



REMEDIATION OF ARSENIC FROM WATER

Author.

Dr. Jyotsna. Lal Professor. , Chemistry Department, Christ Church P G College Kanpur 208001 UP, India
email: jyotsna_lal@yahoo.com

ABSTRACT

Degradation of ground water due to Arsenic , toxic effect to human body and removal of arsenic from water have been studied in this paper.

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INTRODUCTION

The traditional reputation of arsenic as a poison is largely due to the higher toxicity of As(III) ion whereas elemental As , As(V), organic arsenic compounds are commonly considered less toxic. Arsenic is well known for its toxic and carcinogenic properties (class A carcinogen).

The normal amount of As in hair is 0.08-0.25 mg/kg whereas 1mg/kg is an indication of toxicity The normal amount of As in nails 0.043 –1.08 mg/kg The normal amount of As in urine range 0.005-0.40 mg/kg [1.5 D] Maximum permissible limit of As for drinking water is 0.05 mg/l . Arsenic contamination has been found wide spread in different regions of West Bengal is due to dissolution of arsenic containing bedrock. The high arsenic content of the drinking water has become a major public health issue in 6 districts of West Bengal in India, where an estimated 30 million people dependent on tube well water sources have been adversely affected. The district are in the eastern sector of West Bengal, extending 450 km. From north to south bordering Bangladesh. The severely affected districts are Malda, Murshidabad, Nadia, Bardhaman, 24 Parganas (North) and 24 Parganas (South). The arsenic content in the ground water ranges from 0.01 to 0.59 mg/l in as many as 830 villages in this area. Tubewells have been dug to a depth of 120 to 50 feet. Table 1a shows the arsenic levels in drinking water in West Bengal. Table 1b shows a study of arsenic level in the tissue and body fluid of the people of West Bengal

TABLE 1 a OCCURENCE OF ARSENIC IN THE GROUNDWATER OF WEST BENGAL

8 districts /830 villages in 58 blocks Total samples of analysed	Conc of As in water mg/l		
	<0.01	>0.05	> 0.01
34900	43.5	38.7	56.5

TABLE 1 b STUDY ARSENIC LEVEL IN THE HUMAN TISSUE [mg/kg] AND BODY FLUID [ug/1.5D] OF THE PEOPLE OF WEST BENGAL

Total samples of analysed	% of samples above normal level
HAIR	
3530	74.0
NAIL	
3620	74.0
SKIN	
163	[1.2-29.3]
URINE	
7240	83.6

In the last decade, the contamination of soil and groundwater by arsenic compounds has been getting more and more attention. The seriousness of the problem can be seen in Florida where over 2600 sites were found contaminated with arsenic compounds 1 . Similarly, thousands of additional sites exist throughout the U.S. Typical sources of arsenic contamination include mining and processing operations, leakage from

natural rocks, and usage of arsenic based herbicides and pesticides. Arsenic in the form of arsenates is one of the common elements uniformly distributed in major rocks types. Under standard conditions, arsenate migration is limited due to the strong sorption by clays, hydroxides and organic matter 2 . However, the change of redox potential, pH and oxygen can trigger desorption. Also, several strains of bacteria were found to be involved in oxidation as well as in methylation and alkylation of arsenic compounds. The overall toxicity of arsenic depends on the concentration and the ionic form of arsenic salts. Unfortunately, the active components of the arsenic based herbicides; pesticides and wood preservatives were soluble forms, such as sodium arsenate and arsenic trioxide. Also, organic arsenical pesticides pose a danger of releasing toxic inorganic derivatives of arsenic via biotransformation. In general, most of the arsenic-contaminated sites are the result of common application of arsenic based herbicides and pesticides. Until 1960, calcium arsenate was used for boll weevil and cotton leaf worms 10 ; lead arsenate and arsenic trioxide have been used for pest and termite control, and sodium arsenite solutions have been utilized for dipping cattle and sheep. Agriculture utilizes arsenic in the form of methyl arsenate (MSMA), disodium methyl arsenate (DSMA), cacodylic acid and arsenic acid. The MSMA, DSMA and cacodylic acid are used to control Johnson and nusedge grass and weeds in cotton fields. Arsenic acid is used as a desiccant for the defoliation of the cotton ball before harvesting 3 , and arsenic trioxide was widely used as herbicide from the early 1950's to early 1970's to eliminate weeds at industrial facilities and on railroads.

