POLLUTION OF GROUNDWATER BY ARSENIC:
SOME UNANSWERED QUESTIONS


The geochemistry of arsenic revolves around its association with two phases; iron sulfide and hydrous iron oxide. Groundwater processes that destroy either should add arsenic to groundwater, but this doesn’t always happen - and it is the exceptions that we need to understand. Destruction of hydrous iron oxides by reductive dissolution certainly releases arsenic to groundwater in lethal amounts in Hungary, Taiwan, Vietnam, and the Ganges Plain, but the expected chemical signals for this process e.g. a strong relationships between Fe(II) and As in groundwater, are noticeable for their absence. In other aquifers, particularly fractured igneous aquifers, oxidation of arsenical pyrite can give water in which sulfate bears an unequivocal isotope signature of its pyritic origin, but from which arsenic is largely absent, despite it being present in the precursor pyrite. Yet another interest, especially in densely populated areas that are affected by arsenic such as the Ganges Plain, surrounds the role played by human activity in locally, even regionally, influencing arsenic concentrations in groundwater.

In presenting a synthesis of the mechanisms by which arsenic pollutes groundwater, these and other questions will be addressed to show how much we need to discover about such matters.

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ARSENIC OCCURRENCES IN GROUNDWATER FROM THE INDIAN SUBCONTINENT

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Arsenic pollution in the Bengal Basin in West Bengal, India and Bangladesh, in terai region Southern Nepal, and in western parts of Chhattisgarh State, central India, are all essentially natural phenomena. The main sources of arsenic in latter area are from weathered acid magmatic rocks from a Proterozoic rift. Abundant occurrences of magnetite/hematite, biotite-chlorite, other ferromagnesian minerals, and minor occurrences of polymetallic sulfides, especially in the Himalayas are adequate sources for arsenic in Bengal Basin and Southern Nepal. Arsenic sorbed in Fe-oxyhydroxide was preferentially captured in argillaceous, organic rich, early-mid Holocene deltaic sediments in Bengal Basin. Arsenic concentrations are neither high nor much different between adjacentely located polluted or unpolluted aquifers. That reduction of FeOOH is common in affected aquifers in Chhattisgarh area and intense in the affected aquifers of Bengal basin is shown by moderate to high concentration of dissolved iron (Fe = 0.2 – 4.86 and 9-36 mg/L respectively), as against traces or very low concentration of sulfate. Dissolved iron concentration is significantly low (<1 mg/L) in groundwater from the Ganga flood plain upstream of the Rajmahal hills, which are uncontaminated by arsenic. Arsenic appears to be released to groundwater by bio-mediated reductive dissolution of Fe-oxyhydroxide. In Bengal Basin, continued and increased recharging of shallow aquifer because of extensive pumping triggered the reduction process by inducing and enhancing movement of groundwater enriched in highly reducing degraded organic products. Phosphate from degraded organic matter and partly leaked from chemical fertilizers may also aid the process.

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ARSENIC PROBLEM IN GROUNDWATER IN NEPAL

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Recently, Arsenic (As) has been detected in groundwater in southern region of Nepal. So far about 14,000 out of the estimated 200,000 tubewells have been tested and about 5% of the test result is found to have exceeded Nepal's Interim Standard of 50 µgL⁻¹.

This paper presents an analysis of spatial distribution of As concentration in groundwater and identification of vulnerable communities and high risk areas in Nawalparashi, one of the hot spot district in southern Nepal. The analysis is based on the data and information available so far from various studies conducted by DWSS and other agencies. Geographic Information Systems (GIS) and Geostatistical tools are used for the analysis to explore the relationships between As concentrations and socioeconomic scenario.

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ARSENIC CONTAMINATION IN GROUNDWATER IN NEPAL – AN OVERVIEW

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The government of Nepal initiated proactive approach in studying on possible contamination of Arsenic (As) in groundwater in Nepal in 1999. From the laboratory test result of groundwater in 14,536 tubewells conducted by the end of February 2002 in 24 districts, As concentration in about 5% of the tubewells is found to have exceeded 50 ppb which is the interim As standard in drinking water in Nepal. In about 22% of the tubewells, As concentration is found to have in the range of greater than WHO guideline value i.e. 10 ppb to Nepal's interim standard i.e. 50 ppb. The highest As concentration found so far in groundwater of Nepal is of the range of 456 ppb. From the study conducted so far, four mostly As affected districts are Nawalparasi, Rautahat, Bara and Parsa respectively.

A study by the government of Nepal is underway on health issues to find-out the incidences of Arsenicosis in the arsenic affected districts.

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ARSENIC SPECIATION IN MARINE SAMPLES BY ION-EXCHANGE HPLC-ICPMS

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A method for the detection of arsenic species in samples of marine origin is presented. Ion-exchange chromatography was used for the separation of the species and ICPMS was used as an element specific detector.

Emphasis is put on the findings of minor constituents of the arsenic compounds and their role in arsenic metabolism in the marine environment is shortly discussed. Especially the findings of dimethylarsinoyl acetic acid (AA) in various marine sample types are interesting. This compound has so far only been reported found in some shellfish samples. The compound is proposed to be an intermediate precursor in the biosynthetic pathway of dimethylated arsenoribosides to arsenobetaine, the end product of the marine arsenic metabolism. The finding of the compound in several different sample types is a support to this theory.

Also the presence of the “new arsenobetaine” trimethylarsonium propionate (also named arsenobetaine 2, AB2) in several different sample types is of interest. The identity of this compound was first reported by Francesconi et al. in 2000 and its presence supports another scheme for the generation of arsenobetaine(s) proposed.

Identification of these arsenic compounds in different sample types will improve our understanding of arsenic containing natural products and help elucidating the possible biogenic pathways of arsenic.

