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Evaluating the sustainability of methods for mitigation of arsenic contaminated aquifers

Évaluation des méthodes de mitigation des aquifères contaminées à l'arsenic dans un contexte de développement durable

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ABSTRACT

Fresh water is a precious, non-renewable resource that is depleting rapidly in countries such as India, China, and even in the U.S. where rivers are drying up and water table levels are decreasing. To obtain safe drinking water, two direct means are available. These include: (1) treatment of the water as it leaves the pumps (from the tube wells), and (2) application of in-situ geochemical aquifer intervention procedures designed to control and mitigate arsenic pollution. By reducing the concentrations to levels below the critical levels, the risk of arsenic poisoning of the people using the groundwater resource will be minimized or eliminated and thus ensuring the sustainability of groundwater for drinking purposes.

RÉSUMÉ

L`eau douce, cette ressource précieuse et non-renouvelable, diminue rapidement dans les pays comme l'Inde, la Chine et même les États-Unis; les rivières se dessèchent et la nappe phréatique s'abaisse. Pour obtenir de l'eau potable, deux moyens directs sont disponibles, soit (1) le traitement de l'eau des pompes des puits et (2) l'application des techniques in situ d`intervention géochimique dans l'aquifère, établies pour le contrôle et la mitigation de la pollution de l`aquifère. La réduction des niveaux de contaminants en-dessous des niveaux critiques assure la diminution ou l'élimination du risque d'empoisonnement pour la population utilisant cette ressource souterraine comme eau potable.

1 INTRODUCTION

Fresh water is a precious, non-renewable resource and is depleting rapidly in countries such as India, China, and even in the U.S. where rivers are drying up and water table levels are decreasing. Arsenic polluted aquifers provide good examples of a combined "man and nature" impact on the geoenvironment. Arsenic is released into the groundwater from arsenic-rich sulfide ores by either oxidation or reduction processes. It is a toxic element that should not exceed the WHO recommended limit of 10 micrograms per liter. Several million people are affected by arsenic contaminated groundwater.

All through the late 1970s and 1980s there was a move forward to reduce the serious mortality, predominantly among infants, caused by diseases in surface waters in Bangladesh and other countries. International aid agencies such as UNICEF became involved in funding the drilling of shallow tubewells to gain access to groundwater for domestic supply which was uncontaminated by bacteria and otherwise believed to be clean. More than a million such wells were constructed and it was greatly successful in reducing child death in Bangladesh.

Although arsenic is a widely distributed element in the earth's crust, it was not generally found in a water-soluble form and thus does not cause a risk to the safety of drinking water supplies (Smedley and Kinniburgh, 2002). Arsenic problems have long been recognized to occur in sulphide-rich metaliferous strata (principally, therefore, in particular mining areas) and in some geothermal areas. There were also reports in the literature of arsenic occurring in some arid or semi-arid inland basins, for example in parts of Argentina and the United States but the existence of arsenic in soluble form in the anaerobic groundwater of alluvial and deltaic plains was not by and large recognized until 1995. It is, however, the most common contaminant found in Superfund, Department of Defense and Department of Energy sites. Arsenic-containing minerals include arsenopyrite (FeAs), realgar (AsS), orpiment (As_2S_3), niccolite (NiAs) and cobalite (CoAsS) (Boyle and Jonasson, 1973).

Arsenic is a semi-metallic contaminant that originates from and is transported to natural waters through erosion and dissolution of arsenic-containing rocks and soil. Climate and redox potential are significant factors in the transport of arsenic. Arsenic sulfides can be oxidized, releasing arsenic to the environment. Arsenic occurs in natural waters in both organic and inorganic forms but the inorganic arsenic is the most prevalent and is the most likely to exist in significant concentrations. The valence and species of inorganic arsenic are dependent on the oxidation-reduction conditions and the pH of the water. Environmental arsenic is mainly found in two forms: arsenic V (arsenate), and arsenic III (arsenite). Arsenic V is the common oxidized state found in surface water and some groundwater sources and precipitates in the presence of metal ions. Arsenic III is unoxidized and mainly found in deep anaerobic groundwater sources. Under acidic and slightly reducing conditions, arsenic co-precipitates with iron oxyhydroxides. Increases in pH can mobilize the arsenic.

To obtain safe drinking water from the tube wells, two direct means are available. These include: (1) treatment of the water as it leaves the pumps (from the tube wells), and (2) application of in-situ geochemical aquifer intervention procedures designed to control and mitigate arsenic pollution. Procedures for direct treatment of the arsenic-polluted water are presently being applied and others are also under development. Some of the issues involving the impact of human activity on the geoenvironment are illustrated in Fig. 1.

