Mobilization of Arsenic during One-Year Incubations of Grey Aquifer Sands from Araihazar, Bangladesh

Kathleen A. Radloff[1*], Zhongqi Cheng[2], Mohammad W. Rahman[3], Kazi M. Ahmed[3], Brian J. Mailloux[4], Andrew R. Juhl[2], Peter Schlosser[1,2], and Alexander van Geen[2]

[1]Dept. of Earth and Environmental Engineering, Columbia University, New York, NY 10025

[2]Lamont-Doherty Earth Observatory of Columbia University, Palisades, NY 10964

[3]Department of Geology, University of Dhaka, Dhaka, Bangladesh

[4]Barnard College, Columbia University, New York, NY 10025

*Corresponding author:

E-mail: kar2108@columbia.edu

Phone: 845-365-8412; Fax: 845-365-8154

Abstract

Elevated As concentrations in shallow groundwater pose a major health threat in Bangladesh and similarly affected countries, yet there is little consensus on the mechanism of As release to groundwater or how it might be influenced by human activities. The key variable addressed here is the rate at which As is released from Bangladesh sediment. Groundwater and grey sediment were collected simultaneously as diluted slurries between 5 and 38 m, a depth range over which ambient groundwater As concentrations increase from 20 to 100 μg L^{-1}. Arsenic was released to groundwater in slurries from 5 and 12 m depth over one year at a relatively constant rate of 21 ± 4 (2 σ) and 23 ± 6 μg As kg^{-1} yr^{-1}, respectively. Amendment with a modest level of acetate increased the rate of As release only at 12 m (82 ± 18 μg kg^{-1} yr^{-1}). Although the groundwater As concentration was initially highest at 38 m depth, no release of As was observed. These results indicate that the spatial distribution of As in Bangladesh and local rates of release to groundwater are not necessarily linked. Fe release during the incubations did not occur concurrently with As release, suggesting that the two processes may not be directly coupled either. Small periodic additions of oxygen suppressed the release of As from all three depths, indicating that anoxia is a prerequisite for accumulation of As in Bangladesh groundwater.

Introduction

An estimated 100 million people are exposed to elevated levels of As by drinking untreated groundwater throughout South Asia. In Bangladesh, one-third of all wells did not meet the Bangladesh drinking water standard of 50 μg L^{-1} and one-half did not meet the World Health Organization standard of 10 μg L^{-1} before mitigation efforts began (1).

The prevailing view is that elevated As concentrations in anoxic groundwater reflect desorption from Fe oxyhydroxide surfaces as Fe(III) is converted to dissolved Fe(II) or mixed Fe(II/III) surfaces (2,3). Extensive field surveys show, however, that the relationship between dissolved As and Fe concentrations is highly scattered in Bangladesh, even though their concentrations in the sediment are highly correlated (2,4). Significant As mobilization has also been shown to occur with only moderate reduction of Fe(III) minerals and before significant releases of Fe(II) into solution (5-8). More recently, detrital sulfide phases have been proposed as an alternative shallow source of As that is mobilized through redox cycling in near-surface sediments (9).

In addition to uncertainties about the importance of Fe reduction, the role of As reduction *per se* for mobilization is still unclear. Studies using natural sediments and model sediments inoculated with Fe- or As- reducing microbes have shown enhancement of mobilization when arsenate is reduced to As(III) (5,10). But the need for As reduction for mobilization has been questioned as more studies have shown that As(III) is not necessarily more mobile than As(V) under all environmental conditions, particularly when phosphate is present (11,12).

It is generally accepted that microbial activity influences As release, either through direct interactions or indirectly via Fe oxyhydroxides, although the nature of this relationship remains poorly understood (2,13). This is supported by the observation that As mobilization is enhanced by organic substrate additions in both field and incubation studies (10,14,15). Islam and colleagues also observed major shifts in the population of dominant groups of the microbial community in response to organic substrate additions (15). The extent to which large substrate additions are representative of the process of As release under natural conditions is unclear. The provocative notion that irrigation pumping might have enhanced the supply of organic substrate to shallow aquifers in recent decades, hereby triggering the release of As, has also been put forth (14,16).