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DEVELOPMENT OF A FIELD DEPLOYABLE METHOD FOR ARSENIC SPECIATION IN DRINKING WATER

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Simple determination of total arsenic concentration in environmental samples does not reflect the level of hazard of this element. It is becoming increasingly important that the concentrations of the various forms of arsenic be determined to provide a much clearer view of the risk associated with arsenic exposure in the environment. Therefore, the aim of this study is to provide an optimized and a relatively simple, rapid, sensitive, accurate and relatively inexpensive technique for the speciation of the As (III), As (V), monomethylarsonic acid (MMA) and dimethylarsenic acid (DMA) using ion-exchange chromatography cartridges.

The approach of the method is based on selective retention of arsenic species on specific ion-exchange chromatography cartridges followed by selective elution and quantification using graphite furnace atomic absorption spectroscopy (GFAAS). The use of a set of three cartridges allowed to on-site separation of arsenic species, thus avoiding any storage and preservation problems. The different arsenic species were collected from the sample one by one. The appropriate variations on the developed procedure were fully investigated. The detection limit for the four species tested were similar and at about 0.37 μg/L.

Quality assessment of analytical determination is most properly based on the use of certified reference materials. As a consequence of lack of proper certified reference materials the accuracy of the results was checked using spiked samples. A set of ten repetitions of analysis with spiked samples of concentrations between 1 to 60 ppb was performed. Arsenic recoveries ranged from 79-112 %. To study ions interference spiked well and tap water were used. The recoveries for arsenic were 84-105 % indicating that there are no ion interferences in the developed method.

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ARSENIC ANOMALIES IN SOILS AND SEDIMENTS OF GOLD MINING AREAS IN MINAS GERAIS, BRAZIL – ENVIRONMENTAL IMPLICATIONS

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Soils and sediments around gold ore deposits and mining sites in the Iron Quadrangle present positive As anomalies (median concentrations > 100 mg kg$^{-1}$) and wide ranges (< 20 to > 2000 mg kg$^{-1}$) even in densely populated areas. These anomalies can be related to geological structures, to hydrothermal ore deposits and to their continuous exploitation over the past three centuries. The paper presents new data on both As geochemistry and soil and sediment geochemistry in general and discusses consequences of the encountered anomalies, their reasons and potential effects and how they compare with current screening and threshold values. While soil As-values are reproducible at a given site, sediment anomalies show a strong seasonal variation that can be explained by tropical hydrological variances. Suggestions are presented on how to minimize the As-load in densely populated areas that otherwise pose a potential health risk.

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STUDY OF AMBIENT AIR POLLUTION WITH ARSENIC IN THE REGION OF A COPPER SMELTER PLANT

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In the region of the town of Elisejna, situated in North-Western Bulgaria, there is a copper smelter plant. The ore, used for copper production, has high contents of arsenic. Such productions emit in the atmosphere particulate matter, sulfur oxides, nitrogen oxides, airborne arsenic, lead, cadmium and other metal aerosols and toxic substances.

In this region the ambient air pollution with particulate matter, sulfur dioxide, nitrogen dioxide, airborne arsenic, cadmium and lead is monitored at three sampling sites. The level of arsenic in ambient air varies within broad ranges depending on the type of input materials. The paper presents the results of the study on arsenic concentration in ambient air in table and graph form.

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SPECIATION OF ARSENIC IN BIOLOGICAL SAMPLES FROM ARSENIC AFFECTED AREA BY HPLC-ICP MS

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Speciation of arsenicals in biological samples is an essential tool to get an insight into its distribution in tissues and its species-specific toxicity to target organs. Although 60-75% of ingested arsenic is excreted in a few days through urine a certain part of it is excreted through hair, nail, skin scale, feces, and sweat. Biological samples (urine, hair, nail) examined in this study were collected from 47 people of West Bengal, India who were drinking arsenic contaminated water with 248 ± 59 µg As/L, whereas 59 blood samples were collected from a population who stopped drinking their arsenic contaminated water 2-7 yrs before the blood collection. Speciation of arsenicals in urine, water-methanol extract of freeze-dried RBCs, TCA treated plasma, and water extract of hair and nail was carried out by HPLC-ICP MS.

Urine contains 12.5, 6.5, 3.2, 14.4, 2.8, and 70.2% iAs\text{III}, iAs\text{V}, MMA\text{III}, MMA\text{V}, DMA\text{III}, and DMA\text{V}, respectively. Our results suggest that after ingestion of iAs human body can metabolize it into six different arsenicals. RBCs contains 21.6, 78.4 % AsB, and DMA\text{V}; blood plasma contains 12.4, 25.9, 30.3, 31.4 % AsB, iAs\text{III}, MMA\text{V}, and DMA\text{V}; hair contains 60.9, 33.2, 2.2, 3.7 % iAs\text{III}, iAs\text{V}, MMA\text{V}, and DMA\text{V}; nail contains 58.6, 21.5, 7.7, 9.2, 3 % iAs\text{III}, iAs\text{V}, MMA\text{V}, DMA\text{III}, and DMA\text{V}, respectively. The proportion of iAs\text{III} is more than that of iAs\text{V} in hair and nail. No MMA\text{III}, DMA\text{III}, and iAs\text{V} are found in any plasma and RBCs samples, but urine contains all of them.

So, further studies are necessary to conclude their fate in human body using blood specimens collected from the population who are currently using arsenic contaminated drinking water and other roots or to develop a speciation technique of insoluble protein-bound arsenic in biological samples.

