (arsenate), +III (arsenite), 0 (arsenic), and –III (arsine) (Clifford and Zhang, 1993). In oxygenated waters, the predominant form of arsenic is arsenate, which hydoxylizes into anionic forms - $H_2AsO_4^-$, $HAsO_4^{2-}$, or AsO_4^{3-} (5 < pH <12). Under anoxic conditions, arsenite is most abundant. Arsenite species H_3AsO_3 and anionic H_2AsO^{3-} dominate below and above pH 9.2, respectively. Although it is possible to predict arsenic speciation at different redox conditions from thermodynamics (Refer Figure 1), in natural environments the kinetics of transformation reactions are subject to chemical and microbial influences (Edwards, 1994).

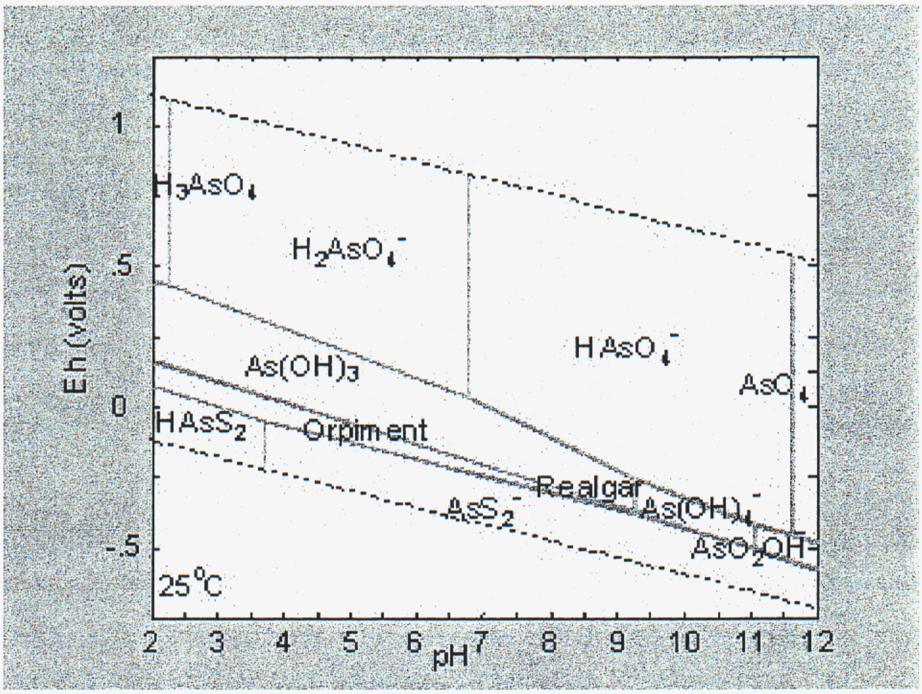

Figure 1. Eh-pH Diagram for Arsenic Species. Source: Brady et al. (1999).

2.0 Fundamentals of Arsenic Removal

2.1 Coagulation Using Metal Salts

Edwards (1994) defines arsenic removal from water by coagulation as the conversion of dissolved arsenic to insoluble products by the combined mechanisms of precipitation, co-precipitation and adsorption. Precipitation is the insolubilization of contaminants by exceeding the solubility product. Co-precipitation is the incorporation of soluble arsenic species into a growing hydroxide phase, via inclusion, occlusion, or adsorption and adsorption is the formation of surface complexes between soluble arsenic and the solid oxyhydroxide surface sites (Edwards, 1994), e.g:

$\equiv Fe\text{-}OH + H^+ + H_2AsO_4^- \rightarrow \equiv Fe\text{-}H_2AsO_4 + H_2O$ (arsenate sorption)

$\equiv Fe\text{-}OH + H^+ + H_2AsO_3^- \rightarrow \equiv Fe\text{-}H_2AsO_3 + H_2O$ (arsenite sorption)

Where: $\equiv Fe\text{-}OH$ denotes a surface site exposed at the oxyhydroxide-water interface. The trivalent metal salts used for arsenic removal by coagulation are alum and ferric salts (see reviews of Harper et al., 1992, Cheng, et al., 1994, Edwards, 1994, Scott et al., 1995, Hering and Elimelech, 1996, McNeill and Edwards, 1997 and Banerjee et al., 1999). Under comparable conditions (pH, arsenic concentrations, coagulant type and coagulant dosage), As (III) removal efficiency is less than that of As (V). This is because below a pH of 9.2, arsenic (III) exists in the uncharged form as H_3AsO_3 and thus is not electrostatically favored to form to any great extent to the positively charged metal oxyhydroxide surface. In the treatment of groundwater containing arsenic (III) and arsenic (V), species oxidation is therefore, a required pre-treatment step.

