

03-216

Environment Canada

Water Science and
Technology Directorate

Direction générale des sciences
et de la technologie, eau
Environnement Canada

Remediation of Arsenic-Contaminated Groundwater

By:

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NWRI Contribution # 03-216

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Remediation of Arsenic-Contaminated Groundwater

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Abstract

Selection of a method for the remediation of arsenic-contaminated groundwater requires knowledge of the physical hydrogeological setting, chemical composition of the groundwater and aquifer solids, and concentrations and forms of arsenic in the dissolved and solid phases. There are a number of new approaches for remediating groundwater contaminated with inorganic substances. These approaches typically involve manipulation of the geochemistry of the aquifer or contaminant plume such that the inorganic contaminant is either mobilized and extracted from the aquifer or stabilized to remove it from the water. Successful deployment of a technology depends on site-specific characteristics, the expense of the technology, including costs covering the initial infrastructure and ongoing maintenance costs, and the technical performance requirements to meet the desired remediation objectives. Hydrogeological and geochemical factors which require consideration in selecting a remediation system, and remediation technologies available for treating arsenic contaminated groundwater are described.

Assainissement des eaux souterraines contaminées par l'arsenic

Carol J. Ptacek et David W. Blowes

Résumé

Lorsqu'il s'agit de choisir une méthode d'assainissement d'eaux souterraines contaminées par l'arsenic, il faut connaître les caractéristiques physiques et hydrogéologiques du site en jeu, la composition chimique des solides qui se trouvent dans les eaux souterraines et dans l'aquifère, ainsi que les concentrations et les formes d'arsenic en solution et dans la phase solide. On compte un certain nombre de nouvelles méthodes d'assainissement des eaux souterraines contaminées par des substances inorganiques; elles consistent habituellement à modifier les propriétés géochimiques de l'aquifère ou du panache de contaminants de manière soit à rendre les contaminants organiques mobiles pour ensuite les extraire de l'aquifère, soit à les stabiliser pour les retirer de l'eau. L'efficacité de la technique utilisée dépend des caractéristiques propres au site, du coût de la technique, y compris les frais associés à la mise en place initiale de l'infrastructure et les dépenses pour l'entretien permanent, de même que de la qualité technique nécessaire pour atteindre les objectifs d'assainissement visés. Dans l'article, nous décrivons les facteurs hydrogéologiques et géochimiques qu'il faut prendre en considération au moment de choisir une méthode d'assainissement, ainsi que les techniques d'assainissement existantes pour traiter les eaux souterraines contaminées par l'arsenic.

NWRI RESEARCH SUMMARY

Plain language title

Remediation of Arsenic-Contaminated Groundwater.

What is the problem and what do scientists already know about it?

Arsenic is present in groundwater as a result of natural processes and anthropogenic releases. Ingestion of arsenic-contaminated groundwater leads to the development of cancer and other health concerns. There is abundant knowledge on the occurrence of arsenic in groundwater in many parts of the world. Additional studies have been conducted on mechanisms controlling the release, transport and remediation of arsenic in groundwater. There remain, however, numerous localities and geological systems where very little is known about the transport and remediation of arsenic in groundwater.

Why did NWRI do this study?

This study was undertaken to gather information on various remediation approaches to remove arsenic from groundwater. There are many remediation approaches available. Application of a remediation approach requires knowledge of site-specific geochemical mechanisms controlling arsenic behaviour. This study involved a review of various remediation approaches and includes a detailed background discussion on geochemical and physical processes controlling arsenic release, transport and attenuation. New data collected from a mine site in Ontario is used as an illustrative example.

What were the results?

The majority of studies in the field of arsenic remediation are site specific. It is critical to understand the site geochemistry prior to implementation of a remediation approach. The remediation approaches typically rely on manipulation of physical and chemical processes. This manipulation can potentially exacerbate the original problem, therefore, a thorough understanding of the geochemical processes occurring at a given site is required prior to implementation of a remediation system. Examples are provided to illustrate the need to have sufficient background information prior to selection of a remediation option.

How will these results be used?

The information can be used to control the release of arsenic to prevent widespread contamination of drinking water supplies and release of arsenic to the environment.

Who were our main partners in the study?

University of Waterloo.

Sommaire des recherches de l'INRE

Titre en langage clair

Assainissement des eaux souterraines contaminées par l'arsenic.

Quel est le problème et que savent les chercheurs à ce sujet?

L'arsenic présent dans les eaux souterraines provient de processus naturels ou de rejets anthropiques. L'ingestion d'eau souterraine contaminée par l'arsenic cause des tumeurs et d'autres problèmes de santé. On dispose d'une grande quantité de données sur l'arsenic dans les eaux souterraines de nombreuses régions du monde. Des études ont également été menées sur les mécanismes gouvernant la libération et le transport de l'arsenic dans les eaux souterraines, de même que l'assainissement des eaux contaminées par cet élément. Cependant, il reste bien des localités et des systèmes géologiques pour lesquels on en sait très peu au chapitre du transport de l'arsenic dans les eaux souterraines et de l'assainissement de celles-ci.

Pourquoi l'INRE a-t-il effectué cette étude?

La présente étude a été entreprise dans le but de rassembler de l'information sur diverses méthodes d'assainissement permettant de retirer l'arsenic des eaux souterraines. Il existe de nombreuses méthodes d'assainissement. L'application d'une méthode donnée exige que l'on connaisse les mécanismes géochimiques, propres au site, gouvernant le comportement de l'arsenic. Durant l'étude, nous avons examiné diverses méthodes d'assainissement. En outre, dans l'article, nous discutons en détail les processus géochimiques et physiques qui déterminent la libération et le transport de l'arsenic ainsi que la réduction des concentrations de cet élément. De nouvelles données recueillies à un site minier en Ontario nous permettent d'illustrer nos propos.

Quels sont les résultats?

La plupart des études sur le terrain portant sur l'élimination de l'arsenic sont propres au site considéré. Il est essentiel de comprendre la géochimie du site en question avant d'y appliquer une méthode d'assainissement. Les méthodes d'assainissement consistent habituellement à modifier des processus physiques ou chimiques. Cette intervention peut aggraver le problème initial; c'est pourquoi il importe de connaître parfaitement les processus géochimiques en jeu à un site donné avant d'y mettre en place un système d'assainissement. Nous fournissons des exemples montrant la nécessité d'avoir en main suffisamment de données sur le site avant de choisir une méthode d'assainissement.

Comment ces résultats seront-ils utilisés?

L'information présentée peut être utilisée afin de limiter les rejets d'arsenic, de manière à prévenir la contamination à grande échelle des sources d'approvisionnement en eau potable ainsi que la libération d'arsenic dans l'environnement.

Quels étaient nos principaux partenaires dans cette étude?

Université de Waterloo.