The motivation for the present study is that some of these unsettled issues could be addressed by knowing the rate at which As is released in aquifers in different geological settings. Previous incubations using natural aquifer sediment have provided valuable mechanistic insights, but intentional or unintentional manipulations of the material have hampered the interpretation of the measured rates as an indication of ambient conditions. A distinguishing feature of the incubations described here is that groundwater and sediment were collected simultaneously using a device coined the needle-sampler, placed immediately under anaerobic conditions in the field, and maintained under such conditions for the 11-month duration of the experiment (17).

Materials and Methods

Site Description. The sampling site (90.6˚ E, 23.8˚ N) is at the center of an expanse of rice paddies in Araihazar upazila, 30 km east of Dhaka. Grey aquifer sediments extend to a depth of 48 m at this location. The radiocarbon age of two wood fragments recovered during drilling of the site in January 2005 (6970±45 and 7180±55 ^{14}C yr at 40 and 44 m depth, respectively) confirm that the aquifer is of Holocene age (18). The generally sandy section of grey material is interrupted by a clay layer between 27 and 34 m depth (Figure 1). Numerous mechanized irrigation wells are distributed across the surrounding rice paddies and draw their water through screens that span the entire shallow grey aquifer (19).

A set of monitoring wells (site F in ref. (20)) located 1 km northwest of the sampling site shows a gradual increase in As concentrations in groundwater of the area from <10 μg L^{-1} at 6 m depth to ~50 μg L^{-1} at 15 m (20). Over the same depth range, groundwater dating using the ^{3}H-^{3}He technique indicates an increase in the age of groundwater from <1 to ~5 yr at the same location (21). Between 15 and 20 m depth at site F, the concentration of As increases rapidly to 200 μg L^{-1} and the age of groundwater increases by several decades.

Collection and Transport. A preliminary survey of the area was conducted in January 2005. A single well was drilled using the traditional "sludger" method and a series of groundwater and sediment samples were collected to a depth of 21 m using the needle-sampler (17). The diffuse spectral reflectance of the cuttings collected every 1.5 m was measured soon after collection in the field to indicate the Fe speciation in the solid phase (20).

The samples used for the long-term incubations were collected in April 2005. The needle sampler was used to obtain slurry samples from three depths: 5, 12 and 38 m. The starting material from each depth consisted of groundwater and sediment contained in two needle sampler tubes collected within 0.3 m of each other vertically. Preferential uptake of groundwater relative to sediment with the needle-sampler increased the porosity of the collected slurries. The porosity of sandy aquifers typically varies between 0.2 and 0.3 in the study area, while the material collected by the needle sampler had a porosity ranging from 0.7 to 0.8. The collected sediment may also have been biased towards the finer fraction because the coarsest particles are excluded by the entry holes to the needle (17).

Before sampling, each needle-sampler tube was flushed with N_2 and then evacuated to <300 mbar. To minimize oxygen exposure, the headspace of each tube was also flushed with N_2 immediately after collection. Aliquots of the collected groundwater were gently pressure-filtered in the field, also under N_2. The needle-sampler tubes were then stored in plastic, N_2-flushed, air-tight anaerobic boxes (Becton-Dickinson) containing pouches that generate H_2 and consume O_2 with a catalyst. Three days after collection, the needle-sampler tubes were transferred to an anaerobic chamber (Coy Laboratories) where they remained for the duration of the experiment. The chamber was kept under a N_2 atmosphere containing 1-2% H_2.

Amendments. The two needle sample slurries from each depth were combined, homogenized and divided into five 60-mL amber glass bottles in the anaerobic chamber 17 days after collection. Each microcosm bottle contained between 8 and 15 mL of groundwater and 5-11 g of sediment. The sample bottles were sealed with a butyl rubber stopper and an aluminum crimp. The bottles were gently agitated over the next 11 months and kept at room temperature. To minimize the potential effect of H_2 contained in the anaerobic chamber on the incubations, the bottles were flushed with ultra-high purity N_2 for several minutes after every sampling.