Figure 2 summarizes the process of arsenic removal using alum and ferric salts. The treatment train for arsenic removal from groundwater requires a preoxidation step to convert all the arsenite to arsenate, pH adjustment for enhancement of coagulation followed by coagulation to convert dissolved arsenic to insoluble products and settling/micro-filtration to remove the insoluble products of coagulation. In general, greater arsenic removal is achieved by coagulation with ferric salts with than alum salts at near neutral pH. Greater process attention is required for aluminum than iron salts because of the differential solubility of $Al(OH)_3$. In general, operational features critical to arsenic removal from groundwater using coagulation settling/microfiltration are pH, coagulant type and dosage, oxidant addition, efficient mixing of coagulant with source water and rate of settling/filtration. Naturally occurring organic matter, competing ions such as sulphates, nitrates, phosphates and silicates can decrease the removal efficiency of arsenic by coagulation. by interfering or by competing for binding sites on the coagulant surface. Researchers have also shown that presence of certain divalent metal ions in water can enhance the process of removal of arsenic from water by coagulation settling/microfiltration.

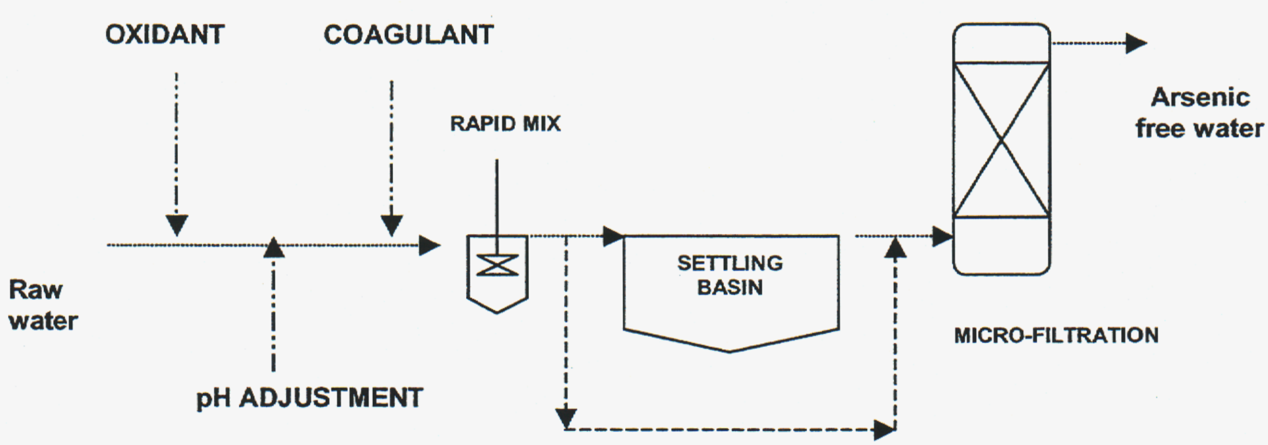

Figure 2. The coagulation settling/microfiltration process for arsenic removal.