Most water treatment technologies suitable for the developed countries are expensive for towns and small industries of the developing countries because of limitation of funds and lack of skilled man power . The process of adsorption has a an edge over other methods,,due to its simplicity and

sludge free clean operation . There is a increasing search for low cost adsorbent material made from locally available waste material and plant substrata [Table 2] Some low cost adsorbents selective for As (III), As(IV) have been tested in the laboratory by batch and column technique , pH , concentration and flow rate , break- through capacity have been investigated .

The objective of this paper is to review the existing methods that are or could be applied to remove arsenic compounds from the soil and groundwater. Some of those technologies could be used for soil and groundwater treatment as well. However, because of a different matrix (soil or water), even the same application will carry different limitations, costs and overall effectiveness. For these reasons we are presenting separate reviews of the methods used for the soil treatment and for groundwater clean up

TABLE 2 ADSORBENTS FOR REMOVAL OF As [III] , As[V] FROM WATER

ADSORBENT	REFERENCE
Ferric Sulphide bed	Lee et al 1972
Bauxite sample	Balient Ambro 1974
Chitin mixture	Elson CM et al 1980
Amorphous iron oxide	Pierce et al 1982
Manganese Dioxide soil	D.W Huang 1983
Lake sediments	Takamastu 1985
Coal fly Ash	Sen AK and Arnab K.De 1987
Coral limestone	Shigeru 1992
Activated carbon impregnated with metallic Ag,Cu	Rajakovic 1992
Ganga sand	Vaishaya RC 1993
Synthetic Birnessite	Scott 1995
Basic Yttrium carbonate	Warsy SA 1996
Iron coated spent catalyst	Liu JC ;Huang 1997
Chemically treated sawdust	Nag A 1998
Manganese Dioxide coated sand	Sanjeev Bajpai 1999
Lanthanum impregnated Sawdust carbon	Raji .C; Anirudhan1999
Ferric oxide impregnated carbon	Brian Reed 2000

INDUSTRIES RELEASING ARSENIC

Arsenic is used in metallurgical industry, glassware and ceramic industries, dye and pesticide manufacturing industries, petroleum refineries. Chemicals containing Arsenic are used in manufacture of herbicides and pesticides. In West Bengal, a factory manufacturing acetocopper arsenite, a pesticide, polluted drinking water in the southern part of Calcutta. Arsenic is released into the surface water through mining and burning of coal as well as copper smelting. Rivers flowing through the coal fields of Bihar have been reported to carry large amounts of arsenic, responsible for downstream arsenic poisoning in West Bengal. According to studies, the arsenic contamination of the Damodar in West Bengal is mainly due to dumping wastes from the coal mines along the river bed.

HEALTH HAZARDS DUE TO HIGH ARSENIC INTAKE

As (III) ion causes arsenicosis in human beings. Symptoms of arsenic poisoning are abdominal pains, vomiting, diarrhoea and pain in the extremities followed later by numbness and tingling of the extremities, palmoplantar hyperkeratosis, Mees lines on the finger nails, deterioration in motor and sensory responses, chronic arsenicalism includes dermal lesions, peripheral neuropathy, skin cancer, peripheral vascular disease. Bony fishes and fresh water mussels contain a high content of As [Table 3]. Non-cancerous effects of arsenic exposure might show up as skin lesions such as hyperkeratosis, hyperpigmentation and depigmentation. Prolonged exposure could cause central nervous dysfunction, cardiovascular diseases, digestive and respiratory system diseases, liver and spleen swelling and liver cirrhosis.