Chapter 12

Remediation of arsenic contaminated groundwater

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Abstract

Selection of a method for the remediation of arsenic contaminated groundwater requires knowledge of the physical hydrogeological setting, chemical composition of the groundwater and aquifer solids, and concentrations and forms of arsenic in the dissolved and solid phases. There are a number of new approaches for remediating groundwater contaminated with inorganic substances. These approaches typically involve manipulation of the geochemistry of the aquifer or contaminant plume such that the inorganic contaminant is either mobilized and extracted from the aquifer or stabilized to remove it from the water. Successful deployment of a technology depends on site-specific characteristics, the expense of the technology, including costs covering the initial infrastructure and ongoing maintenance costs, and the technical performance requirements to meet the desired remediation objectives. Hydrogeological and geochemical factors which require consideration in selecting a remediation system, and remediation technologies available for treating arsenic contaminated groundwater are described.

Keywords: groundwater, remediation, arsenic, barrier systems

Introduction

Groundwater contaminated with arsenic can be remediated by pumping the water from the aquifer followed by above-ground treatment, or it can be remediated *in situ*, minimizing the need for groundwater extraction and the construction of above-ground treatment facilities. Elevated concentrations of arsenic in groundwater can occur from anthropogenic discharges of arsenic, natural releases of arsenic, or a combination of natural and anthropogenic processes. Selection of a remediation technique requires knowledge of solid-phase sources and forms of arsenic and aqueous-phase concentrations and species of arsenic. An understanding of how these forms change over time as groundwater flows through the aquifer and as geochemical conditions change is also required. Information on the geochemistry of arsenic in the solid and aqueous phases, together with information on the hydrogeological and geochemical setting, are required to develop a site-specific remediation approach. Large advances in understanding of arsenic release, transport and cycling in groundwater have been made

Aquatic arsenic toxicity and treatment, pp. 139–157

Edited by T. Murphy & J. Guo

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over the last two decades. At the same time, new technologies have been developed for remediating groundwater, many of which can be applied to sites contaminated with arsenic. This paper describes processes controlling the release, transport and attenuation of arsenic in groundwater and various approaches available for remediating metal-contaminated groundwater, many of which can be applied for sites with arsenic contamination.

Background

Geochemical processes controlling the release of arsenic to groundwater

Arsenic occurs in a variety of aqueous and solid-phase forms in aquifers and at waste sites. Arsenic can be present in minerals including sulfides, oxides and hydroxides, incorporated directly in the crystal structure, or it can be associated with mineral surfaces through adsorption or coprecipitation reactions. Arsenic is released to groundwater as a result of natural processes, as a result of anthropogenic perturbations, such as excessive groundwater exploitation, or through the direct release of arsenic at waste sites or through intentional land application. Once in the groundwater, a series of reactions influence the transport of arsenic (Table 1). Selection of a groundwater remediation system requires knowledge of the processes leading to arsenic release to groundwater. A remediation approach for a system where the release of arsenic is expected to be short-term will be quite different than one selected for a system where the release is expected to be a long-term or ongoing process. This paper describes mechanisms controlling the release and attenuation of arsenic and options for remediating arsenic contaminated groundwater. The focus of the paper is on the remediation of distinct plumes of contaminated groundwater. There are locations where diffuse distributions of arsenic can be treated in a manner similar to distinct plumes. Transfer of the remediation technologies for distinct plumes for treating diffuse arsenic contamination is expected.

Natural release of arsenic

Arsenic is present naturally in a variety of geological materials, including hydrothermally influenced and volcanogenic materials or alluvium derived from arsenic rich

Table 1. Geochemical processes influencing the mobility of arsenic in groundwater.

Geochemical process	Example reaction
Sulfide oxidation and arsenic release	$\text{FeAsS}_{(s)} + 11/4 \text{O}_2 + 3/2 \text{H}_2\text{O} = \text{Fe}^{2+} + \text{SO}_4^{2-} + \text{H}_3\text{AsO}_3^0_{(aq)}$
As(III) oxidation coupled to reductive dissolution of oxide	$\text{MnO}_{2(s)} + \text{H}_3\text{AsO}_3 + 2\text{H}^+ = \text{Mn}^{2+} + \text{H}_3\text{AsO}_4 + \text{H}_2\text{O}$
Adsorption	$\text{SOH}_{(s)} + \text{H}_3\text{AsO}_3 = \text{SAsO}_3^{2-}_{(s)} + \text{H}_2\text{O} + 2\text{H}^+$
Sulfide formation	$2\text{H}_3\text{AsO}_3^0 + 3\text{H}_2\text{S} = \text{As}_2\text{S}_3_{(s)} + 6\text{H}_2\text{O}$

source areas. Arsenic can be bound in sulfide minerals, or a variety of other phases. Weathering of the source minerals, primarily through oxidation reactions, leads to the release of arsenic to flowing groundwater. Extensive sulfide oxidation occurs in the unsaturated zone of aquifers, where oxygen is plentiful. Below the water table, oxidation is less extensive because of the relatively low solubility of oxygen in water. There are numerous examples worldwide documenting the natural occurrence of arsenic in a variety of geological settings (Smedley and Kinniburgh 2002).

As the arsenic is released, a series of attenuation reactions can lessen the extent to which arsenic migrates from source areas. These reactions are highly dependent on the pH and oxidation-reduction processes in groundwater. Inorganic forms of arsenic predominate in groundwater, principally as As(III) and As(V) valence states. Inorganic As(III) and As(V) hydrolyse to form oxyanions or neutral species. The As(III) valence state occurs as H_3AsO_3 , H_2AsO_3^- , HAsO_3^{2-} species, with the neutral species H_3AsO_3 predominant below pH 9.2. The As(V) valence state occurs as H_3AsO_4 , H_2AsO_4^- , HAsO_4^{2-} and AsO_4^{3-} species, with the H_2AsO_4^- and HAsO_4^{2-} species predominant over the normal groundwater pH range. Other inorganic aqueous species such as the thioarsenate species ($\text{H}_2\text{AsO}_3\text{S}^-$) form under reducing conditions, but can be relatively short-lived (Rochette et al. 2000). Organoarsenic compounds, such as dimethylarsinic acid (DMA; $(\text{CH}_3)_2\text{AsO}(\text{OH})$) and monomethylarsonic acid (MMAA; $\text{CH}_3\text{AsO}(\text{OH})_2$), are less common in groundwater. A combination of adsorption and desorption reactions, mineral precipitation and dissolution reactions, and microbiologically mediated reactions control the mobility of these dissolved species.

Inorganic As(III) and As(V) species adsorb onto many mineral surfaces, including ferric and manganese oxyhydroxide solids [lepidocrocite, goethite (Farquhar et al. 2002), and birnessite (Manning et al. 1998, 2002)], clay minerals [kaolinite, illite, montmorillonite, and amorphous aluminum hydroxide (Manning and Goldberg 1997)], and sulfides [mackinawite and pyrite (Farquhar et al. 2002)]. Arsenic is generally more mobile under reducing conditions. This greater mobility is attributed to the weaker adsorption exhibited by As(III) species, and the loss of adsorption capacity as a result of abiotic and microbial reductive dissolution of iron and manganese (oxy)hydroxides.