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DEVELOPMENT OF A COMBINED ION EXCHANGE- ADSORPTION PROCESS FOR ARSENIC REMOVAL FROM WATER - A CASE STUDY

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New low-waste technology was developed by combination of ion exchange and adsorption methods. It is appropriate for selective removal of ammonium, iron, manganese and arsenic ions, as well as humic acids from drinking water. Natural zeolite was mined in Hungary and clinoptilolite content was found 65-70 m/m % by XRD analysis. Na-form of clinoptilolite was produced by 20 BV 20 g NaCl/L solution, then washed with distilled water. The manganese-form was prepared from Na-form with 20 BV of 1 mol/L MnSO₄, and 20 BV of 10 g/L KMnO₄. Granulated activated carbon (Filtrasorb 300) was produced by Chemviron Carbon. Al₂O₃/Fe(OH)₃ adsorbent was prepared from granules of 0,3-1,0 mm of activated Al₂O₃ and Fe(OH)₃ was freshly precipitated onto the surface of particles. The resulting Fe(OH)₃ impregnated porous adsorbent was dried at room temperature.

Laboratory and field experiments were carried out by 3,2 cm i.d.*15 cm and 8 cm i.d.*90 cm columns. Bed volumes (BV) of ion exchangers and adsorbents were chosen as 80 mL in laboratory, and 4 L at field experiments. The average exhaustion flow rate was about 5 BV/h.

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POSSIBLE INVOLVEMENT OF ORGANIC LIGANDS IN ARSENIC DETOXIFICATION AND HYPERACCUMULATION IN THE BRAKE FERN PLAN

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Brake fern, a recently discovered arsenic hyperaccumulator, is a potential plant for the phytoremediation of arsenic-contaminated soil. A better understanding of the arsenic uptake, hyperaccumulation, species transformation and tolerant mechanisms enhance the use of the Brake fern in phytoremediation and provide useful information for arsenic tolerant mechanism in other plants. In order to investigate the organic ligands possibly involved in arsenic detoxification in Brake fern, thiol-containing compounds, amino acids, and organic acids (carboxylic acids) were analyzed in the plant samples. Results show that arsenic can stimulate the synthesis of total thiols and acid-soluble thiols in roots, rachis and leaflets. The thiol levels in leaflets increase with arsenic concentration in soil. However, this trend is not obvious in roots and rachises. Acid-soluble thiols are the major components of total thiols. Sulfur in soil can increase the level of total thiols and acid-soluble thiols in roots when 1000 ppm arsenic is present in soil, but cannot increase the thiol levels in rachises and leaflets. Without arsenic, sulfur itself slightly increases the thiol levels in leaflets. The levels of phosphor are almost the same in roots, rachises and leaflets. The concentrations of total phosphorous in roots and leaflets increase significantly in 1000 ppm arsenic group compared with control group. Our preliminary results show that concentration of hydroxyproline, the major amino acid of the cell wall glycoproteins, is much higher in the plants from arsenic-contaminated soil than those from control. Identification of the major thiols has been conducted. The possible roles played by these thiols and amino acid for arsenic hyperaccumulation by Brake fern will be discussed in this paper.

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DEVELOPMENT OF A ROUTINE ANALYTICAL METHOD FOR ARSENIC SPECIATION IN HUMAN URINE SAMPLES

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Urinary arsenic levels are useful biomarkers for assessing the level of arsenic exposure and for investigating arsenic metabolism in humans. The determination of arsenic species, including arsenite, arsenate, dimethylarsinic acid and monomethylarsonic acid species, is often performed by approaches that are based on liquid chromatographic separation combined with arsenic selective detection by atomic absorption, atomic fluorescence or atomic mass spectrometries.

In this work, an arsenic speciation method based on ion pair chromatography combined with hydride generation atomic fluorescence spectrometry has been developed which is suitable for routine analyses of human urine samples. The separation technique utilizes ion pairing of anionic arsenic species with tetrabutylammonium hydroxide and reversed phase separation on a C\textsubscript{18} column. Consideration has been given to optimizing instrumental parameters for analyte sensitivity and selectivity as well as minimizing measurement times. Separation of the four major arsenic species is performed in approximately 8 min. with limits of detection of approximately 1-2 ng/ml for each specie.

Recent efforts have demonstrated the utility of the method for arsenic speciation in urine. Approximately 150 urine samples collected during three different studies of human populations have been analyzed for arsenic speciation and have revealed differences in urinary arsenic profiles that appear to be associated with the major sources of environmental arsenic exposure, e.g. drinking water or food sources.

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DEVELOPMENT OF CHROMATOGRAPHIC SEPARATION TECHNIQUES FOR DETERMINATION OF ARSENIC SPECIATION IN URINE SAMPLES

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The toxicological and biological properties of arsenic depend on its chemical form, and speciation of arsenic metabolites is essential to the understanding of arsenic metabolism and health effects. Chromatographic separation coupled with spectrometric detection is a favorable technique to quantitatively determine arsenic species present in environmental and biological samples.

Several chromatographic separation techniques coupled with hydride generation atomic fluorescence detection were developed for arsenic speciation analysis. Ion pair chromatography was used to determine dimethylarsinous acid (DMA$_{\text{III}}$) and monomethylarsonous acid (MMA$_{\text{III}}$), as well as arsenite (As$_{\text{III}}$), arsenate (As$_{\text{V}}$), dimethylarsinic acid (DMA$_{\text{V}}$), and monomethylarsonic acid (MMA$_{\text{V}}$). Detection limits with hydride generation atomic fluorescence were 0.5-2 μg/L.

Trimethylarsine oxide (TMAO) was separated from other arsenic species using anion exchange chromatography.