TABLE 3 ARSENIC CONTENT OF SOME FOOD ITEMS

Food items	Arsenic content [ppm]
Bony fish	2-8
Oyster	3-10
Mussels	120
Shrimps	42
Prawns	174
Cows milk	0.03-0.06
Vegetables	0.001

As, Hg, Pb, Cd are highly toxic even at low concentrations. Zn (II) in some metallo-enzymes is substituted by Cd (II). Arsenic acts on the biological system in the following ways: Trivalent toxic As inhibits enzyme activity by reacting with ligands containing sulphhydryl groups. As (III) attacks -SH groups of an enzyme, i.e. inactivation of pyruvic dehydrogenase by complexation with As (III), preventing the generation of ATP in the citric acid cycle. Pyruvic dehydrogenase is highly sensitive to As because of its interaction with two -SH groups of lipoic acid, leading to pyruvate accumulation. As (III) ion readily crosses the placental barrier and causes foetal damage. Arsenic when absorbed in the biological system can undergo bio-transformation under in vivo conditions. As (V) ion or arsenate can

be reduced into As (III) ion or arsenite The half life of this bio-transformed arsenite is three to five days , when it can cause harm to the system .

Arsenicosis is a chronic disease with a significant latency period for non-cancer and cancer effects. There appears to be discrepancy in the literature regarding latency, with some reports of 2 years being the minimum for hyperpigmentation and keratosis. Researchers in Bangladesh suggest that 5 years is the minimum latency, whilst some other estimates suggest that this is 9 years. Latency for cancers is also unknown, but it is estimated to be of the order of 20 years. The proportion of a population exposed to elevated arsenic that will develop arsenicosis is uncertain, but may be significant. Medical treatments for arsenicosis are not fully developed. There is indication that switching to arsenic-safe water and anti-oxidants may reverse symptoms in early stages. The proportion of a population exposed to elevated arsenic from drinking-water that will go on to develop arsenicosis is unknown. WHO have modelled the progression of arsenicosis using data from Samta, Bangladesh. The range of those affected over 30 years was 15.75% in the lowest estimate scenario to 29.25% in the highest estimate scenario. Variation in the estimates of mortality from cancers was between 5.0 and 6.5%. This implies a significant overall health burden for those affected. It is not clear whether early removal of arsenic- contaminated water would reduce the onset of cancers, but it is assumed that it would have some impact because of the cumulative nature of the risk. More recent work suggests that anti-oxidants within vitamins A, C and E and possibly compounds containing zinc and selenium also work to reverse symptoms. A recent controlled trial was performed in Bangladesh, but this remains to be published and may require further controlled clinical trials. However, this does indicate the necessity of combining both environmental and medical interventions for arsenicosis.

PREPARATION OF ADSORBENTS

Washed and dried river Ganga sand [geometric mean size 0.49mm] sand was washed with distilled water until the runoff was clear , dried stored

Manganese dioxide coated sand was prepared by forming manganese dioxide by oxidation of manganous ion by permanganate in presence of sand .Washed and dried river sand [geometric mean size 0.49mm]250g was added to 250 ml 0.1 M KMnO₄ solution , to this 250 ml of 0.4 M NaOH to make the pH alkaline thereafter 250ml 0.3m MnCl₂ was added dropwise while the mixture was mixed in magnetic stirrer .dried in oven for 24 hr The coated sand was washed with distilled water until the runoff was clear , dried stored .

The method adopted for iron III hydroxide impregnation on coal. Bituminous coal crushed to geometric mean size of 0.102 mm was washed several times with distilled water and dried in the oven The proximate analysis of coal was moisture at [60% RH] 4.7%,ash 18.2% , volatile 32.0% and fixed carbon 45.1% coal 20 g was slurried in 100ml distilled water to which 2 g of iron III chloride was added ,using 2N sodium hydroxide solution the pH was then adjusted to 11.5 and the volume made upto 200ml .The slurry was allowed to settle for 24 hrs . The adsorbent following pretreatment /impregnation was removed, washed several times with distilled water dried and dessicated.