The slurries from each depth were treated in three different ways: without amendment, with the addition of acetate, and with periodic, small oxygen additions, hereafter referred to as unamended, acetate and oxygen incubations. The acetate addition (14 μmole per sample, which corresponds to ~2 μmole g^{-1} sediment or ~1.5 μmole L^{-1} groundwater) supplied enough electron donors to potentially reduce approximately 10% of the acid-leachable Fe present in the sediment.

The oxygen additions occurred at each sampling point, adding approximately 1.2 mL of air (i.e. 10.3 µmole of O_2, equivalent to ~0.21 mg O_2 L^{-1} groundwater). The oxygen addition to the groundwater could in principle oxidize 18 µg of Fe after each sampling, which provides enough oxygen to remove more than 1.5 mg L^{-1} Fe from solution. The porosity of the different slurries was gradually reduced from 0.6-0.9 to 0.4-0.8 by sampling water 7 times over the course of the experiment.

Analyses. Groundwater samples collected from each incubation on days 17, 23, 40, 99, 160, 252 and 340 provided a detailed time series of variations in the concentrations of 10 dissolved constituents including As, Fe, Mn, S and Si as well as the major cations Na, K, Mg, Ca (Supporting Information). Aliquots of 1.2 mL were drawn from the slurries, filtered through 0.45 µm syringe filters, acidified to 1% HCl (Optima) and analyzed by High-Resolution Inductively Coupled Plasma Mass Spectrometry (HR ICP-MS) (22).

Aquifer sediment was analyzed on day 17 after collection, when the slurries were divided into amendments, and again on day 340 at the completion of the experiment. Two extractions were used to assess the availability of As in the solid phase. A 1 M phosphate solution was used to estimate the amount of adsorbed As (23). A hot 1.2 M HCl solution was used to release As more tightly bound within amorphous Fe minerals (24). The extracts were analyzed for As using HR ICP-MS (22). Iron speciation of the HCl leachates were quantified on day 17 and day 340 using the ferrozine method (20). Arsenic speciation in the phosphate extract was also determined after dilution using anion exchange columns (25), but only on at the completion of the incubations.

Results

Site Profile. Dissolved As concentrations generally increase with depth from 20 to 100 μg L^{-1} in the grey aquifer, although there is considerable variability between some closely spaced samples (Figure 1). The absence of oxygen in the groundwater is indicated by detectable levels of dissolved Fe and Mn concentrations over the entire depth range (Supporting Information). The profile of sediment reflectance documents the increasingly reduced state of the aquifer as a function of depth and is consistent with a rise from ~0.5 to 0.84 in the proportion of Fe(II) to total Fe in the sediment leachates. The concentration of P-extractable As in the sediment was highly variable in the upper 15 m (0.1-2.7 mg kg^{-1}). The three samples used for the incubations initially contained 40, 72 and 88 μg L^{-1} dissolved As and 0.5, 1.4 and 0.3 mg kg^{-1} P-extractable As at 5, 12, and 38 m depth, respectively.

Iron. The unamended and acetate incubations all experienced 10 to 85-fold increases in dissolved Fe concentrations between day 17 and 252, followed by an almost equally large loss of Fe by day 340 in the majority of samples (Figure 2). Over 90% of the total Fe released occurred after day 99 of the incubations. The proportion of Fe (II) to total Fe in the HCl leachates remained nearly unchanged for the anoxic samples, with the exception of the unamended sample at 12 m for which Fe(II)/Fe decreased from 0.7 to 0.5. After day 40, the oxygen addition (about 20.6 μmole) maintained dissolved Fe concentrations below the detection limit of 0.01 mg L^{-1} at all three depths. The leachable Fe(II)/Fe ratio in the sediment also decreased from 0.6-0.8 to 0.4-0.5 in the incubations amended with oxygen.