Cummings et al. (1999) document the release of arsenic by the dissimilatory Fe(III)-reducing bacterium *Shewanella alga* BrY. As(III) and As(V) adsorbed to hydrous ferric oxide was released, and crystalline ferric arsenate solubilized, as a result of bacteria respiration processes. The increase in dissolved arsenic concentrations observed by Cummings et al. (1999) are attributed primarily to the decrease in adsorption capacity. Manning et al. (2002) document the oxidation of As(III) coupled to MnO_2 reduction, and observed that the adsorption of As(V) after oxidation from As(III) was greater than the direct adsorption of As(V) onto MnO_2 . The enhanced adsorption of As was attributed to freshening of the MnO_2 surface as Mn^{2+} is released into solution.

Many direct microbial reactions also affect arsenic release. As(V) can be reduced to As(III) through a dissimilatory reduction process, where As(V) serves as a terminal electron acceptor for microorganisms (Langner and Inskeep 2000). Here, the reduction of As(V) to As(III) was observed in the absence of reduction or dissolution of Fe(III) solid phase. At least seven bacteria and two hypothermophilic archaea can utilize As(V) as a direct terminal electron acceptor, as summarized by Macur et al.

(2001). Additional species reduce As(V) to As(III) through intercellular detoxification mechanisms. Macur et al. (2001) have suggested this latter pathway may operate in aerobic as well as anaerobic environments.

In groundwater systems, a number of geochemical processes can influence arsenic mobility. These processes depend on the aqueous geochemistry, aquifer mineralogy and microbial activity (Fig. 1). For example, under aerobic conditions, oxidation of arsenical sulfides can lead to the release of arsenic. The released arsenic can be attenuated by adsorption or coprecipitation with Fe(III) oxyhydroxides. Above the water table, oxidation of sulfides occurs readily. In the saturated zone, the rate of oxidation will depend on the transport of oxygen and other oxidants to the sulfide solids. Once released from sulfides and other reduced phases through oxidation processes, arsenic can adsorb to clay minerals and ferric and manganese oxyhydroxide solids. If groundwater containing arsenic encounters layers enriched in organic carbon, sulfate reducing conditions can develop, and arsenic can be restabilized through the formation of secondary sulfides, such as orpiment [As₂S₃]. These secondary sulfides can potentially reoxidize, leading to renewed release of arsenic. In a given aquifer, only one of these reactions might dominate, whereas in another aquifer, multiple reactions can occur over a very short distance. Detailed site characterization is required to fully delineate the extent that arsenic is released, transported from the source area,

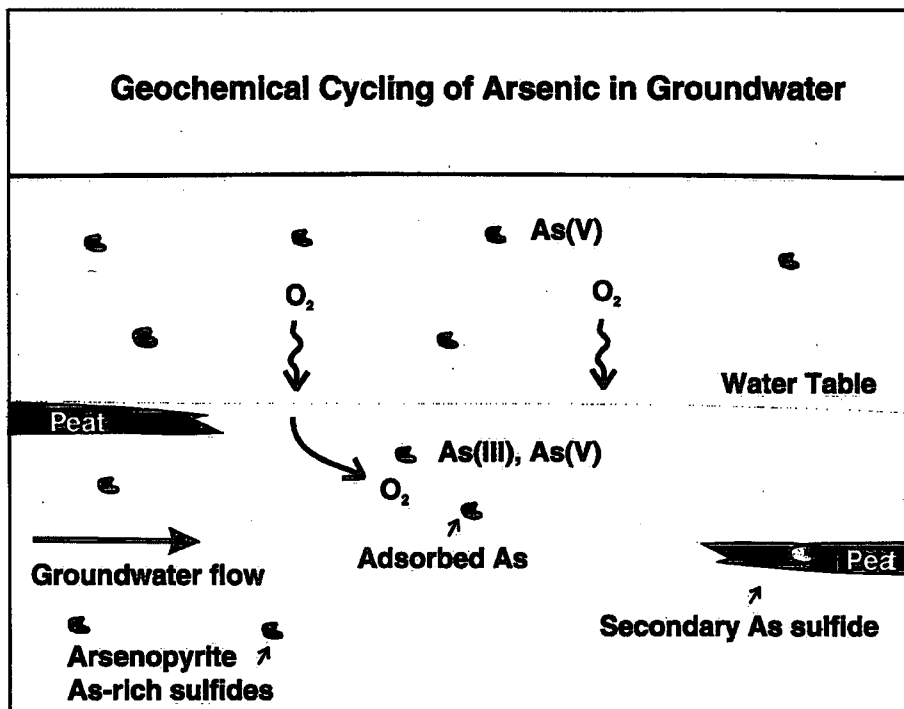


Fig. 1. Schematic diagram showing potential reactions that can occur at various locations in an aquifer.

and the extent that arsenic is restabilized through formation of secondary sulfides.

Cycling of arsenic in groundwater on local scales and on basin scales has been well documented. For example, in western Canada, arsenic occurs as arsenopyrite in hydrothermal veins within metasediments and metavolcanic rocks. Oxidation of the arsenopyrite has resulted in elevated concentrations of arsenic in the groundwater. A portion of the arsenic is adsorbed and later released, with highest concentrations observed at elevated pH conditions (Boyle et al. 1998). In northeastern United States elevated concentrations of arsenic in groundwater are attributed to arsenopyrite oxidation, with the highest concentrations observed at the edges of a large basin (Sidle et al. 2001). In Hanoi, Vietnam, arsenic is released to groundwater from sediments as a result of either reductive or oxidative processes (Berg et al. 2001). Concentrations of arsenic are elevated in a sandstone aquifer in Bavaria and are strongly associated with specific lithofacies (Heinrichs and Udluft 1999). In Mexico, arsenic release to groundwater is attributed to arsenopyrite oxidation, scorodite dissolution and desorption reactions (Planer-Friedrich et al. 2001; Morse 2001). In Bangladesh, arsenic release has been attributed to a number of geochemical processes, including reductive dissolution of Fe and Mn oxyhydroxides and microbial oxidation of organic matter (e.g. Mandal et al. 1996; Anawar et al. 2002). These, and other examples, demonstrate the wide range of geochemical processes that can be active under natural conditions in aquifers.