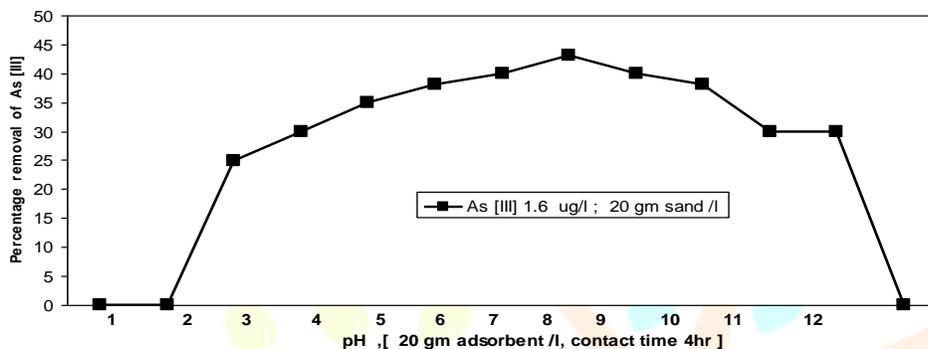
METHODS AND MATERIALS

Stock solutions were prepared using AR grade Sodium Arsenate and Sodium Arsenite in distilled and tap water . 0.1 MKOH and 0.2 M HCl used to adjust the pH . Batch adsorption studies were performed in glass stoppered conical flasks at room temperature in rotary shaker. Column studies were done using 1.25 ID glass column loaded with adsorbent ,and As (III) and (V) was percolated downward at flow rate ~5ml/minute .The operation was stopped when As was detected in effluent [break-through point] Arsenic was determined in the test solutions spectrophotometrically using Perkin Elmer Spectrophotometer.

RESULTS AND DISCUSSION

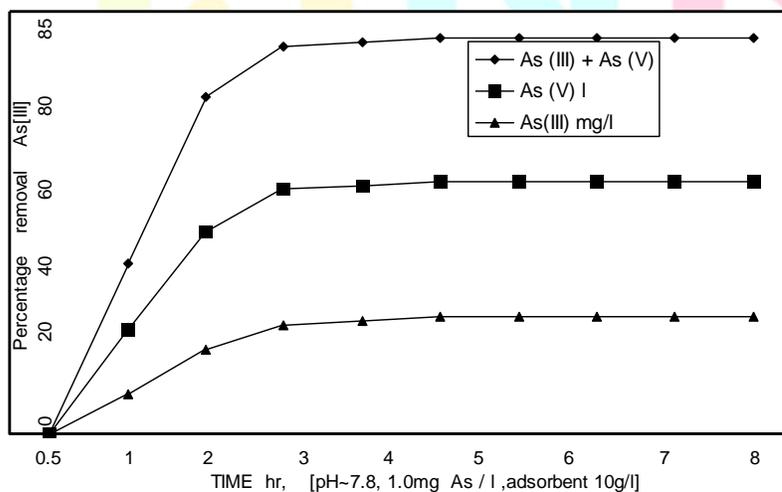
Ganga sand can be used as low cost adsorbent for treating As (III) at low levels . The removal of As (III) was determined to be 85 µg /g for initial concentration of 1.6 mg/l at an adsorbent dose of 20g/l, adsorption of As (III) on ganga sand is pH dependent and maximum percent removal of occurs between pH 7-9, followed by a marginal decrease at pH range 9-12 [Figure 1] The is that As (III) is found in aqueous in form of arsenious acid .Being undissociated weak acid with no charge the uptake of As (III) may be due to combination of electrostatic attraction between sorbate and sorbents and Vander Waals attraction .Column studies were done at 0.8mg As (III)/l concentration in tap water which produced arsenic levels below WHO guideline value for As .Sodium hydroxide is a efficient regenerant chemical for removing As from ganga sand.