The techniques to control, or significantly reduce the release of arsenic at the source points in the aquifers will be examined in this paper and compared to groundwater treatment methods such as ion exchange to compare the sustainability of the various procedures for dealing with arsenic mitigation. A sustainable environment for society must be safe from metal pollution.

Figure 1. Land environment issues and impact of various activities on the geoenvironment (adapted from Yong and Mulligan, 2004).

2 EX SITU TREATMENT OF GROUNDWATER

Some of the procedures for water treatment designed to capture the arsenic in the water when it is delivered from the tube wells include coagulation/filtration, activated alumina, reverse osmosis, electrodialysis reversal, ion exchange, and a whole host of other processes. To evaluate the sustainability of these methods, several factors including materials, energy, transportation and waste management requirements for the treatment process need to be taken into consideration. One of the principal methods is ion exchange. However, removal of arsenic for As(III) is not sufficient by ion exchange alone. Oxidation of this form to As(V) must be required and performed with a preoxidation filter. Although this method is highly efficient, disposal of a toxic arsenic waste from the regeneration of these filters and the ion exchange resins as a result of the water treatment procedures impacts the sustainability of these processes as these activities generate significant wastes that can severely impact the environment, causing more harm than good. Due to the problems of arsenic in the groundwater, economic solutions need to be found to ensure the safety of the drinking water. Several common treatment technologies are used for removal of inorganic contaminants, including arsenic, from drinking water supplies. Large-scale treatment facilities often use conventional coagulation with alum or iron salts followed by filtration to remove arsenic. Lime softening and iron removal also are common, conventional treatment processes that can potentially remove arsenic from source waters. In small communities small-scale systems often use ion exchange adsorption because of their ease of handling and sludge-free operations (Clifford, 1999). Treatment options identified by EPA include ion exchange, reverse osmosis, activated alumina, nanofiltration, electrodialysis reversal, coagulation/filtration, lime softening, greensand filtration and other iron/manganese removal processes, and emerging technologies not yet identified (USEPA, 2003).

In West Bengal, purification units can be utilized for treatment of the well water. The units cost $14 US, in excess of a farmer's monthly wage (Siegel, 2002). Treatment facilities or alternative water sources are required. We can see that this treatment method, along with the others does not eliminate the arsenic source before delivery and that other methods must be examined to accomplish this. Other groundwater methods such as oxidation can lead to the formation of toxic by-products and sludge, which require disposal.

3 MANAGEMENT OF ARSENIC CONTAMINATION

3.1 Retention and release of arsenic

Recently, Twelve Principles of Green Engineering have been suggested to engineers as a way to improve the sustainability of processes (Anastas and Zimmerman, 2003). The second principle is particularly relevant which says that "It is better to prevent waste than to treat or clean up waste after it is formed". Therefore, in this case more sustainable measures need to be structured to arrest the production and delivery of arsenic in the ground itself as complete elimination or removal of the arsenic in the aquifers is not feasible or practical as shown by ion exchange processes. The release of arsenic in the groundwater provides a continuous pollutant source. Managing the pollution once it has occurred is highly complex and thus it is preferable to avoid the pollution from the beginning.

Two models exist in respect to possible mechanisms for release of arsenic from the arsenic-bearing materials: (a) reduction mechanisms, and (b) oxidation processes. In the former process, it is reasoned that reductive dissolution of arseniferrous iron oxyhydroxides releases the arsenic responsible for pollution of the groundwater. The other model for arsenic release from the alluvium relies on oxidation of the arsenopyrites as the principal mechanism. This occurs when oxygen invades the groundwater because of the lowering of the groundwater from the abstracting tubewells. Changes in the redox conditions by dredging of sediments can also affect the release of arsenic from wastes. For example, an experiment by Forstner (1996) showed that arsenic could be released from an industrial solid waste oxidatively at pH 5 over a period of 5 weeks. From 20 to 90% of pyrite-bound arsenic could also be released in periods up to a day under oxidative conditions in sea water. Fe(III) respiring bacterial strains such as *Sulfurospirillum arsenophilum* and *Sulfurospirillum barnessii* can also release arsenic by transformation of As(V) to As(III). Production of organic acids by *Thiobacillus* can also extract arsenic. *Penicillium* methylate monomethylarsonic acid to trimethylarsenine, which is highly volatile. Another species, *Pseudomonas arsentioxidans,* can oxidize arsenite. In general, oxidation and reduction of the arsenic is not a sustainable remediation method as the product is toxic. A better understanding of the biological reactions of arsenic with sorption phenomenon may enable natural attenuation to be utilized but the level of understanding is currently low (Yong and Mulligan, 2004).