Release of naturally occurring arsenic through anthropogenic perturbations

An important step in establishing a remediation program is to determine the geochemical processes leading to the release of arsenic. Anthropogenic activities can alter geochemical conditions in aquifers and consequently enhance the release of arsenic to groundwater (Figs. 2, 3). These actions can lead to the development of distinct plumes of arsenic contaminated groundwater, which may require remediation, or larger-scale arsenic contamination beyond the scale that can be economically remediated. An example of an anthropogenic activity that can lead to diffuse arsenic contamination of groundwater is the excessive extraction of groundwater leading to the development of drawdown cones and enhanced oxidation of naturally occurring arsenic sulfides and other reduced arsenic phases (Fig. 2). An example of an activity where distinct plumes of arsenic contamination can develop is the unintentional or intentional release of reducing water to the subsurface such as from waste disposal sites or municipal landfills (Fig. 3). The release of reducing water can promote the dissolution of iron and manganese oxyhydroxide minerals, liberating adsorbed or coprecipitated arsenic. If left uncontrolled, extensive groundwater contamination and degradation of drinking water supplies can ensue. Through similar geochemical mechanisms, induced infiltration of organic rich river waters or agricultural waters can cause the release of arsenic (Fig. 3). Extraction of groundwater from wells installed adjacent to rivers or lakes can lead to the ingress of water containing elevated concentrations of organic matter. Similarly, increased flooding, due to widespread deforestation and other anthropogenic perturbations, can result in the deposition of organic rich sediment above vulnerable aquifers. The quality of groundwater that is extracted

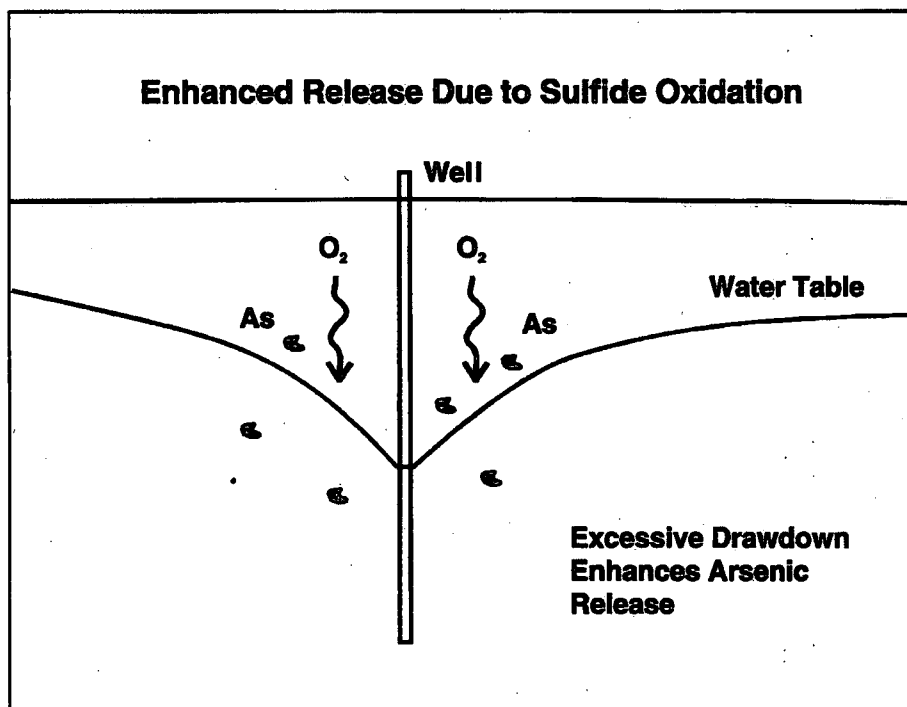


Fig. 2. Schematic diagram showing enhanced oxidation of sulfides, such as arsenopyrite or arsenian pyrite, as the result of drawdown of the water table around a well field.

from these aquifers can degrade due to enhanced release of arsenic from the infiltration of dissolved organic matter.

Anthropogenic releases

Metal-mining environments

Many ore deposits, in particular gold-bearing ore deposits, contain elevated concentrations of arsenic in the ore and host rock. Arsenic, commonly in the form of arsenopyrite [FeAsS], or arsenian pyrite [(Fe,As)S₂], requires management during and after mining and ore processing. Elevated concentrations of arsenic can occur in effluent discharging from mine workings. Arsenic concentrations observed in the discharge portal at the Iron Mountain site, California are up to 340 mg/L (Nordstrom et al. 2000). Waste rock with elevated concentrations of As can be generated during ore production. Arsenic can also be released in heap-leach operations or refining processes. Heap-leach operations include leach pads containing crushed ore through which leaching fluids are circulated. Although these pads are lined, to assure collection of

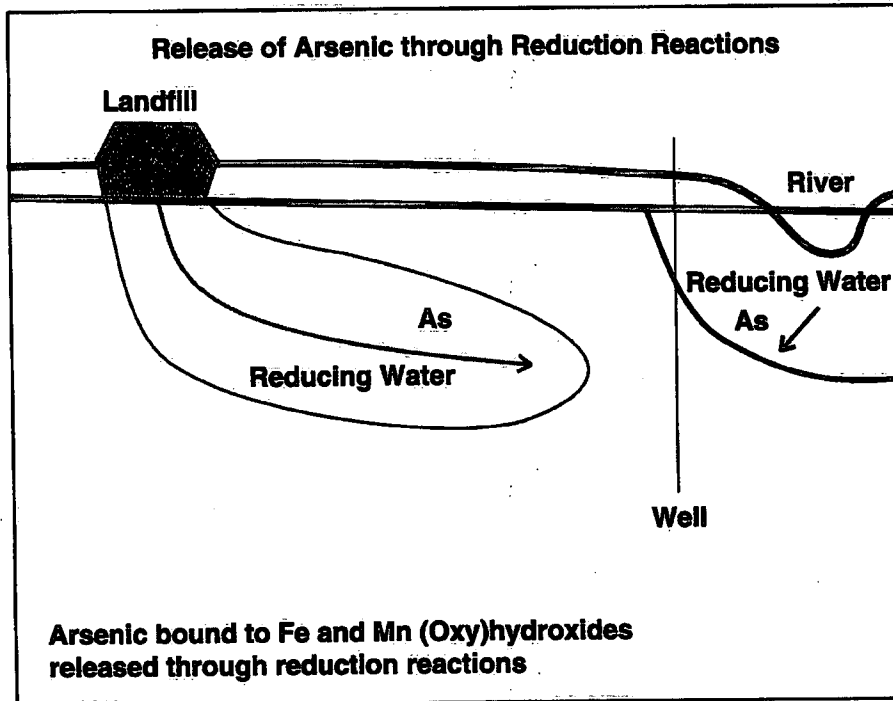


Fig. 3. Enhanced release of arsenic in response to the ingress of water containing high concentrations of dissolved organic matter.

the leaching solution, arsenic may escape during operation or after active heap leaching ends and fluid circulation ceases.

In conventional gold recovery operations the ore is finely ground. The gold-bearing fraction of the ore is separated by gravitational settling or differential flotation. Once separated, refining techniques, including roasting or pressure oxidation using autoclave methods are used to recover gold. Mill tailings, the residue from the concentration process, have little commercial value and are typically disposed of in dedicated impoundments. Refinery wastes are disposed of in separate impoundments, or co-disposed with the tailings. As waste rock, mill tailings and refinery wastes weather, elevated concentrations of arsenic can be leached from the wastes and released to groundwater.