FIGURE 1 As [III] REMOVAL BY GANGA SAND



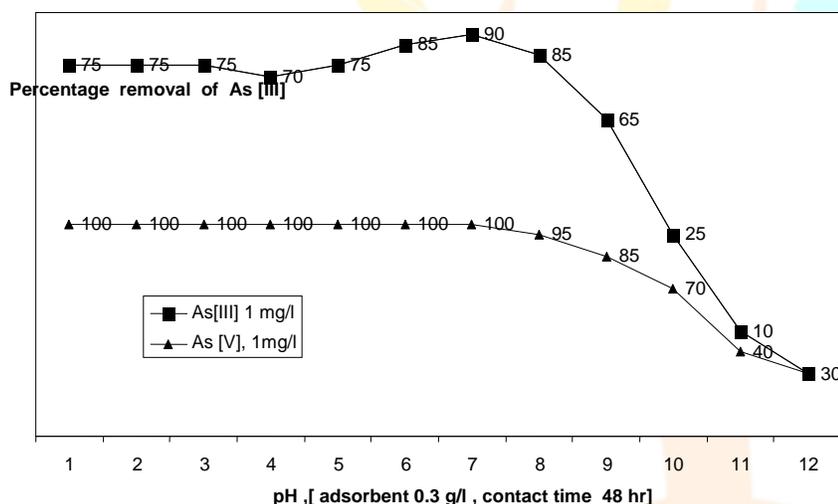
Manganese dioxide coated sand [Figure 2] Arsenite is oxidized to arsenate by manganese dioxide [$H_3AsO_3 + MnO_2 = HAsO_4^{2-} + Mn^{2+} + H_2O$]; the released Mn^{2+} adsorb on the manganese dioxide, giving it a positive surface and leading to an enhancement in the removal/adsorption of arsenate present initially or produced as a result of arsenite oxidation. The performance of manganese dioxide coated sand in As removal in 10 cycles in column experiment was good [0.5mgAs(III)/l, 0.5mg As (V) /l influent] no As (III) or Mn (II) leaching in the effluent. Breakthrough volume was at WHO guideline 0.01mg/l, 85% As was recovered during regeneration.

FIGURE 2 REMOVAL OF As(III), As(V) BY MnO2 COATED SAND



On Iron III hydroxide impregnated coal.[Figure 3] , For anionic As (V) removal decreased with increasing pH while As(III) removal is not a strong function of pH ,increased to maximum at pH~7. Arsenic (V) and (III) form inner sphere complexes with iron hydroxide . As (V) speciation between pH 4-10 $H_2AsO_4^- = HAsO_4^{2-} + H^+$ $pK_a = 7$,at this pH range the net surface is negative , producing a repulsive electrostatic force between $HAsO_4^{2-}$ and adsorbent .The chemical interaction force must be greater Than the repulsive electrostatic force for removal to occur . As (III) in pH range 3-11, $H_3AsO_3 = H_2AsO_3^- + H^+$ $pK_a = 9.2$, the predominant species is H_3AsO_3 ,the electrostatic force is zero the removal is attributed to chemical interactions [inner sphere complexation] at pH ~ 8.5 about 20 % of the the aqueous As (III) exists as $H_2AsO_4^-$, a repulsive electrostatic force exists ,this repulsive electrostatic force contributes to the decrease in removal with increasing pH

FIGURE 3 REMOVAL OF As [III], As[V] BY FE- OXIDE IMPREGNATED COAL



Iron (III) hydroxide loaded coral was used as an adsorbent for Arsenic (V) and (III) Iron (III) coexisted in the aqueous solution , no effect of presence of Cl^- , NO_2^- , SO_4^{2-} , CH_3COO^- but PO_4^{3-} greatly decreased the adsorption.

As (III) adsorption on Lanthanum impregnated saw dust carbon .[Figure 4 increased 10.3 %-- 86.0 % by increasing the pH from 1.0-12 for the initial concentration of 50 mg/l .The anionic forms $H_2AsO_3^-$ and $HAsO_3^{2-}$ are the predominant species in this pH range. It seems that the chemical reaction and the less soluble precipitates $LaHAsO_3$ or $La(OH)H_2AsO_3$ on the carbon surface was most likely adsorption process , the predominant species is H_3AsO_3 in this acid range .

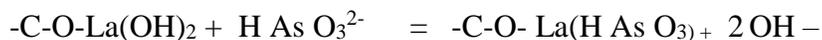
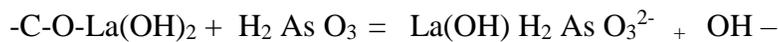
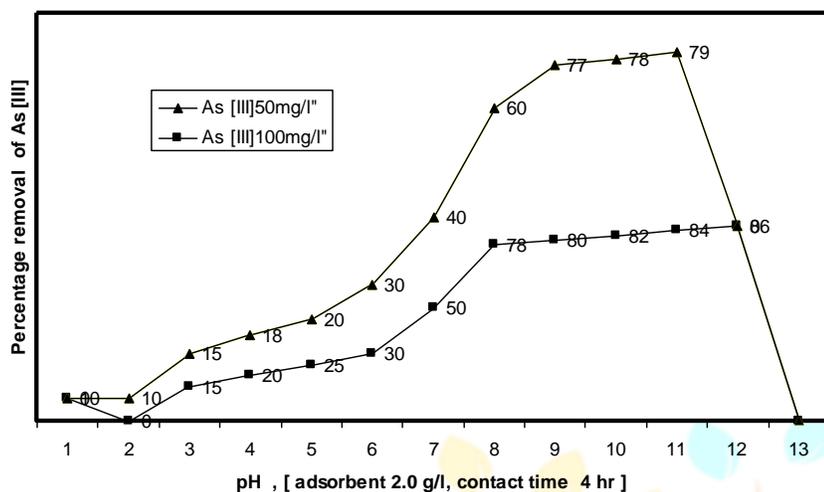