Arsenic. From the time of collection to day 17, the concentration of dissolved As declined slightly from 40 to 28 μg $L^{-1}$and 72 to 38 μg L^{-1} in the slurries from 5 and 12 m depth, respectively. Dissolved As then gradually increased to 50-90 μg L^{-1} in the unamended and acetate samples from the same depths (Figure 2). The loss of dissolved Fe on day 340 was accompanied by a slight decrease in dissolved As in the unamended incubation from 5 m and in the acetate incubation from 12 m. There was no systematic difference in As concentrations between the unamended and acetate incubations at 5 m, while at 12 m the acetate amendment roughly doubled the amount of As released. At 38 m, As concentrations in the amended and acetate samples increased by ~25 μg L^{-1} between day 0 and day 17 and remained constant or slightly decreased thereafter. After day 99 of incubation, the oxygen additions maintained dissolved As concentrations <6 μg L^{-1} at all depths.

The concentration of P-extractable As in the sediment was determined at day 17 and 340; HCl-extractable As was determined only at day 340 (Supporting Information). In sediment from 5 m, the P-extractable As concentration remained steady at 0.5 mg kg^{-1}. Samples from 12 m had the highest level of P-extractable As of 1.4 mg kg^{-1}, which decreased to about 0.6 mg kg^{-1} at the end of the experiment. At 38 m, the P-extractable fraction decreased slightly from 0.30 to 0.24 mg kg^{-1} between day 17 and 340. The observed decreases in P-extractable As concentrations in the solid phase can not be explained by the measured releases of As to groundwater because the releases are equivalent to only 2-6% of the total present in P-extractable form. At the end of the incubations, the P-extractable fraction contained predominantly As(V), the proportion of which increased with depth from 50% to 80% in the anoxic samples. There was no discernable difference in As speciation for the anoxic amendments at each depth. The oxygen incubations from each depth, on the other hand, all contained ~90% As(V) in the P-extractable fraction. The

HCl-extractable As concentration in the solid phase at the end of the experiment was comparable at the three depths (1.3-1.5 mg kg^{-1}).

Discussion

Iron Release and Precipitation. To the best of our knowledge, these incubations are the first to document the release of Fe from natural aquifer solids over an extended period of time under conditions close to those encountered *in situ*. In relation to subsequent changes, dissolved Fe concentrations remained constant during the first 17 days of the incubations, suggesting that the system was not excessively perturbed by sampling, transport and manipulation. By day 99, oxygen additions had already drastically decreased dissolved Fe concentrations to <0.01 mg L^{-1} due to oxidation and precipitation of Fe(II) initially present in the groundwater. A 1.5 to 15 μg L^{-1} rise in dissolved S concentrations in the oxygen incubations before day 99 (Supporting Information) suggests the first aliquots of oxygen were also consumed by labile sulfides, which have been observed in Bangladesh aquifer material (14). The addition of oxygen likely suppressed the microbially-mediated reduction of Fe. The sharp contrast in the behavior of Fe in solution suggests that anoxic conditions were successfully maintained over the course of the other amendments.

The large release of Fe observed in the six anoxic incubations between day 99 and 252 suggest an evolution of the system relative to initial conditions in the aquifer, which are presumed to be at steady-state. The changes can be interpreted in the light of previous laboratory incubations using model solids. Such studies have shown that Fe(II) may not be released during the early stages of Fe(III) reduction but can instead adsorb onto the oxide surface or form secondary minerals (26,27). The large increase in dissolved Fe concentrations that followed and continued until day 252 may therefore indicate that Fe(II) was released directly to solution only after binding sites on the solid became saturated. The delay in this release suggests saturation had not been reached *in situ*, presumably because groundwater flow maintains a lower total level of Fe(II) in the aquifer. The maximum Fe concentrations reached during the anoxic incubations are higher than typically observed in Bangladesh groundwater, but similar levels have been observed in Vietnam (28). The rise in Fe concentrations corresponds to a release of 0.2-50 mg Fe kg^{-1} of sediment and corresponds to only 0.01 to 0.8 % of HCl-extractable Fe. The limited impact of the acetate amendment suggests that organic substrate availability does not limit Fe reduction in these shallow aquifers (29).