The methods were successfully used to measure arsenic species in human and rat urine samples. As$_{\text{III}}$, As$_{\text{V}}$, MMA$_{\text{V}}$, DMA$_{\text{V}}$, MMA$_{\text{III}}$ and DMA$_{\text{III}}$ were detected in human urine samples. TMAO and DMA$_{\text{III}}$ were found in urine samples from rats administered DMA$_{\text{V}}$ in the diet.

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DETERMINATION OF A COMPLEX BETWEEN MONOMETHYLARSONOUS ACID AND 2,3-DIMERCAPTO-1-PROPANE SULFONATE IN HUMAN URINE

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Sodium 2,3-dimercapto-1-propane sulfonate (DMPS) has been used as an antidote to treat acute arsenic and mercury poisoning, presumably by chelating inorganic arsenic and mercury species to increase their excretion from the body. However, the expected complex of DMPS with arsenic or mercury has not been detected in human urine. Here we describe a technique for the determination of a complex between DMPS and monomethyolarsonous acid (MMA\text{III}) and show the presence of DMPS- MMA\text{III} complex in human urine after administration of DMPS.

The technique is based on high-performance liquid chromatography (HPLC) separation, on-line digestion, and hydride generation atomic fluorescence detection (HGAFS). The DMPS- MMA\text{III} complex did not form volatile hydride with borohydride treatment. A digestion procedure using 0.1 M NaOH decomposed DMPS-MMA\text{III} complex, making it amenable for subsequent HGAFS detection. Arsenite (As\text{III}), arsenate (As\text{V}), monomethylarsonic acid (MMA\text{V}), dimethylarsinic acid (DMA\text{V}), MMA\text{III}, and DMPS- MMA\text{III} complex were analyzed in urine samples from human volunteers ingested 300 g DMPS. Previous studies suggested that formation of DMPS-MMA\text{III} complex might inhibit the methylation from MMA\text{V} to DMA\text{V}, leading to the observed increase in MMA\text{V} and decrease in DMA\text{V} in urine, although DMPS-MMA\text{III} complex had not been detected. The finding of DMPS- MMA\text{III} complex reported here provides direct evidence in support of this hypothesis.

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THE RELATIONSHIP BETWEEN ARSENIC INDUCED HYPERTENSION AND NITRIC OXIDE SYNTHASE AND SUPEROXIDE DISMUTASE GENE POLYMORPHISM IN BLACKFOOT DISEASE HYPERENDEMIC AREA IN TAIWAN

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To explore the relationship among nitric oxide synthase (eNOS) and superoxide dismutase (MnSOD) gene polymorphism, arsenic methylation capability, and the prevalence of hypertension. Three villages, Homei, Fuhsin and Hsinming of Putai Township on the southwestern coast of Taiwan Island were selected as the study area. The residents aged 30 or more years old, who lived at least 5 days a week in the villages were recruited into this study from January to February 1989. Each study subject was personally interviewed by well-trained in standardized interview techniques and uses a structured questionnaire. The history of living in the arseniasis endemic area and duration of drinking artesian well water, together with life style variables including alcohol drinking, cigarette smoking and dietary habit, as well as personal and family history of disease were obtained in interview. In addition, the study subjects were received physical examination and the fasting bloods were collected. Serum and buffy coat were separated and stored in 70°C for the analysis of clinical biochemistry index and gene polymorphism. The serum samples of 61 hypertension patients and 266 healthy controls were collected from 1993 who had urinary arsenic species data were randomly recruited as study subjects in this study. To extract DNA from buffy coat and use polymerase chain reaction (PCR) and restriction fragment length polymorphism (RFLP) to analyze eNOS and MnSOD gene polymorphism. We found that the higher the cumulative arsenic exposure and the lower arsenic metabolism capability increase the risk of hypertension after age, sex, BMI, eNOS gene polymorphism adjustment. In the wild/wild eNOS gene type, the odd ratio of hypertension was related with age, sex, BMI, and arsenic metabolism capability. But, the wild/mutant and mutant/mutant eNOS gene type was not related. On the other hand, in the wild/wild eNOS gene type, the higher the cumulative arsenic exposure and the lower arsenic metabolism capability increase the risk of hypertension after age, sex, BMI, eNOS gene polymorphism adjustment. The same result was shown in the wild/mutant and mutant/mutant eNOS gene type. The result of MnSOD will be presented in the conference.

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ARSENIC MOBILITY CONTROLLED BY THREE REDOX ZONES

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Redox control on arsenic mobility has been demonstrated in numerous aqueous environments, as one might expect based on thermodynamic principles. Recently, the concept of the three redox zones critically for arsenic mobility was further refined, based on experimental results of incubation of water treatment sludge as well as equilibrium modeling. Briefly, the three redox zones are: a) an adsorption zone at pe>0, which is characterized by strong adsorption of As(V) on ferric oxyhydroxide; b) a mobilization (transition) zone at -4.0<pe<0, where arsenic is released due to reduction of ferric oxyhydroxide to ferrous iron and As(V) to arsenite [As(III)]; and c) a reductive fixation zone at pe<-4.0, where arsenic is immobilized by pyrite and other reduced solid phases.

Groundwater data obtained in Araihazar, Dhaka, Sripur, Sonargaon, Ramganj and Senbag of Bangladesh clearly demonstrate that arsenic was mobilized within the transition zone at -4.0<pe<0, assuming that the Eh measurements by Pt electrodes represent the equilibrium conditions. Iron was also liberated in the mobilization zone. We are currently exploring the causes for a wide range of As concentrations (2 μg/L - 1189 μg/L) in a very narrow range of pE (-4<pE<0) using thermodynamic models. One plausible explanation is the variation of As in the solid phases contributes to the range of As concentrations. Competitive sorption of other anions, such as PO4, onto limited Fe-oxyhydroxide surface may also contribute to part of the variability. Understanding the redox conditions and the liquid-solid phase interactions should have broad implications for the global occurrence of arsenic in groundwater.