Sorption-desorption of As(V) to iron oxide isotherms were non-linear (Williams et al., 2003). The factors of pH, pore velocity and then phosphate, demonstrated increasing influence on arsenic mobility. Therefore, the fate and transport of arsenic within the soil environment must be thoroughly understood as the soil properties have a tremendous influence on sorption of arsenic.

3.2 Remediation technologies

3.2.1 Extraction methods

Remediation technologies for arsenic-contaminated soils can be divided into *in situ* and *ex situ* technologies. These are summarized in Fig. 2. Washing is a method for remediating arsenic-contaminated soils by using solutions to extract the arsenic from the soil. Legiec et al. (1997) found that washing with alkali or acidic solutions enhanced arsenic removal. However, the strong acids or bases can destroy the soil iron oxides due to the extreme pH used. High pH solutions can also leach organic matter.

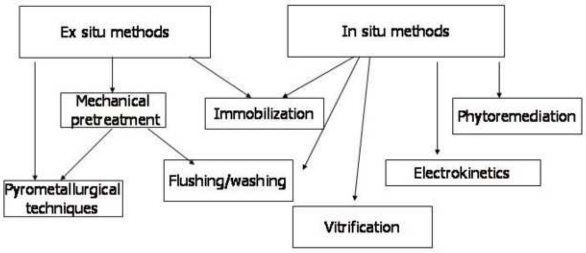

Figure 2. Remediation technologies for arsenic-contaminated soils

A different approach was used by Tokunaga and Hakuta (2002) who showed that acid washing with sulfuric or phosphphoric acid removed As(V) and that damage to the soil was limited if the treatment was less than 2 h. Addition of lanthanum, cerium and iron(II) oxides immobilized arsenic so that less than 0.01 mg/L leached from the soil. Therefore, it was deemed that removal of the bulk amount of arsenic before immobilization would be an optimal strategy.

Alam et al. (2001) determined that 40% of the arsenic could be removed by 0.9 M phosphate. They showed that potassium phosphate (pH 6 to 8) can be an inexpensive and environmental friendly method of extracting arsenic from Al- and Fe-bound forms as shown by sequential extraction. This is illustrated in Fig. 3. Residual arsenic was not extracted. Increasing the temperature also increased the extraction.

Several researchers have examined electrokinetics for decontaminating arsenic-contaminated soils. Panaytova and Zabchev (1998) determined that arsenic was not mobilized by electrokinetics. However, Sidoli O'Connor et al. (2003) found that the combination of electrokinetics and phytoremediation could remove 230 μg/g of arsenic.

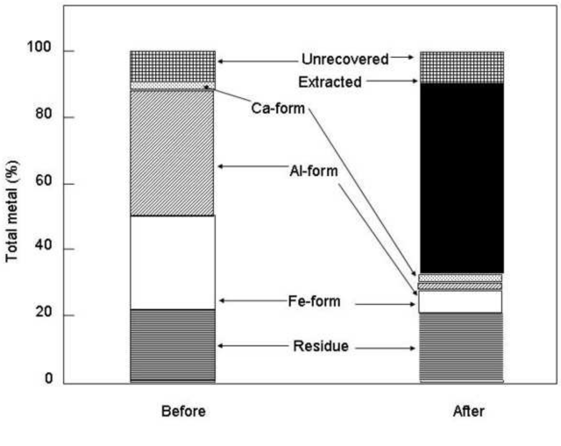

Figure 3. Sequential extraction of arsenic before and after phosphate treatment (adapted from Alam et al., 2001)

The effect of various forms of organic matter on arsenic mobilization has also been studied, with some differing views on this subject. Tessema and Kosmos (2001) examined the effect of humic, phthalic, oxalic and citric acids on the release of arsenic. The extraction of arsenic by humic acid decreased as pH decreased, whereas other acids showed an increase in extraction and the pH decreased. Fe and Al were also released indicating the complexation of these acids with these elements.

Using an IAEA-5 soil 5, Slejkovec et al. (2003) evaluated the extraction of the mobile fraction of adsorbed arsenic. Sonication for less than 1 h at 30°C or 1M NaH_2PO_4 , pH4 showed 31.9% extraction. The remaining amount was deemed to not easily extractable. The K_D was decreased from 465 to 163 L/kg as the phosphate ion concentration increased from 0.01 to 1 M. They also found that as the pH increases, the charge of ferrihydrite decreases, the combination of arsenic and iron increases and phosphate charges decreases. No displacement of arsenic by phosphate occurs as the pH increases. However, repulsion by ferrihydrite at pH of 8 to 10, increases and desorption thus increases. This is illustrated in Fig. 4.