Increases in dissolved Fe concentrations may also have been inhibited by another process. Laboratory studies have shown that the rate of microbial Fe reduction is primarily controlled by the surface site density of Fe(III) minerals (30). Re-sorption of Fe(II) onto the oxide surface can occur to such a degree that further Fe(III) reduction is inhibited (26). Either mechanism, or a combination thereof, could have buffered dissolved Fe concentrations within a certain range over the course of the incubations.

The large drop in dissolved Fe concentrations for 5 out of 6 anoxic incubations between days 252 and 340 suggests that saturation with respect to a solid phase had been reached (27). Solubility calculations and a concurrent 2 to 40 μg L^{-1} reduction in dissolved P concentrations suggest the formation of vivianite or another Fe-P mineral (Supporting Information). It is noteworthy that the decline in dissolved As in response to significant Fe precipitation was modest, probably due to competition with other groundwater constituents for limited binding sites. Secondary minerals are typically also more crystalline and provide a smaller surface area for sorption (12,31).

Effect of Changing Porosity on As Adsorption. We use a formulation equivalent to a Langmuir isotherm to estimate the potential effect of increasing the porosity from 0.2 to 0.3 in the original aquifer to 0.6-0.9 in the needle-sampler slurry, and then back to 0.4-0.8 by the end of the incubations. Adsorption of As is assumed to be controlled by a simple mass-action law of the form: S + A = SA and an adsorption coefficient K_{ads} = [SA]/([S][A]), where S represents the concentration of surface sites in the slurry and A is the concentration of dissolved As. In addition, the two forms of As must satisfy mass-balance: AsT = A + SA, where AsT represents the total concentration of As in the slurry. The concentration of dissolved As (A) was measured in the sample filtered in the field right after collection; the concentration of P-extractable As is used as a proxy for adsorbed As (SA). For each of the three depths, adsorption coefficients were calculated assuming equilibrium at the initial conditions in the aquifer assuming a range of saturations for surface sites (Supporting Information). Using this model, the decline in As concentrations observed between day 0 and day 17 in slurries from 5 and 12 m depth can be attributed solely to the increase in porosity assuming a site saturation of ~95%. Such a high degree of saturation of surface sites with respect to As adsorption is consistent with independent observations (32). The same adsorption model predicts that dissolved As concentrations should be relatively insensitive to changes in porosity in the 0.3 to 0.7 range. Consequently, we attribute the increases in As concentrations observed after Day 17 to an active mobilization process rather than mere desorption.

Implications for Arsenic Release Mechanism. The noticeable impact of oxygen additions on the incubations of aquifer material from all three depths confirms that the formation of fresh Fe(III) adsorption sites is sufficient to significantly limit the release of As to solution. Anoxia therefore appears to be a prerequisite for accumulating As in groundwater of Bangladesh. This does not mean that releases of As and Fe to groundwater are necessarily coupled or that anoxia alone is sufficient to cause As mobilization. The time course of the anoxic incubations provides new evidence of decoupling between As and Fe mobilization. The release of As at 5 and 12 m was evident by day 40, while Fe release did not begin until day 99 and rapidly increased after day 160. The lack of Fe release could be due to Fe re-sorption, as described above. The large release of Fe to groundwater by day 252, however, did not cause a concomitant increase in As concentrations. Again, this could be because the adsorption capacity of the solid phase for Fe had been exceeded or, alternatively, because As was not distributed uniformly within Fe oxyhydroxide surfaces. In either case, the observations suggest that the processes regulating As and Fe release may have little in common once an aquifer becomes anoxic.