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REVIEW AND EVALUATION OF THE TOXICITY AND CARCINOGENICITY OF METHYLATED ARSENIC COMPOUNDS

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We have reviewed the scientific literature on monomethyl and dimethyl arsenic compounds (MMA and DMA), focusing on: (1) metabolism, (2) toxicity, and (3) mode of action of DMA carcinogenicity in the rat.

(1) Metabolism. Direct administration of methylated arsenic compounds leads to a metabolism pattern significantly different from that of methylated arsenic compounds formed in the body as a result of metabolism of inorganic arsenic. When MMA or DMA are administered directly, only a small fraction of the dose is further methylated with the rest remaining unchanged.

(2) Toxicity. Although the role of methylation in arsenic toxicity is currently under scientific debate, the in vivo toxicological relevance of recent in vitro findings suggesting higher cytotoxicity and genotoxicity of trivalent methylated metabolites remains an open issue. The stability of trivalent methylated metabolites and their persistence in the body, particularly at the site of toxicity, is unclear; there is no evidence indicating they are present in vivo at key target locations for a sufficient duration to cause toxicity.

(3) DMA Mode of Action. DMA has been shown to be a bladder carcinogen in rats. The available data suggest that the mode of action involves cytotoxicity followed by regeneration, therefore, the carcinogenicity of DMA should be modeled using a non-linear dose-response relationship.

Overall, based on metabolism and toxicity studies, MMA and DMA should be distinguished from each other, and not equated with inorganic arsenic, in human health risk evaluations. In addition, DMA carcinogenicity in the rat should be evaluated using a threshold, or non-linear dose-response relationship.

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ARSENIC SPECIATION IN HUMAN URINE


Humans are exposed to arsenic in different ways with drinking water and food being two major sources. There have been regulations in U.S. and Canada with respect to the daily allowable amount of arsenic intake from drinking water for humans. Common food products from marine sources, such as seafood and seaweed, generally contain high amounts of arsenic. The speciation studies of arsenic absorbed and eliminated by human bodies are important though the former is difficult to achieve.

High performance liquid chromatography-inductively coupled plasma mass spectrometry (HPLC-ICP-MS) is used to determine the arsenic speciation in urine. Studies of the ingestion of seafood and seaweed are described. The results give a better understanding of the risk of dietary intake of arsenic for humans.

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DOUGLAS-FIR TREES AS ARSENIC ACCUMULATORS

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Douglas-fir trees have been shown to accumulate arsenic to a remarkable degree and it has been suggested that they could be used as a biogeochemical indicator for precious metal deposits, particularly gold. The total arsenic content of Douglas-fir trees from the Bridge River area was determined by using ICP-MS and speciation information was obtained by using HPLC-ICP-MS. The results were compared to those of other tree species sampled in the same vicinity. The affinity of Douglas-fir for arsenic was confirmed. Most samples were found to contain inorganic arsenic but DMAA was detectable in some cones.

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PHASE I EVALUATION OF SEVEN ARSENIC REMEDIATION TECHNOLOGIES IN NEPAL

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Massachusetts Institute of Technology

Over the past two years, a coalition of NGOs and water agencies in Nepal has focused attention chiefly on the occurrence of arsenic and its public health effects. A team of Masters of Engineering students at the Massachusetts Institute of Technology has been a partner in this effort, being the first to investigate tubewell treatment options for Nepal. To date, seven different technologies have undergone a Phase I evaluation. Phase I assessment criteria include performance under field conditions in Nepal (i.e. total arsenic removal below the interim Nepal guideline of 50 ppb), cost, sludge, and several aspects of social acceptability (i.e., simple to construct and operate, use of local materials). The technologies investigated to date include five adsorption systems and two co-precipitation/filtration systems: 1) three-gagri with iron filings, 2) jerry can with iron filings, 3) iron-coated sand, 4) & 5) two different systems using activated alumina metal oxides, 6) iron co-precipitation with a ceramic candle filter, and 7) a community-based arsenic treatment plant. Of the seven technologies assessed, the four top-ranked technologies are: three-gagri, iron-coated sand, one of the activated alumina metal oxide systems and iron co-precipitation with a ceramic candle filter. Phase II assessment under pilot project field conditions will involve additional performance criteria (As(III) vs. As(V) removal, microbial contamination), social acceptability to women, the primary users of these arsenic remediation systems, and the economic sustainability of the project.

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ARSENIC(V) REMOVAL WITH RARE EARTH DOPED IRON OXIDE ADSORBENTS

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Ce-, La-, or Zr-doped iron oxide adsorbents were prepared through co-precipitation, and their respective performance for arsenic(V) removal was compared. The cerium-doped iron oxide adsorbent with an Fe$^{3+}$:Fe$^{2+}$:Ce$^{4+}$ molar ratio of 2:1:0.8 demonstrated the highest adsorption capacity. The arsenic(V) removal performance and stability of the new adsorbent was further evaluated under various conditions. Under an initial arsenic(V) of 1.0 mg l$^{-1}$, the adsorption capacity of the cerium-doped iron oxide adsorbent was constant around a value of 16 mg g$^{-1}$ over a wide pH range (3.0-7.0), while the maximum adsorption capacity of 8.3 mg g$^{-1}$ for activated alumina, a conventional adsorbent, was obtained over a narrow pH range around 5.5. Phosphate seriously inhibited the removal of arsenic(V) while fluoride did not compete with arsenic(V) even at an F/As molar ratio as high as 30, suggesting that the adsorption sites for arsenic(V) and fluoride were different. Salinity, hardness, and other inorganic anions such as Cl$^-$, NO$_3^-$, and SO$_4^{2-}$ had no apparent effect on arsenic(V) adsorption.