2.2 Adsorption Processes

Adsorption is a mass transfer process in which a substance is transferred from the liquid phase to the surface of a solid where it becomes bound by chemical or physical forces. In the case of oxyanions, such as arsenate and arsenite, adsorption occurs on the oxide water interface by forming a complex with surface sites that may be positively charged, such as a protonated surface hydroxyl group. In other instances the reaction may involve a ligand exchange mechanism in which the surface hydroxyl group is displaced by the adsorbing ion (AWWA Research Foundation, 2000). The adsorption reaction mechanism of arsenic species onto solid metal oxyhydroxide surfaces may be generically represented by (AWWA Research Foundation, 2000 and Edwards, 1994):

$$\equiv\overline{S\text{-}OH} + H^+ + H_2AsO_4^- \rightarrow \equiv\overline{S\text{-}H_2AsO_4} + H_2O \text{ (arsenate sorption)}$$

$$\equiv\overline{S\text{-}OH} + H^+ + H_2AsO_3^- \rightarrow \equiv\overline{S\text{-}H_2AsO_3} + H_2O \text{ (arsenite sorption)}$$

Ion-exchange is a special case of adsorption where ionic species in aqueous solution are removed by exchange ions of a similar charge attached to a synthetic resin surface.

Adsorption processes commonly used in water treatment are: adsorption onto activated alumina, ion-exchange, iron oxyhydroxides and manganese dioxide coated sand. (Banerjee, et al., 1999, Torrens, 1999). Figure 3 summarizes the typical treatment set-up for sorption process for arsenic removal. The efficiency of each media depends on operating conditions, such as: pH, the presence of interfering ions, speciation of arsenic, system dependent parameters (e.g., empty bed contact time, surface loading rates, bed-porosity etc.) and the use of oxidizing agent(s) in the pre-treatment train. In general, As (V) is easier to remove from water, since it has a residual negative charge above a pH of 2.2 and is attracted to positively charged metal hydroxide surfaces. As (III) is uncharged in most natural waters below pH 9.2 and has no charge affinity to surfaces. The charge neutrality makes it difficult to remove As (III) from natural waters (Edwards, 1994).

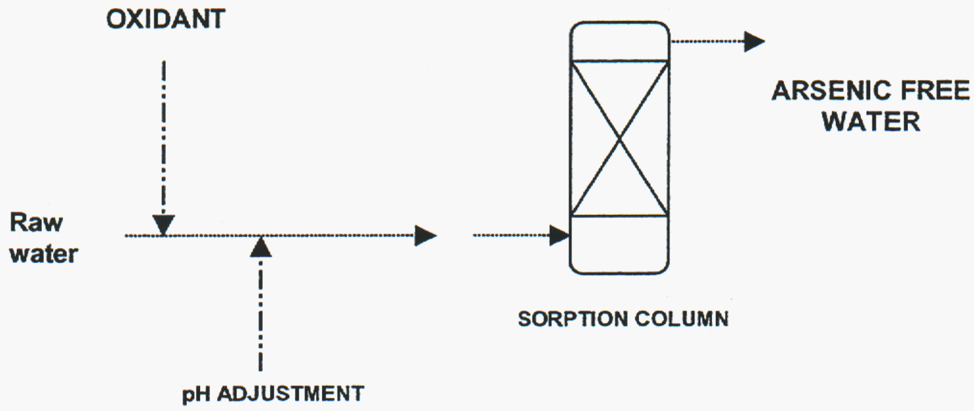

Figure 3. Sorption process for arsenic removal.

2.2.1 Activated Alumina

Activated alumina is known to be effective in the removal of arsenate, but pH has a strong effect. The optimal pH range for arsenate removal is 5.5 to 6.0. Above a pH of 8.2 (point of zero charge (ZPC) below which activated alumina surface has a positive charge and above which activated alumina surface has a negative charge and acts as a cation exchanger) arsenate removal efficiency decreases greatly (Clifford, 1990). For feed water at or near neutral pH, adjustment of pH may be necessary for effective arsenate removal. The adsorption capacity and bed breakthrough is dependent on the pH of the influent water and the concentration of arsenic in the feed water.

The ability of activated alumina to remove arsenite is considerably less than arsenate and breakthrough occurs faster (Frank and Clifford, 1986). Adsorption capacity is dependent on pH. Maximum adsorption occurs at a pH of 6, while around neutral pH, the adsorption capacity decreases greatly (Clifford and Lin, 1991). Surface oxidation of arsenite to arsenate may occur to a small degree at the activated alumina surface (Driehaus et al., 1995).