Average rates of As mobilization were calculated for each anoxic incubation by excluding samples showing a large loss of Fe from solution. The entire 340 days were considered for the acetate incubation at 5 m since the decrease in Fe was minimal at the last sampling. Only the first 252 days were included for the unamended incubation at the 5m depth and both anoxic incubations from 12 m and 38 m (Figure 3). Rates of release were determined by least-squares regression; the uncertainty is listed as two time the standard deviation (σ) of the slope. The combined rate of As release was 21 ± 4 μg As kg^{-1} sediment per year (r^2= 0.91, n= 9) for the two anoxic incubations from 5 depth; the calculated rates were not statistically different between the types of amendment. At 12 m, the acetate addition increased the rate of As release nearly 4-fold from 23 ± 6 μg As kg^{-1} yr^{-1} (r^2= 0.93, n= 4) to 82 ± 18 μg kg^{-1} yr^{-1} (r^2= 0.95, n= 4) for the unamended and acetate incubations, respectively. The combined set of observations indicates that the process of As mobilization in Bangladesh is probably microbially mediated, although not necessarily limited by the availability of organic substrate. The speciation data for As in the P-extractable fraction at the end of the incubations indicates that aquifer anoxia does not necessarily result in As(V) reduction in the solid phase and that such reduction is not a requirement for mobilization.

The incubations unambiguously demonstrate that As is not necessarily released at higher rates in shallow grey aquifers of Bangladesh where dissolved As concentrations are elevated. Whereas groundwater As concentrations increased with depth at the sampling site (Figure 1), As release was only observed in aquifer material from 5 and 12 m depth. The sample from 38 m had the highest initial As concentrations in groundwater and yet no release was observed, even though the release of Fe was comparable at all depths. The low level of P-extractable As contained in aquifer material from 38 m suggests that As release was limited by the lack of a sufficient pool of mobilizible As in the solid phase. According to the rates of As release measured at 5 and 12 m depth, an initial pool of ~10 mg kg^{-1} mobilizible As (2,33) could be lost from an aquifer due to groundwater flow, as previously proposed to explain low groundwater As concentrations in older grey sediment (18,34), in as little as 500 years.

Re-Interpreting Spatial Patterns of Groundwater As. Porosity differences and adsorption must be considered when estimating the release rate observed in incubations to increases in dissolved As concentration in the aquifer. The lower *in situ* porosity of aquifers means that the release will have a greater affect on dissolved As concentrations than observed in the incubations. This is balanced to some extent by the adsorptive re-equilibration that governs the partitioning of As between the groundwater and aquifer solids. Considering these two competing effects on the basis of the Langmuir model described above, the observed release rates of As would be equivalent to 57 μg L^{-1} per year at 5 m and 87 μg L^{-1} per year at 12 m (the increase in groundwater concentrations at 5 m is lower than at 12 m, although the release rates are similar due to the higher adsorption coefficient of the sediment at this depth; see Supporting Information). Assuming that a shallow aquifer is recharged with surface water that is initially low in As, the implication is that the As content of groundwater only a decade old could already pose a serious health threat. The short time scale of As release suggests that the distribution of As in shallow aquifers could change over time, particularly in a region like the present study area where irrigation pumping is likely to have drastically affected flow patterns (19).

A recent study provides evidence that the predicted rise of As concentrations in groundwater over time, indicated by these long-term incubations, may be realistic. Shallow groundwater was dated by the ^{3}H-^{3}He technique at a set of six nests of monitoring wells in the same general area of Araihazar (21). Whereas the age of groundwater increased with depth at all six sites, the rate of increase varied considerably from one site to the other most likely in response to variations in local recharge. Comparison of groundwater ages ranging from <1 to over 30 years with groundwater As concentrations of 1 to 520 μg L^{-1} measured in the same set of wells, however, results in a relationship that is fairly consistent across all sites and corresponds to a release rate of 19 ± 2 μg L^{-1} As per year. The entirely independent measure of the rate of As build-up in groundwater is within a factor of 3 to 5 of the rates measured in the unamended incubations. Elevated groundwater As concentrations observed at 38 m depth at our study site and the absence of As release could therefore be an indication of transport of As released at shallower depths. In accordance with the present findings, the groundwater age study also found little relationship between the rate of As release and other aquifers parameters, such as dissolved Fe or the proportion of Fe(II) in the leachable Fe fraction, that were measured (21).