The surface properties of the new cerium-doped iron oxide adsorbent were investigated with XRD, FTIR, XPS, and some other methods. FTIR spectra of cerium-doped iron oxide adsorbent before and after arsenic(V) adsorption demonstrated that ion exchange between M-OH groups on the adsorbent and arsenic(V) ions in the solution occurred during adsorption.

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EVALUATION OF A NEW ARSENIC-ADSORBING RESIN (WASP)

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The ability of a newly-developed resin to remove both arsenate and arsenite from solution has been investigated. The resin, called WASP, has been developed by a Singaporean company. Both batch and column studies were used in the investigation.

The resin is a mixed inorganic oxide which removes arsenic through an adsorption process. The resin is capable of reducing arsenic from high concentrations (1,000 mg/L) down to low levels (<10 ug/L.) The resin adsorbs both arsenate and arsenite, with adsorption versus pH profiles similar to those for hydrous iron oxides. Arsenate adsorption is greatest at low pH, while arsenite adsorption is highest at neutral pH values. The resin will adsorb around 30 mg As/g at pH 7, with the same capacity for both arsenate and arsenite. The arsenic does not readily desorb from the resin once it is bound, suggesting that it would be difficult to regenerate the material. The results indicate that the resin has good potential for treating arsenic-contaminated water.

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ARSENIC REMOVAL FROM UNDERGROUND WATER


A novel method for the removal of arsenic from underground potable water is suggested using ultrasonic power and a simple chemical treatment. The removal of arsenic even up to 50 ppb level (WHO) has been achieved without much difficulty and within limited resources of instrumental facilities, suiting well to economically weaker people of the arsenic prone areas of the world, mainly, Bangladesh, Vietnam & India. Chemical treatment for the removal of arsenic is usually possible only at very low pH but using this technique, arsenic level could easily be brought to safe drinking limits just by simple chemical treatment and filtration but without changing the pH and other drinking quality parameters of the underground water.

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AN ARSENIC-IRON FILTER FABRICATED FROM NATURAL MATERIALS

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An arsenic filter was developed that uses laterite iron concretions to remove arsenic. The goal was to make an arsenic-iron removal system for less developed countries that costs little to operate, and is fabricated with locally obtained supplies. Laterite from Ghana and Brazil, which ranges in composition from 29 to 50% SiO$_2$, 24 to 40% Fe$_2$O$_3$, and 12 to 17% Al$_2$O$_3$, was studied by means of column experiments. Sorption capacity for 2 mm grains is about 300 bed volumes of 1 ppm arsenic water. Contact times of 10 to 15 minutes reduce arsenic concentrations by about a factor of 100 to 1000, which allows the fabrication of fast-flow filters. Iron sorption was observed during our first field tests. Sorption capacity is > 200 bed volumes for five ppm Fe solutions.

A 20-liter bucket filter, 100-liter bucket filter and two, meter-size filters that directly treat hand-pumped well water were field tested in Ghana. Laterite was hand-crushed, sized with window screen, and washed. Inclusion of abundant fines increases the arsenic sorption capacity. There was no indication that high-silica tropical waters poison the filter. Bucket-filters comprising a plastic bucket, piece of window screen, laterite and board have flow rates about 0.5 to 1 liter/min. Laterite filters remove turbidity and off-tastes from drinking water and do not impart any new taste, hence the product water is readily accepted for drinking. Gravity-flow pump-filters made with cement blocks, window screen, and PVC pipe are at present being tested over a year time and the results will be presented.

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SUPERIOR PERFORMANCE OF NOVEL ARSENIC SORPTION MEDIA AGAINST FERRIC COATED ACTIVATED ALUMINA AND GRANULAR FERRIC HYDROXIDE

Nadim R. Khandaker, Ph.D., David M. Teter, Ph.D., James L. Krumhansl, Ph.D., Patrick V. Brady, Ph.D., Sandia National Laboratories

Public water utilities in the United States of America must achieve compliance with the new arsenic maximum contamination level (MCL) of 10 ppb by 2006. Treating drinking water to this level with existing arsenic removal techniques (coagulation/microfiltration, fixed bed filtration, reverse osmosis), is expected to cost billions of dollars. Therefore, a pressing need exists for arsenic removal methods that are substantially more efficient and cost-effective. Sandia National Laboratories (SNL) has used molecular modeling of arsenic-surface complexation interactions to develop novel sorption media with enhanced anion exchange capacities.

This paper will report on the performance of the SNL media-using batch arsenic sorption tests, Freundlich isotherms and rapid column evaluation against commercially available ferric-coated activated alumina (AA-FS50) and granular ferric hydroxide (GFH). Freundlich isotherm and column data indicate that the SNL-media performance is far superior (six folds higher sorption capacity for arsenic at circum neutral pH) to that of AA-FS50 and GFH.

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FIELD TEST RESULTS OF A NEW ARSENIC REMOVAL FILTER IN BANGLADESH

Professor M. Fakhrul Islam, Ph.D., Department of Applied Chemistry and Chemical Technology, Rajshahi University, Rajshahi, Bangladesh in collaboration with Abdus Sobhan, Sad Ahmad, David B. Nunley, International Development Enterprises, Bangladesh

A locally developed, inexpensive, household-level arsenic removal filter was tested in arsenic-contaminated areas of Bangladesh. The filter media is composed of activated burned clay materials of prescribed sizes. Each filter device requires 20kg of media and can filter up to 32 liters of water per day. The filter is manufactured locally using local materials.