Competitive ions have a less pronounced effect on activated alumina adsorption than they do for ion exchange systems. Phosphates and fluorides compete directly for the exchange sites with arsenate. Sulphates, silicates, and chlorides also reduce adsorption of arsenate to activated alumina (Rosenblum and Clifford, 1984).

Activated alumina is typically regenerated with a 2-5% sodium hydroxide solution and the bed is then flushed with acid to re-establish a positive charge on the grain surface. Regeneration of activated alumina is more difficult and less effective than ion-exchange regeneration (Clifford, 1986). With activated alumina, sites are saturated by arsenate ions that are irreversibly adsorbed to the sorption surface. Regeneration also reduces the active bed volume due to dissolution of alumina (Ghurye et al., 1999).

2.2.2 Ion-exchange

Ion-exchange medium, typically synthetic resins, are made up of cross-linked polymer matrices possessing charged functional groups attached by covalent bonding (Clifford, 1990). Both strong and weak base functional groups are used to prepare ion exchange resins. Strong base resins operate over a wide pH range in contrast to weak base resins which are effective at acidic pH ranges (Clifford, 1990). Theoretically, anionic ion exchange resins can remove only arsenate. Pre-oxidation is required for arsenite removal (Frank and Clifford, 1986). Anion exchange resins are regenerated by flushing with solutions containing weakly sorbing anion species such as chloride.

Ion selectivity is a key performance issue in ion exchange processes. For this reason, anion exchange is not an attractive process for arsenic removal at high total dissolved solids (>500 mg/L) and high sulphate concentrations (> 25 mg/L). At elevated total dissolved solids and sulphate concentrations, ion-exchange systems are prone to fouling, decrease in performance cycle length and chromatographic peaking (i.e. effluent concentrations are greater than influent concentrations during a cycle of operation). For example, elevated concentrations of sulphate in the feed water can lead to dumping of arsenic in the effluent 1.3 to 6 times higher than influent arsenic concentrations) (Clifford, 1990). A selectivity sequence for the treatment of groundwater using anion exchange as reported by Clifford et al. (1986) is as follows:

$HCrO_4^- > CrO_4^{2-} > ClO_4^- > SeO_4^{2-} > SO_4^{2-} > SeO_4^{2-} > NO_3^- > NO_3^- > Br^- > (HPO_4^{2-}, HAsO_4^{2-}, SeO_3^{2-}, CO_3^{2-}) > CN^- > NO_2^- > Cl^- > (H_2PO_4^-, H_2AsO_4^-, HCO_3^-) > OH^- > CH_3COO^- > F^-$.

2.2.3 Adsorption to Iron-oxyhydroxide Surfaces

The generic mechanism for the removal of arsenic by sorption onto metal oxide—oxyhydroxide surfaces has been explained earlier. Both arsenate, and to a limited degree arsenite, are removed by iron oxyhydroxides. Adsorptions of arsenate and arsenite have different sensitivities to pH. Lowering the pH enhances the removal of arsenate, whereas arsenite removal is not affected by pH change (Edwards, 1994). The sorption capacity for arsenate can be twice that of arsenite. Studies with packed columns containing iron oxyhydroxide showed that arsenite breakthrough occurs considerably faster than arsenate (AWWA Research Foundation, 2000). For efficient arsenic removal from groundwater, the addition of oxidant and lowering of pH around 5.5-6.0 should be a standard operational procedure. Regeneration of iron oxyhydroxides can be achieved with sodium hydroxide, but complete removal of all the arsenic attached to the surface may not be possible (AWWA Research Foundation, 2000). As in the case with activated alumina, competitive ions have a less pronounced effect on arsenic adsorption to iron oxyhydroxide surfaces than they do for ion exchange systems. Phosphates and fluorides directly compete for the exchange sites with arsenate. Sulphates, silicates, and chlorides also reduce adsorption of arsenate to iron oxyhydroxide surfaces (Rosenblum and Clifford, 1984).

2.2.4 Manganese Dioxide Coated Sand

Manganese dioxide coated sand (MDCS) is prepared by the oxidation of manganese ions coated on sand surfaces (Murry, 1974). Researchers have shown MDCS has the ability to remove arsenic from water (Viraraghavan et al., 1999 and Bajpai and Chaudhuri, 1999). The mechanism is thought to involve oxidation of arsenite to arsenate followed by sorption (Oscerson et al., 1983 and Takamatsu et al., 1985, Bajpai and Chaudhuri, 1999). Arsenic removal with MDCS is still in the development stage and success of this process at commercial scale is yet to be determined.