The rate of arsenic leaching from mine wastes depends on a variety of factors, including the hydrological, and geochemical conditions that prevail in the waste disposal facility, and the form of the arsenic in the wastes (Fig. 4). Arsenic can be present in the wastes in reduced forms or oxidized forms. In flotation tailings derived from sulfide ore bodies arsenical sulfides, including arsenopyrite and arsenian pyrite may be abundant. In oxidized waste rock piles, oxidized tailings impoundments, or in impoundments containing oxidized residues such as autoclave wastes, arsenic bearing iron and manganese oxides may be abundant.

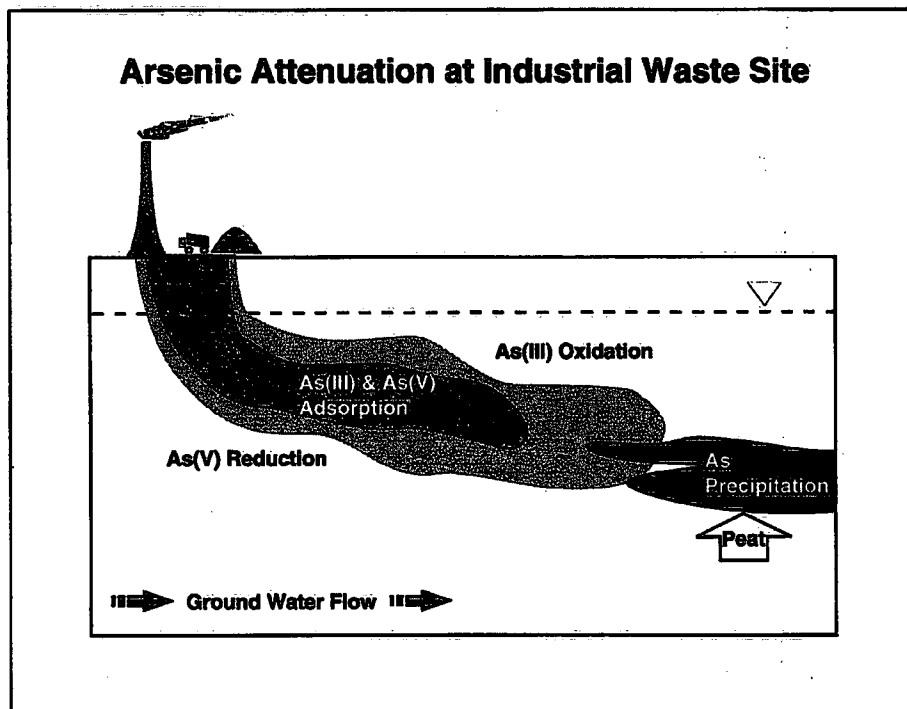


Fig. 4. Industrial release of arsenic and processes influencing arsenic transport.

Release of arsenic from mine waste sites has been well documented. Arsenic release can occur directly from waste rock piles and tailings impoundments (e.g. McCreadie et al. 1998, 2000; Ross et al. 1999), and from wastes that have spilled into rivers and lakes (e.g. Cherry et al. 1986; Macur et al. 2001). Al et al. (1994) report the presence of high concentrations of arsenic in the porewater of the Kidd Creek Mine tailings impoundment, near Timmins, Ontario, Canada. The production of Zn from sulfide ore includes the removal of iron from the zinc-bearing solutions through the formation of a jarosite residue which is rich in arsenic. The jarosite residue contains jarosite $[KFe_3(SO_4)_2(OH)_6]$, sodium jarosite $[NaFe_3(SO_4)_2(OH)_6]$, hydronium jarosite $[H_3OFe_3(SO_4)_2(OH)_6]$, goethite $[\alpha FeOOH]$, and lepidocrocite $[\gamma FeOOH]$. This jarosite residue is co-blended with sulfide-rich flotation tailings and discharged to a conical pile within the tailings impoundment. Dissolution of the jarosite residue is favoured under the geochemical conditions prevalent in the tailings cone. As dissolution occurs, arsenic contained within the jarosite residue is released to the tailings porewater.

At the Campbell Au mine, Red Lake, Ontario, roaster wastes produced during the production of gold are deposited with the tailings. Studies conducted at this site indicate that arsenic-bearing gold-refinery wastes are unstable when blended with sulfidic tailings (McCreadie et al. 2000). Arsenic bound to the roaster wastes is released to tailings porewater and subsequently migrates from the wastes into an un-

derlying sand aquifer (Ross et al. 2000). The arsenic rich plume is migrating from the tailings pile at a rate of approximately one-half of the groundwater velocity (Ross et al. 1999). At some locations where peat layers are present at the base of the tailings impoundment, arsenic is stabilized as an arsenic rich sulfide, with a formula consistent with orpiment (Stichbury et al. 2000a,b).

The release and attenuation of arsenic at the Campbell Au mine are complex. At the surface of the impoundment in the vadose zone, oxidation of arsenopyrite releases dissolved arsenic. Above the peat layer, reducing conditions enhance the release of arsenic through the reductive dissolution of arsenic rich ferric iron minerals coupled with the oxidation of dissolved organic carbon. Within the peat layer, in the absence of Fe(III) oxides, sulfate reduction coupled with the oxidation of organic carbon results in the precipitation of arsenic sulfides. At this tailings site, a number of geochemical processes are occurring at different localities. Selection of a remediation approach for this site requires consideration of the processes controlling arsenic release and transport within the tailings, within and underlying the peat layer and within the adjacent aquifer. Selection of an inappropriate remediation system could promote the release of arsenic resulting in an increase in concentrations rather than the desired reduction in arsenic concentrations.

Arsenical pesticides and other industrial releases

Other sources of arsenic can lead to distinct plumes of arsenic contaminated groundwater. A number of industrial activities involve direct handling of arsenic bearing solids and solutions, resulting in contamination of groundwater. Example sites where elevated concentrations of arsenic are present in groundwater at sufficient concentrations to require remediation include tannery sites (Davis et al. 1994), and pesticide manufacturing sites (Mariner et al. 1996).

Remediation of arsenic contaminated groundwater

There are two main settings where remediation of arsenic contaminated groundwater is undertaken. In the first setting, treatment of groundwater is required prior to distribution of arsenic free drinking water. In the second setting, concentrations of arsenic are elevated due to anthropogenic releases. For this setting, treatment of arsenic contaminated groundwater is carried out to prevent widespread contamination of aquifers and discharge of arsenic-contaminated groundwater to surface water bodies.