3.2.2 *Immobilization methods*

Most *in situ* remediation techniques are potentially less expensive and disruptive than ex situ ones, particularly for large contaminated areas. Natural or synthetic additives can be utilized to enhance precipitation, ion exchange, sorption and redox reactions (Mench et al., 2000). The sustainability of reducing and maintaining the reduced solubility conditions is key to the long term success of the treatment. *Ex situ* techniques are expensive and can disrupt the ecosystem and the landscape. For shallow contamination, remediation costs, worker exposure and environmental disruption can be reduced by using *in situ* remediation techniques.

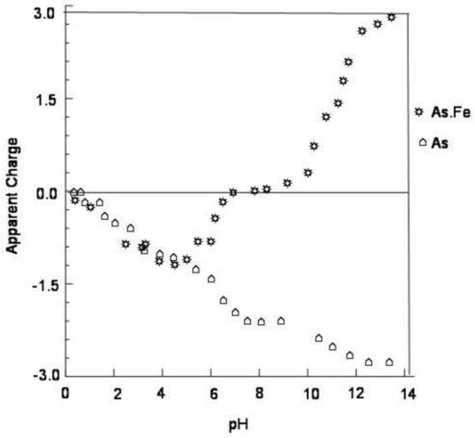

Figure 4. Apparent charge of arsenate and the combination of arsenate and ferrihydrate (Adapted from Slejkovec et al., 2003)

According to the United States National Research Council (NRC), the sustainability of natural attenuation is dependent on the sustainability of the mechanisms for immobilizing or destroying contaminants while the contaminants are being released into the groundwater. A mass balance analysis can be used to estimate the long-term destruction or immobilization rates (NRC, 2000). For hydrocarbons, the availability of electron acceptors or donors may be evaluated to determine the sustainability of remediation techniques such as natural attenuation for hydrocarbons. However, in the case of metals and metalloids such as arsenic, this approach is only applicable if the attenuation is biologically driven.

Information regarding the sustainability of arsenic immobilization could be obtained for sequential extraction techniques will be obtained to evaluate the adsorption, sequestration and bioavailability of arsenic in soils (Yong and Mulligan, 2004) before and after treatment with additives such as iron, aluminum, calcium and manganese, cement, lime and pozzolanic materials as was previously shown for phosphate treatment.

Various field trials have been performed to evaluate the addition of steel shots and beringite for reducing arsenic solubility (Verkleij et al., 1999). The corrosion of the steel shots enhances the formation of Fe/Mn oxides while beringite affects soil pH (Vangronseld et al., 1999). Boisson et al. (1999) amended the soil with various additives and determined that 1% steel shot with 5% beringite was the most effective for reducing arsenic mobility due to adsorption and precipitation with iron oxides. However, hydroxyapetite addition could enhance mobility due to competition with phosphate. Distilled water was used as the extractant for evaluating mobility. Warren et al. (2003) later found, however that bioavailability was not reduced by amending with iron grit. It was postulated that smaller particle sizes and larger specific surface areas may be more effective.

Ferrous sulfate addition was also evaluated by due to in situ synthesis of iron oxyhydroxides (Voigt et al., 1996). Warren et al. (2003) also evaluated addition of ferrous sulfate and lime in solution to produce ferrous oxides. Significant reductions in arsenic bioavailability were achieved.

As sequential extraction does not predict plant uptake or phytotoxicity, a follow-up study was performed by Mench et al. (2003) to evaluate the long term sustainability of various amendments at a former gold mine including 5% compost with steel shot (CSS), 5% compost with 5% beringite and 1% steelshot (CBSS), 5% compost (C) and others. The CBSS and CS treatments were very successful in terms of revegetation and decreased leaching compared to compost only.

Other types of iron oxy-hydroxides which are industrial byproducts have also been evaluated as immobilization additives (Lombi et al., 2004). They include water treatment sludges, red muds and red gypsum. Risk assessment of the remediation technologies needs to include assessment of the pathways and sensitivity of the plants and microorganisms to the treatment. The water treatment sludges decreased arsenic mobility and bioavailability, and enhanced plant and microbial growth. However, in the event of a pH reacidification, the treatment may not be stable as the arsenic becomes exchangeable.