2.3 Membrane Separation Processes

Pressure driven membrane separation process involves the forced passage of water through a selective membrane, which rejects undesirable species (refer to Figure 4). The potential that controls the flux of water across the membrane (i.e. the driving force), is the difference in pressure across the membrane. In terms of increasing selectivity, pressure driven membrane processes include: microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). Separation in MF and UF membranes occurs via mechanical sieving, whereas capillary flow or solution diffusion is responsible for separation in NF and RO. It should be noted that as membrane selectivity increases, the required driving pressure increases (AWWA Membrane Technology Research Committee, 1992).

Both RO and NF membranes have high rejection rates at high flux for As (V). In the case of As (III), only RO and tight NF membranes have high rates of rejection, but the rejection rate decreases with flux. Pre-oxidation enhances the rate of removal of As (III) by NF. Substantial arsenic removal may be obtained with coagulation as a pre-treatment for use with membranes with larger pore size (Brandhuber and Amy, 1998).

Removal of arsenic by membranes is independent of the pH of the influent. However, an operating pH of 5 to 6.5 is preferred to prevent deterioration of the cellulose acetate membrane. Co-occurring ionic species generally do not affect membrane processes; however, control of scale formation on membrane in hard waters by pH adjustment (pH< 6) and control of organic fouling may be necessary for prolonged operation of the membrane systems (Waypa, et al., 1997).

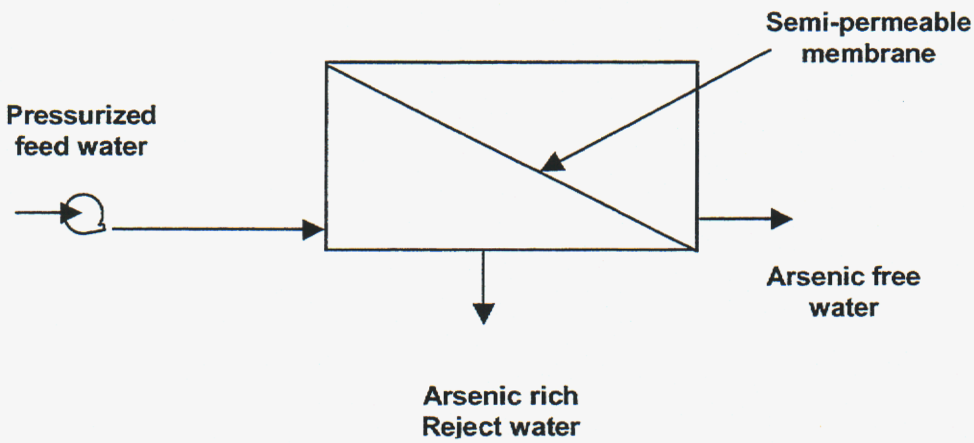

Figure 4 Membrane Systems for Arsenic Removal.

3.0 Oxidizing Agents Used in Preoxidation of Arsenite to Arsenate

Arsenite can be oxidized to arsenate by ozone, chlorine and permanganate under given appropriate oxidant dosages and residence times, with ozone being the superior oxidant on an equivalent dosage basis. Generally complete conversion of arsenite to arsenate can be achieved by the three oxidants mentioned above if an excess of the stoichiometric amount of oxidant is provided. The theoretical stoichiometric equivalent oxidant for arsenite oxidation by oxidants are: 0.64, 1.41 and 0.99 mg of oxidant/mg of arsenite respectively, for ozone, potassium permanganate, and sodium hypochlorate.