Remediation of arsenic contaminated groundwater can take place *in situ* or *ex situ*. If arsenic free drinking water is required, the conventional approach to providing a water supply is to pump groundwater to the ground surface followed by treatment. Above-ground treatment approaches range from small hand-held devices (e.g. Sengupta et al. 2001) to large treatment plants. Treatment options include adsorption based systems, reverse osmosis systems and precipitation based systems. The selection of an appropriate treatment system will depend on scale of the treatment

Table 2. Summary of approaches to remediation of inorganic contaminants in groundwater, including potential advantages and disadvantages in their application.

Remediation method	Advantages	Disadvantages
Pump-and-treat	Treated water can potentially be used Contaminant is removed from aquifer	High initial capital costs Ongoing operating costs Long treatment times Energy intensive Treated water requires disposal Treatment byproducts (sludge) require disposal
Enhanced pump-and-treat	Contaminant is removed from aquifer Potential for long-term (positive) changes in aquifer properties	High initial capital costs Ongoing operating costs Difficulties in delivering reagents Aquifer properties changed in undesirable manner Treated water requires disposal Deleterious effects of reagents
In-situ stabilization: injection systems	Contaminant is stabilized in aquifer - cannot be pumped to wells or released to surface waters Potentially low capital costs	Contaminant is not removed from aquifer - potential for remobilization Difficult to deliver reagents Ongoing operation costs Reduction in aquifer permeability Deleterious effects of reagents
In-situ stabilization: permeable reactive barriers	Contaminant stabilized in aquifer No ongoing maintenance costs No ongoing energy costs No disposal of treated water	High initial capital costs Not suitable for deep systems Media may require replacement Contaminant not removed from aquifer

system or plan, the composition of the groundwater and time available for treatment.

At an industrial site undergoing remediation, pumping of groundwater to ground surface may or may not be required. In this case, either *ex situ* or *in situ* groundwater remediation techniques can be applied. As in drinking water treatment systems, selection of an effective remedial approach requires consideration of the scale of the remediation project, the groundwater flow rates, and the composition of the groundwater. For *in situ* treatment systems, information on the composition of the aquifer solids is also required.

Groundwater remediation systems have been developed for a broad range of anthropogenic contaminants, including organic solvents, metals, nutrients, radionuclides, pesticides, and more recently arsenic and selenium. Large advances in implementing and operating full-scale groundwater remediation systems have been made over the past few decades (Table 2). Many of these advances can be applied for treating arsenic contaminated groundwater, including both *in situ* or *ex situ* approaches.

Pump-and-treat systems for remediating arsenic in groundwater

Traditionally, single wells or a series of wells are installed into or adjacent to the contaminant plume, and groundwater is pumped to ground surface where it is treated

(Fig. 5a). This approach, pump-and-treat, has been demonstrated to be effective for a limited number of contaminated sites. For the majority of sites, it is increasingly recognized that the conventional pump-and-treat approach is limited by a combination of physical and geochemical processes (Mackay and Cherry 1989; Blowes 2002). These processes result in the retention of small masses of contaminants within the aquifer either as a discrete phase or as dissolved constituents within fine-grained portions of the aquifer. As a consequence, long operating times are expected. In many cases the contaminants can be contained but complete aquifer remediation cannot be achieved in a reasonable length of time (Mackay and Cherry 1989).

Applications of pump-and-treat systems to remove arsenic from contaminated groundwater have been documented. For example, Mariner et al. (1996) describe a pesticide manufacturing site where a pump-and-treat system is operating to extract arsenic contaminated groundwater ($As > 500$ mg/L) to prevent release to tidal flats. At this site, the groundwater pH is elevated and adsorption reactions limited. Arsenic, therefore, is likely available for release by pumping. For most arsenic contaminated sites, long treatment times are expected because of the ongoing release of arsenic that is associated with the aquifer solids. In areas where arsenic occurs naturally, arsenic may be pumped from aquifers at unacceptable concentrations for many volumes of water (years of pumping) with no apparent diminishment in concentrations observed. For a contaminated site, the release of arsenic can also potentially last for many years, suggesting that long pumping times would be required to meet site remediation goals. In addition to information on physical hydrogeological setting, detailed knowledge of the distribution and form of arsenic in the aquifer solids, and geochemical and microbiological processes is required. Operation of treatment systems for decades or longer can be expected.

Enhanced pump-and-treat systems

The use of enhanced pump-and-treat systems is becoming more widespread for remediating a variety of contaminated sites, including sites contaminated with organic solvents and those contaminated with metals. These systems rely on delivery of a reagent to the subsurface through the use of injection wells (Fig. 5b). The reagent promotes the transfer of the contaminant in the solid-phase to the aqueous phase to shorten the duration of pumping. For arsenic, there are a variety of reagents that potentially could be used to promote the release of arsenic from the solid-phase into the solution phase. For example, reagents such as phosphate-bearing solutions to promote arsenic desorption, or weak acids or a reductant to promote dissolution of adsorbing phases and indirect release of arsenic, could potentially be used in an enhanced pump-and-treat system. Selection of the reagent requires an initial understanding of the form and distribution of arsenic on the solid phase. Peters et al. (1994) describe the potential use of sodium carbonate, potassium phosphate (-dibasic), citric acid, oxalic acid, phosphoric acid, polysodium vinyl sulfonate and three commercially provided reagents for arsenic extraction in enhanced pump-and-treat systems (in Redwine 2001). Citric acid was observed to remove up to 100 times more arsenic from soil in field applications than could water alone (Redwine 2001). Enhanced pump-and-treat systems could be utilized at sites where a spill of arsenic occurred and removal of ar-