Amendments such as goethite, iron grit, iron(II) and (III) sulfate (with lime) and lime were evaluated by Hartley et al. (2004). Several leaching tests were used to determine the short and long term immobilization ability of the amendments. In terms of efficiency for As immobilization, the following order was determined: Fe(III)>Fe(II)>iron grit>goethite> lime. Although iron oxides attenuated arsenic in the soils, they led to increase mobility of Pb and Cd.

Vitrification is applicable for arsenic-contaminated soils arsenic is of low volatility. Melting ability depends on the soil's silica content. The maximum allowable oxide content for arsenic is 5% (Smith et al., 1995). It is the best demonstrated available technology (BDATs) for RCRA wastes.

Addition of immobilizing agents that are insoluble such as hydroxyapetite, zeolites or illitic clays can be problematic, particularly to depths below 50 cm. A solution to this may be to utilize permeable reactive barriers within the groundwater. Zeolite, hydroxyapetite, elemental iron and limestone have been used, particularly for chromium reduction (Vidac and Pohland, 1996). Other materials such as surfactant-modified zeolite may also be promising for their ability to sorb arsenic from soil leachates (Sullivan et al., 2003). These materials adsorb arsenic due to ion exchange phenomena with the counterions of the surfactant head groups and arsenic may also complex with the organic carbon in the surfactant bilayer. The media, regeneration of the median and retention time through the barrier would need to be optimized. Another alternative is the use of passive flow-through contaminant barriers. These have been installed within the migrating plumes (Fuller et al., 2002).

3.4 Evaluation of the sustainability of remediation alternatives

As we have seen, there are a variety of remediation technologies available for arsenic. Pollution of groundwater from arsenic is a major threat to the health of humans. Therefore, measures are required to mitigate this problem. In choosing the technologies. we need to consider carefully what are the targets, exposure routes, future land use, acceptable risks, legislation, and emissions. Other factors are seen in Fig. 5. We have discussed and seen the applications of some of the tools that can be utilized to evaluate the sustainability of the technologies. Specific comments are included in Table 1 for the various technologies. Other factors that need to be considered to evaluate site remediation technologies include disturbance to the environment, energy use and consumption, solid wastes generated, emissions of contaminants and greenhouse gases into the air, water and materials used.

4 CONCLUSIONS

Therefore, based on the mechanisms for release from the geoenvironmental engineering perspective, and from the viewpoint of environmental management, *in situ* geochemical procedures need to be structured to counter arsenic release from the source materials. The *in situ* geoenvironmental approach is substantially more sustainable than the *ex situ* water treatment one as it does not produce wastes that need to be further treated.

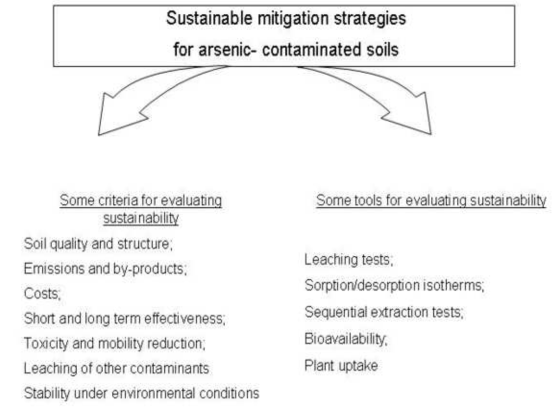

Figure 5. Some of the criteria and tools for evaluating technologies and protocols for environmental management of arsenic-contaminated soils

Table 1: Comparison of remediation technologies for arsenic in soil and groundwater (adapted from Evanko and Dzombak, 1997)

Technology	Cost*	Long-term effectiveness	Reduction of toxicity	Reduction in mobility
Capping	Good	Low	Low	Good
Solidification (in situ)	Avg.	Avg.	Low	Good
Solidification (ex situ)	Good	Avg.	Low	Good
Vitrification	High	Good	Low	Good
Biological treatment	Good	Low	Good	Good
Soil washing	Avg.	Good	Low	Low
Pyrometallurgical extraction	High	Good	Low	Low
Electrokinetics	Avg.	Good	Low	Low

* High is in the range of \$300 to 900/tonne, average from \$100 to 300/tonne and low is up to \$100/tonne. Avg. denotes average.

By reducing the concentrations to levels below the critical levels, the risk of arsenic poisoning of the people using the groundwater resource will be minimized or eliminated and thus ensuring the sustainability of groundwater for drinking purposes, a basic need for society.

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