FIGURE 4 As [III] REMOVAL BY LANTHANUM IMPREGNATED SAWDUST CARBON



Impregnation of activated carbon by metallic Cu improves the sorption process for As (III) in pH range 4 - 9 which is the optimum pH for apparent affinity between the carbon surface and the arsenic species H_3AsO_3 and $H_2AsO_4^-$ predominant at this pH

A strong anion exchange resin DOWEX -1X8 can be used to separate a mixture of Arsenic (V) and (III) anions at pH 7.7 [Table 4a]. As (III) is as unionized H_3AsO_3 while As (V) is present as $H_2AsO_4^-$, $HAsO_4^-$ anions which are retained on the resin bed which is eluted from the column with 0.1M HCl. The toxic As (III) can be oxidised to As(V) by Chlorine water and removed [Table 4 b].

TABLE 4a RECOVERY OF ARSENIC FROM A MIXTURE OF As(III), As(V) IN DIFFERENT RATIO AFTER SEPARATION THROUGH ANION EXCHANGE COLUMN

As (III) Taken μg	As(V)	As (III) Recovered μg	As(V)
2.0	0	1.8	0.01
0.0	1.8	0.01	1.9
1.0	0.45	0.95	0.5
1.4	0.35	1.4	0.35
0.4	0.25	0.3	0.35
0.2	0.35	0.1	0.35
0.2	0.2	0.15	0.2
0.1	0.09	0.1	0.1
0.04	0.04	0.04	0.05
0.02	0.02	0.015	0.015

TABLE 4b RECOVERY OF As (V) FROM As (III) AFTER OXIDATION WITH OXIDATION WITH Cl₂ WATER AND PASSING ION EXCHANGE COLUMN

Amount of As (III) μg	Amount of Cl ₂ water (ml) added	Amount of As(V) μg recovery	% Recovery
100	nil	0	--
100	2.0	92	92
100	4.0	90	90
100	8.0	89	89

A study indicated that Al in concentration of $80 \mu\text{g} \setminus \text{L}$ or more results in Dialysis dementia and risk of Alzheimer's disease [Gupta AB et al 1999]. Activated alumina could achieve over 95% Arsenic (V) and (III) removal twice as effectively as activated bauxite and 12 times more effectively than activated carbon. Activated alumina removed As (V) over 20 times more effectively than As (III) along with Fluoride at pH 5.0. The adsorption of As (V) on alumina is governed by the diffusion within the particles. Better adsorption capacity can be achieved by standing the adsorption process for over a period of more than 12 hr. The smaller the particle size the better will be the kinetic performance. Activated alumina can be regenerated by 4-5% NaOH, 70% capacity can be regenerated after first use. Ganga sand and Manganese dioxide coated sand are better low cost adsorbents than others. Both show promise for cheap and effective removal of Arsenic (V) and (III) from water.

SOIL TREATMENT METHODS

There are number of known technologies that could be used for the treatment of soil contaminated with arsenic. These techniques allow for either physical removal of the arsenic compounds from the soil matrix, or permanent or temporary "lock" of arsenic compound on site. The second group of technologies focuses on stopping migration of arsenic through soil and leaching to groundwater. In the last decade, the contamination of soil and groundwater by arsenic compounds has been getting more and more attention.

1. Excavation.

a. Description: Soil, removed from the contaminated site, is hauled to landfill or treated ex-situ.
b. Advantages: Permanent removal from the site; no future leaching.

c. Disadvantages: Transfer of contaminated soil to other location.

d. Cost (with disposal): \$46 – 70/ton 1 .