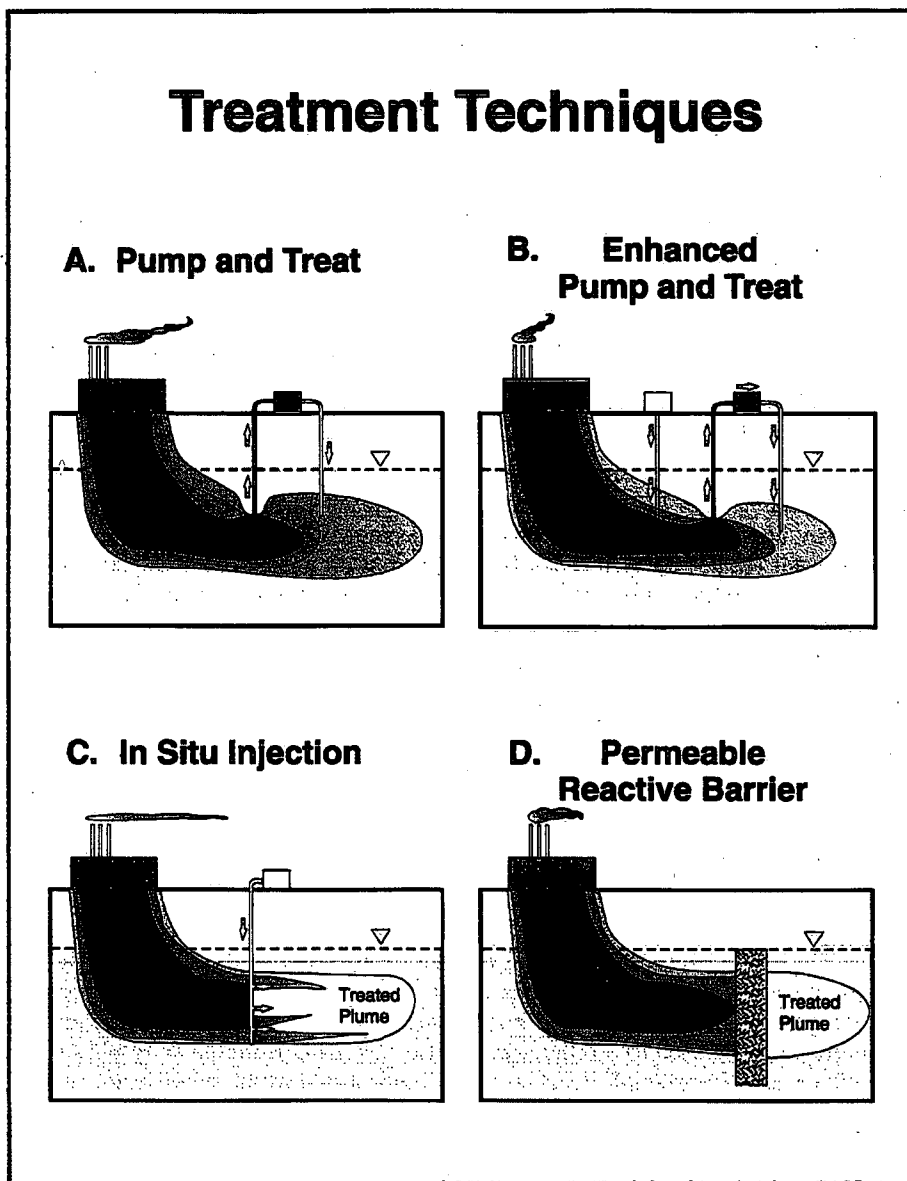


Fig. 5. Schematic diagram showing a) pump-and-treat system and b) enhanced pump-and-treat system, c) in-situ stabilization system and d) permeable reactive barrier system for remediating groundwater contaminated with arsenic.

senic from the site is required. Because of the number of potential reactions that could take place as chemicals are injected, bench- and small field-scale trials would be required.

In-situ stabilization treatment systems

Alternative treatment approaches have been developed and implemented which promote the stabilization of the contaminant *in situ* to prevent its release into flowing groundwater. To stabilize contaminants, reagents can be injected through wells as solutions (Fig. 5c) or as nanoparticles, or solid-phase reactants can be added by auger mixing, jetting or excavation and replacement techniques (Fig. 5d). The reactants can impede groundwater flow, such as reactive grouting (also referred to as soil stabilizers) or the groundwater can be allowed to flow through the reactant to promote contact between the contaminant and the reactant. If the groundwater is allowed to flow through the reactant, the installation is referred to as a permeable reactive barrier (Blowes and Ptacek 1992, 1994, 1996; Blowes et al. 2000).

Solidification/stabilization techniques use the addition of a binding agent to solidify the soil or waste and render the contaminants immobile (Redwine 2001). Binding agents include portland cement, or cement with chemical additives that decrease the leachability of contaminants. Redwine (2001) describes the application of soil solidification/stabilization techniques to two field sites. At both field sites, a ferrous sulfate slurry was added to the soils followed by a cement slurry 24 hours later. Following post-treatment testing of both sites, regulators declared "no further action" was required (Redwine 2001).

Permeable reactive barriers have been demonstrated to successfully treat a range of inorganic contaminants, including dissolved metals, arsenic and selenium, radionuclides, nitrate and phosphate (Blowes and Ptacek 1994, 1996; Blowes et al. 2000). Aquifer material is excavated and replaced with a reactive media. Permeable reactive barriers are approximately 1 m wide, up to 20 m deep and of sufficient length to intercept the plume (10's to 100's of meters). Groundwater is allowed to flow passively through the barrier, typically for many decades, to provide ongoing treatment in an energy efficient and cost-effective manner. Auxiliary pumping can be added to direct water through the reactive mixture if desired.

Selection of the reactive media will depend on the target contaminant, the groundwater chemistry and the groundwater velocity. For example, zero-valent iron has been used in permeable reactive barriers to treat Cr(VI) contaminated groundwater (Blowes et al. 1997, 1999a,b). As Cr(VI)-bearing groundwater flows through the zero-valent iron, the Cr(VI) is reduced to Cr(III) and removed as insoluble hydroxides and oxide, thus preventing its migration. Once stabilized, Cr(III) is no longer released to flowing groundwater at concentrations of concern. Organic carbon containing permeable reactive barriers have been described by Blowes and Ptacek (1994, 1996) and Blowes et al. (1994) and installed at large industrial sites for treating metal-contaminated groundwater by promoting sulfate reduction and metal sulfide formation. A variety of organic carbon sources, including municipal compost, wood chips and pulp waste were effective at lowering metal and sulfate concentrations and increasing alkalinity in an aquifer receiving mine drainage (Waybrant et al. 1998, 2002; Benner et al. 1997, 1999, 2002). Using a similar mixture, McGregor et al. (2002) and Ludwig

et al. (2002) describe the removal of metals to very low concentrations at an industrial facility. Baker et al. (1997, 1998) describe the use of steel foundry wastes and aluminum oxides to treat phosphate contaminated groundwater. A comparison of costs associated with a pump-and-treat system and a permeable reactive barrier indicates that permeable reactive barriers can yield large cost savings over conventional pump-and-treat systems (Blowes 2002).

For groundwaters containing arsenic and selenium, a number of reactive solids have been demonstrated to effectively lower concentrations to below water quality guidelines. These materials include zero valent iron (Blowes and Ptacek 1994, 1996; McRae et al. 1999; Su and Puls 2001a,b; Melitas et al. 2002; McGregor et al. 2002), activated alumina (McRae et al. 1999) and other lower cost materials, including blast oxygen furnace oxides produced at steel foundries (Blowes et al. 1996; McRae et al. 1999). McRae et al. (1999) observed that materials suitable for arsenic removal were also effective at removing selenium from groundwater. In column experiments, concentrations of arsenic were below detection after flowing through a column containing 10 wt % zero valent iron for more than 800 pore volumes of flow (Fig. 6b), and through a column containing 20 wt % of activated alumina for more than 600 pore volumes of flow (Fig. 6a). Concentrations of arsenic declined from 1000 $\mu\text{g/L}$ to <20 $\mu\text{g/L}$ (the analytical detection limit) using 10 wt % blast oxygen furnace oxides for more than 100 pore volumes of flow at typical groundwater velocities (Fig. 6c). Permeable mixtures containing higher percentages of reactive media remove arsenic for even longer times (Bain et al. 2002).