Following extensive, successful laboratory testing using arsenic-contaminated tube-well water, the filter was submitted to tests in households. Approximately 400 filters have been tested in areas where people have few or no alternative sources of safe drinking water. The input and filtered water were analyzed on a regular basis for arsenic and other water parameters, including phosphate, dissolved silica, iron, chloride, sulphate and manganese. The input water varied in its composition of all parameters. The filtered water was monitored regularly for arsenic using the Merck Kit and the results were verified in independent laboratories.

Overall, the filters removed arsenic to below the Bangladesh minimum approved level (0.05ppm) from approximately 25 liters of water per day for an average of 3-4 months. As with every absorption technology, competing ions, in particular phosphate, was shown to negatively affect the filter's performance. A simple pre-treatment was developed and implemented in these households. Further testing with pre-treatment showed that it restored the filter's effectiveness. Leaching tests on the expired media were also conducted using water with pH4-9.5. They showed the expired media to have no negative environmental impact. Field tests are ongoing and field and laboratory results are being independently verified.

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ARSENIC ABSORPTION PROPERTIES OF SOME IMPREGNATED SILICATE MATERIALS

Professor M. Fakhrul Islam, Ph.D. and Md. Mokhlesur Rahman, Department of Applied Chemistry and Chemical Technology, University of Rajshahi, Rajshahi, Bangladesh

Commonly available silicate materials, such as brick sands and sand-cement composites were impregnated with selected compounds and tested for As absorption properties. The materials tested included: (1) FeSO\(_4\)-treated, roasted brick sands, (2) Fe\(_2\) (SO\(_4\))\(_3\)-treated, roasted brick sands, (3) FeSO\(_4\) and Na\(_2\)CO\(_3\)-treated brick sands, (4) Fe\(_2\) (SO\(_4\))\(_3\) and Na\(_2\)CO\(_3\)-treated brick sands, (5) ZnSO\(_4\) and Na\(_2\)CO\(_3\)-treated brick sands, (6) FeSO\(_4\) and Na\(_2\)HPO\(_4\)-treated brick sands, (6) FeSO\(_4\)-treated, roasted waste cement plasters, and (7) iron oxide sand-cement composites. The As absorption capacity of the materials depended on the As concentration of the solution, the time of contact with the material and the interval between contact periods. Absorption capacities for high concentrations of As were increased with longer contact periods. With short contact periods absorption capacities even for low concentrations of As were diminished. The materials absorbed As (III) in solutions with both high and low pH levels. The presence of phosphate in the solution decreased As absorption capacity of the materials.

The most effective materials at removing As with a 20-minute contact period were: (1) Fe\(_2\) (SO\(_4\))\(_3\)-treated, roasted brick sands, (2) FeSO\(_4\)-treated, roasted brick sands, and (3) ZnSO\(_4\) and Na\(_2\)CO\(_3\)-treated brick sands. For a six-hour plus contact period the most effective materials were: (1) FeSO\(_4\)-treated, roasted brick sands, (2) Fe\(_2\) (SO\(_4\))\(_3\)-treated, roasted brick sands, and (3) FeSO\(_4\) and Na\(_2\)CO\(_3\)-treated brick sands. Leaching tests were conducted on arsenic-loaded brick sands using different common chemicals. The tests showed that it is very difficult to leach arsenic from arsenic-loaded iron oxide impregnated brick sands.

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THE HEALTH EFFECT OF ARSENIC: ITS IMPACT, TRACE ESTIMATION AND REMOVAL

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The health exposure and impacts of arsenic due to its presence in drinking water in Eastern India has been evaluated and toxicological effects are discussed. A novel new method for its trace determination in drinking water and ground water was developed. Arsenic is determined in air, water, soil, water, blood and environmental samples with a detection limit of 1 ng per ml in presence of several cations and anions.

A new polymer based calix[6]arene has been synthesized for the removal of arsenic from drinking water as low as ng level. The arsenic was also determined in the blood samples of more than 600 persons exposed to the arsenic poisoning and its impact is discussed.
ARSenic severity in groundwater of HIZLA, BANGLADESH AND THE SUITABLE METHOD OF ITS REMOVAL

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Abstract

Total arsenic concentrations in groundwater of more than hundred hand-pump tubewells were determined, and several soil and biological samples (vegetables, fish, and meat) from potentially arsenic affected area Hizla, Bangladesh; were analyzed. About 80 per cent of shallow (10-30 m) but not deep tubewell (more than 200 m) water was found to be contaminated (As conc. > 0.05 mg/L). Arsenic concentration in groundwater was found to increase proportionally with iron and but decrease inversely with tubewell depth. All analyses except water were executed by X-Ray Fluorescence method at Chemistry Division, Atomic Energy Center, Dhaka, Bangladesh. Total arsenic determination of groundwater was carried out by field kit and cross-checked by Total Reflection X-Ray Fluorescence method. The groundwater is the major source of drinking water in Bangladesh containing significant amount of iron, which substantially helps remove a high amount of arsenic from groundwater while it is treated aptly. Arsenic pollution in drinking water has been observed in numerous countries of the world. Despite this universal occurrence, the provisions of sustainable arsenic removal techniques from contaminated water have been found to be extremely limited. Numerous chemical and biological methods are available with various constraints including high cost, complex operation, and management of highly concentrated arsenic waste. Poverty is one of the grave problems in Bangladesh, which demands the low-cost arsenic removal techniques with safe drinking water. This paper, therefore, presents the suitable method, which is economically viable, socially acceptable, and environmentally friendly for the removal of arsenic from groundwater.