2. Capping.

a. Description: This technique utilizes an impermeable surface barrier for stopping or reducing possible infiltration of the precipitation or stormwater runoff collected at the contaminated land surface. Caps are designed to either redirect rainfall/stormwater or eliminate infiltration.

b. Advantages: No percolation through contaminated site.

c. Disadvantages: High groundwater table, or high fluctuation in groundwater will make this method useless.

d. Cost: \$54.00/ton 5 .

3. Soil Solidification/Stabilization.

a. Description: Uses cement or other binder for solidification of contaminated soil by chemical fixation or encapsulation.

b. Advantages: (1) Encapsulated contaminated soil stays on-site; (2) Soil permeability is reduced below $1 \times 10^{-7} \text{ cm/s}$ and (3) Possibility of leachate is substantially reduced.

c. Disadvantages: (1) Not feasible in residential setting, and (2) Required monitoring for the changing conditions that could lead to the leachate in the future.
d. Cost: \$72-348/ton 5 .

4. Vitrification.

a. Description: The contaminated soil is melted into glass. The technology is based on generating high temperatures on site ($1600 - 2000 \text{ }^\circ\text{C}$) by high amperage current delivered through electrodes inserted into contaminated area.

b. Advantages: (1) Contamination is trapped in the glass; (2) Possible leachate is eliminated.

c. Disadvantages: (1) Air could be contaminated by vapor from the heated site, therefore air above melted area should be collected and treated; (2) Maximum depth of treatment is 6 m; (3) Possible safety hazard because of high voltage and potential splashing of melted medium; (4) High costs.

d. Cost: \$480-1044/ton 5 .

5. Containment (barrier walls).

a. Description: Construction of the walls to inhibit migration of contaminants from the site. Several used options include sheet pile walls, grout curtains and bentonite slurry walls.

b. Advantages: (1) Simple solution; (2) Decreases permeability from surrounding sediments.

c. Disadvantages: (1) Required monitoring for the changing conditions that could lead to leachate in the future; (2) Possible contamination from the wall's material; (3) Not effective without lower clay or confining unit.

6. Soil Flushing.

a. Description: Contaminated soil is purged with a mixture of reagents with addition of co-solvents or surfactants. Soil flushing could be done in the continuous, slug or pulsed mode. By using different extraction solutions, arsenic could be extracted in ionic form, but also as a (1) aluminum bound-, (2) iron bound- or (3) calcium bound- complex. It was found that flushing soil with phosphate solution under controlled pH (4–6) can remove up to 80% of adsorbed arsenic compounds 7,8 .

b. Advantages: Permanent removal from contaminated site.

c. Disadvantages: (1) Possible contamination of site by extraction solution, and (2) additional treatment or disposal of the sludge is required.

d. Cost: Depending on technique: Reported: (1) \$8/ton, including ancillary costs such as mixing chemicals and groundwater pump and treat (1), but also \$72-348/ton 5 .

7. Electrokinetics.

a. Description: Method uses low intensity direct current applied to anode and cathode inserted into contaminated soil. Direct current initiates electroosmosis and ion migration to the appropriate electrode.

b. Advantages: Method could be used in fine-grained geologic materials.

c. Disadvantages: (1) Soil need to be saturated with water; (2) No commercial uses yet.

d. Cost: Estimated <\$120/ton 1 .

8. Phytoremediation.

a. Description: Removal through plant's uptake. Studies shows that water hyacinth (*Eichhornia crassipes*)⁷ , saw palmetto (*S. repens*) and plant moss verbena (*V. tenuisecta*)⁹ can accumulate arsenic. It was reported that phytoextraction by edenfern TM could attain bioaccumulation factor of 193 through uptake and bioaccumulation from the 38.9 mg As/kg of soil to 7526 mg As/ kg fern fronds 10 .

b. Advantages: Depending on the plant used for phytoextraction, it is possible to achieve considerably high bioaccumulation (ratio of plant over soil concentration).

c. Disadvantages: (1) Limited to the growing season; (2) Requires proper biomass disposal/treatment; (3) Litter from plants could be a problem; (4) Uptake possible only up to the depth of root penetration.

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