4.0 Effect of Water Quality on Performance

Groundwater typically includes elements other than arsenic, which greatly influences arsenic removal in either an antagonistic (e.g., anions such as phosphate competing with arsenate in the removal technology), or synergistic way (e.g., iron oxidation and precipitation which enhances arsenic removal). The redox state of water and consequently the oxidation state of the anions and cations will dictate which chemical reactions are dominant. The three aspects of water chemistry, which influence treatment efficiency, are: matrix components, pH and redox state. Note first of all that the parent mineralogy of the soils through which the aquifer flows largely controls all of these through determining the ions present; and the presence or absence of particulate phase organic carbon. Oxidation of organic carbon consumes dissolved oxygen and can change the redox state from aerobic to anoxic. Consequently, waters from anaerobic aquifers will require more oxidative pre-treatment than waters from aerobic aquifers.

Arsenic removal technology performance works better when optimized for a particular water source. The optimization process may include design changes to the technology (i.e. increased hydraulic retention time) or the inclusion of pre-treatment to remove interferences from the influent water. Table 1 is a summary of water quality parameters, which may affect the performance of arsenic removal technologies. The impact of the water quality parameter depends on the degree of deviation from the optimum range. Also potential combined interaction(s) between two or more water quality parameters must be accounted for.

Table 1. Effect of Water Quality Parameters on Treatment.

Treatment	Water Quality Parameter	Effect
Coagulation	Neutral to high pH, variation in concentration and speciation of arsenic, presence of phosphates, sulfates, silicates, fluoride, iron and instances of hard water.	Adjustment in pH, coagulant dosage and oxidant dosage will be required from well to well for optimal removal efficiency and filter life. Presence of iron and hardness ions may enhance arsenic removal efficiency but may irreversibly foul filter media. Presence of phosphate, fluoride, sulfate and silicate ions may compete for oxyhydroxide surface sites.
Adsorption	Near neutral to high pH, variation in concentration and speciation of arsenic, presence of phosphates, fluoride, sulfates, silicates iron and manganese ions, and instances of hard water. High total dissolved solids (TDS) and natural organic matter (NOMs).	Arsenic sorption is highly sensitive to pH and arsenic speciation. Variation in pH and speciation will affect process efficiency and bed life. Phosphate, sulfate, fluoride and silicate ions will compete with arsenic for sorption sites. Irreversible fowling due to high concentrations of iron, manganese, and hardness is of concern. High TDS may cause media fouling and NOMs Presence of NOMs may enhance bio-fouling.
Ion-Exchange	Variation in concentration and speciation of arsenic, presence of phosphates, sulfates, silicates, bicarbonate alkalinity, and fluoride, iron and hard water. High total dissolved solids (TDS), NOMs.	Ion-exchange can only remove arsenic (V), thus pre-oxidation is necessary, pH adjustment is not required between pH of 2 .2 to 11. Anions such as phosphate, sulfate, fluoride, silicate, and bicarbonate will compete for sorption sites and may contribute to chromatographic peaking. Fouling due to the presence of excess iron and hardness is of concern. Precipitation of insoluble iron salts and calcium and magnesium salts may foul media. High TDS may cause media fouling and NOMs Presence of NOMs may enhance bio-fouling.
Pressure-driven membrane processes	Near neutral pH, iron and hard water. Presence of NOM.	The concern is more towards membrane life optimization and preventing membrane fouling. Adequate pretreatment and pH adjustment may be necessary. Presence of NOMs may enhance bio-fouling of membrane.

5.0 Cost of Treatment and Residual Handling

The cost of treatment and residual handling is greatly dependent on the characteristics of the groundwater. Water quality parameters such as pH, concentration of arsenic, speciation of arsenic and the presence of sulfates greatly affect both capital and operating cost of the treatment system. Table 2 illustrates the cost associated with treating groundwater contaminated with arsenic. These treatment cost estimates are based on Albuquerque, New Mexico groundwater with design treatment capacity of 2.3 mgd (Chwirka et al., 2000).

Table 2. Estimated Capital and Operating Costs of Different Arsenic Removal Technologies With The Design Capacity of 2.3 mgd (Chwirka et al., 2000).

Technology	Capital Cost Including Residual Handling ($)	Annual Operating Cost Including Residual Handling ($)	Cost ($) to Treat 1000 gallons
Ion-exchange	5,243,000	447,000	0.53
Activated alumina adsorption	4,557,000	444,000	0.54
Coagulation microfiltration	4,149,000	273,000	0.33

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