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OCCURRENCE OF PUBLIC HEALTH AND ENVIRONMENTAL HAZARDS AND PROPOSED REMEDIATION OF ARSENIC-CONTAINING SOILS AND SURFACE AND GROUNDWATERS AT THE LAVA CAP MINE

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The authors are involved as US EPA-sponsored Technical Assistance Grant advisors to the public on the Lava Cap Mine NPL Superfund site located in Nevada County, California. This mine, which was one of the largest gold mines in California, is a former gold and silver mine that processed an ore with high arsenic content. The ore processing produced large amounts of high arsenic finely divided tailings that, through deliberate discharge and through failure of a log-based tailings dam, have been transported considerable distances in small streams into Lost Lake, which was designed to be a tailings storage area, and beyond Lost Lake into downstream waterbodies. The failure of a log-based tailing stem in January, 1997 resulted in a sudden release of over 10,000 cubic yards of tailings down the stream. Further, there were considerable high arsenic tailings deposited in low-lying areas upstream of Lost Lake. The previous discharges and the failure of the log-based dam resulted in arsenic concentrations in these soils, which range up to about 1,100 mg/kg. The impacted area is a mountainous residential area with private property along the streams and near Lost Lake, having been polluted by tailings containing elevated arsenic concentrations. There is contamination of the groundwaters near the mine tailings deposition area and within the mine with high arsenic. Mine discharge water samples have been found to contain 660 mg/L of arsenic. Contaminated soils had over 7,000 mg/kg of arsenic. The area also has naturally-occurring arsenic in the geological strata which causes some shallow domestic water supply wells to contain elevated arsenic concentrations. The US EPA Region 9 is the lead agency in conducting a remedial investigation feasibility study to define the hazards of the arsenic in the soils, surface waters, and groundwaters, and to develop remediation programs to protect the public and wildlife from the hazards of the arsenic. In November, 2001, the US EPA released for public comment its draft remedial investigation study, and the Agency had just released its draft proposed remediation approaches.

This paper summarizes the results of the US EPA’s field studies, their assessment of the health risk of the arsenic-containing soils and waters, and their proposed remediation approaches. According to the US EPA’s human health hazard assessment, the arsenic concentrations in the soils near Lost Lake are sufficient to be hazardous to those who spend any significant amount of time in these areas. The basic issue that is being addressed is what degree of remediation is appropriate for arsenic-containing soils to protect the public and wildlife from the health hazards of arsenic derived from the mine tailings. A review of these issues will be presented in the papers.
EFFECT OF SPIRULINA ON ARSENICOSIS PATIENTS IN BANGLADESH

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Abstract

A double-blind randomized clinical trial was carried out among Arsenicosis patients of Sonargaon thana of Narayanganj district, to see the effect of Spirulina on Arsenicosis disease resulting from drinking arsenic contaminated ground water through hand pump tubewells in many parts of Bangladesh. Spirulina is microscopic blue green algae, used as a food supplement but it has some therapeutic value in treating some ailments. It is rich in protein, amino acid, beta-carotene, vitamins etc. 50 arsenicosis patients has been identified through simple random sampling from 185 arsenicosis patients of three villages, who had been diagnosed as cases of arsenicosis by the physicians, depending on the presence of visible signs. The patients included male and female of different age. A double blind method was followed during the drug distribution. It was found after distribution that 33 patient got Spirulina and 17 patient got placebo. 3 gm Spirulina per day per person and same dose of placebo was used as drugs for three-month duration and consumption of arsenic free safe water was ensured for both the group (Spirulina and placebo) during the total duration of study. Physical examinations of these patients were done carefully at every fifteen days interval during the intervention using a structured checklist.

After three month it was found that 27 (81.81%) patients showed evidence of improvement by diminishing the visible manifestation among 33 patients who got Spirulina. Statistical analysis showed significant correlation between Spirulina intake and diminishing of visible skin manifestation of chronic arsenicosis (P<0.001).

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CLONING, EXPRESSION, AND MUTATIONAL ANALYSIS OF RAT S-ADENOSYL-L-METHIONINE: ARSENIC(III) METHYLTRANSFERASE

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Methylation of inorganic arsenic (iAs) to yield methyl and dimethyl arsenic is a common feature of its metabolism in many species. In rat liver, a single enzyme, S-adenosyl-L-methionine: arsenic(III) methyltransferase (AsMT), catalyzes both methylation reactions. The predicted amino acid sequence of rat AsMT is similar to those of the predicted product of the cyt19 gene in the human and mouse genomes. The predicted sequences of the rat, mouse, and human proteins contain amino acid motifs commonly found in non-nucleic acid methyltransferases. Noteworthy features of these proteins are the presence of two methyltransferase motif II sequences (IIa & b) and C-terminal regions with conserved cysteines at three of the final eight residues. Rat cyt19 (rcyt19) and human cyt19 (hcyt19) have been cloned into expression vectors; both recombinant proteins methylate iAs, producing methyl- and dimethyl arsenic as minor and major metabolites, respectively. The rate of methylation of iAs is considerably greater for rcyt19 than for hcyt19. Differences in the rate of methylation of the rat and human enzyme may be due to single residue changes in motif IIa and IIb, or to the presence of a six amino acid insert in the hcyt19 sequence. Mutagenesis studies have focused on motif IIa, motif IIb and the cysteine-rich tail of rcyt19. Mutation of the conserved aspartate residue in motifs IIa (D185V) or IIb (D287V) abolishes methyltransferase activity. The I287M mutation in rcyt19 that alters the motif IIb sequence to that of hcyt19 reduces methyltransferase activity to 27.4% of wildtype rcyt19. Mutations C362S, C363S, and C369S reduce protein activity 3.0, 6.2 and 3.6 fold, respectively. Hence, the observed difference in the rate of methylation of iAs in human and rat hepatocytes could reflect between species differences in the amino acid sequence of cyt19 that affect its catalytic function. (This abstract does not necessarily reflect EPA policy.)

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