McGregor et al. (2002) describe the effective removal of arsenic using a permeable reactive barrier at a field site in northern Ontario using reactive mixtures containing zero valent iron and organic carbon. The results of the study by McGregor et al. (2002) show removal of in excess of 90% dissolved arsenic within the reactive barrier during the first three months of operation.

Selection of a reactive media would be based on site-specific conditions. Highly reactive and permeable mixtures would be required for sites with high groundwater velocities. Mixtures containing zero valent iron and organic carbon would be effective at removing arsenic from oxidizing to moderately reducing groundwater flow systems. Activated alumina would provide effective treatment in a wide range of geochemical conditions, but is typically very expensive to utilize at the scales of most groundwater plumes. Basic oxygen furnace oxide contains a mixture of Fe(II) and Fe(III)-bearing solids. There is potential that this material might not be stable under prolonged exposure to very high concentrations of dissolved organic carbon. Permeable reactive barriers containing zero valent iron result in large increases in groundwater pH. If groundwater requiring treatment also contain high concentrations of Ca, Mg and HCO_3 , precipitation of carbonates in the barrier can occur, potentially impeding flow through the barrier and decreasing reactivity leading to ineffective treatment. The costs and treatment capacity of the activated aluminum, zero valent iron, steel foundry wastes and organic carbon vary widely, therefore detailed cost analyses and bench-scale or field trials are an important aspect in the development of effective full-scale installations.

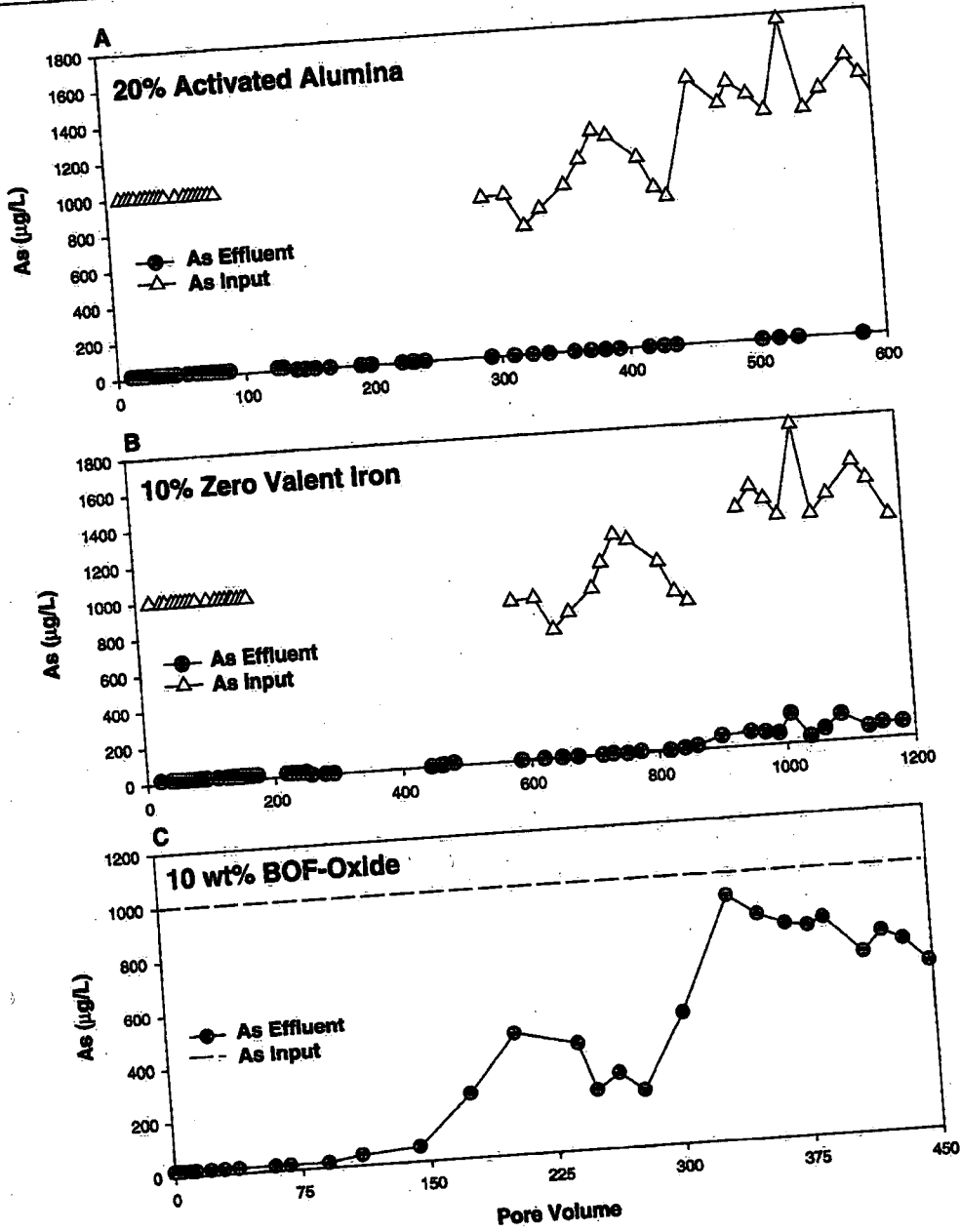


Fig. 6. Treatment of arsenic using permeable reactive media. Results of flow-through column experiments using a) activated alumina, b) zero valent iron, and c) blast oxygen furnace oxide (after McRae et al. 1999 and Bain et al. 2002).

Conclusions

Groundwater concentrations of arsenic can vary both temporally and spatially due to natural and anthropogenic factors. Site specific information on mineralogical, geochemical and hydrogeological factors is required to select an effective remediation strategy. Geochemical conditions in the subsurface can be manipulated to enhance the extraction or stabilization of arsenic. A number of approaches have been developed for remediating arsenic at industrial sites. These technologies can potentially be transferred to treat arsenic in aquifers used for drinking water supplies, provided that no toxic byproducts remain in the water supply. The variations in cost of different remedial options are large, indicating that detailed cost analyses for specific aquifers will be required. Arsenic release from natural sources and industrial sites can occur over long periods of time. Selection of a remediation approach should be consistent with the duration of the source term. For example, permeable reactive barriers are typically designed for several decades of passive operation, after which installation of fresh reactive material may be required. If the source is expected to be of long duration, a technique such as permeable reactive barriers would be suitable. An industrial release may lead to the release of very high concentrations of arsenic to the subsurface. Removal of a large mass of arsenic could be accomplished using a pump-and-treat system or enhanced pump-and-treat system. Following this initial removal, a stabilization technique or long-term passive technique could be used for the residual arsenic. A combination of techniques could provide a cost effective approach for long-term control.

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