Our incubations of Bangladesh sediment at close to *in situ* conditions over an extended period provide new evidence that the concentration of dissolved Fe in groundwater is not necessarily linked to the rate of reductive dissolution of Fe oxyhydroxides. This may be one reason why there is no simple relationship between concentrations of Fe and As in anoxic groundwater (2,9,10,20,32). The limited impact of acetate on incubations of natural aquifer material suggests that the release of both Fe and As is not necessarily limited by the supply of organic substrate. Flow patterns and groundwater age may therefore play a dominant role in setting the highly variable distribution of As in shallow groundwater of Bangladesh.

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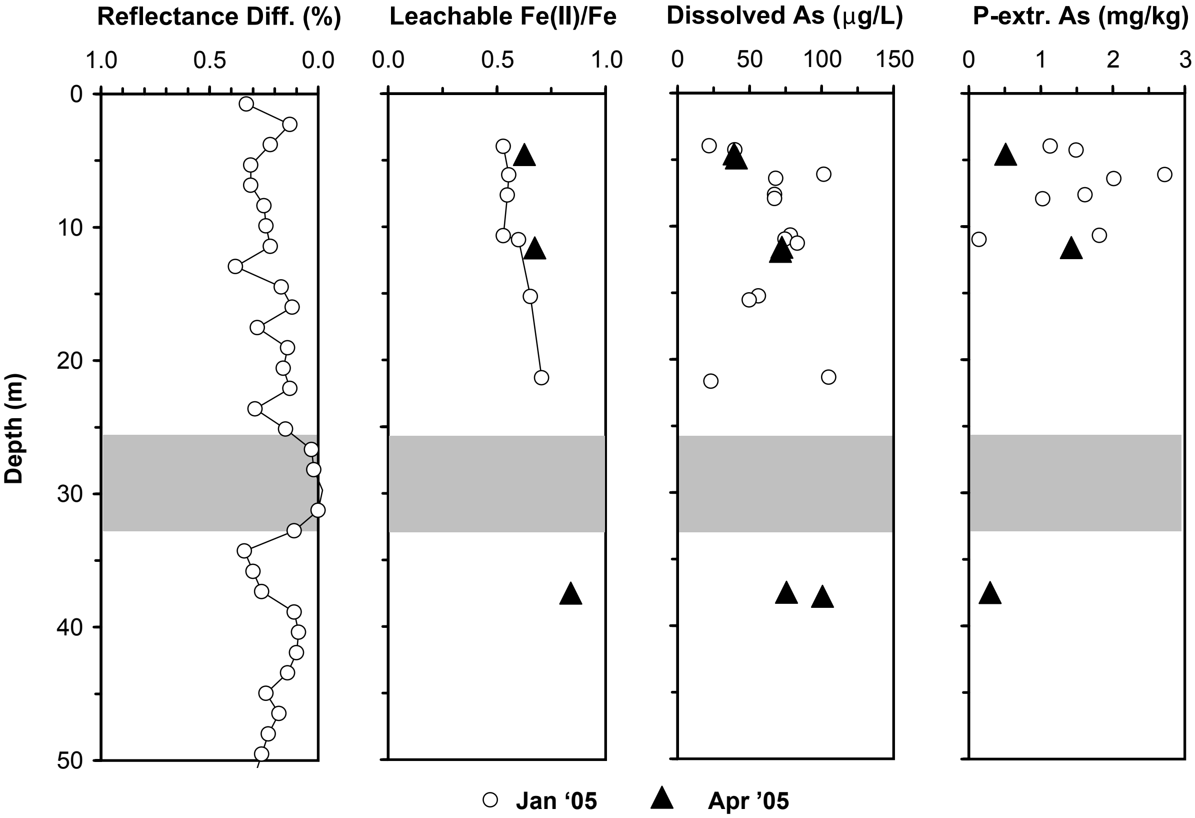
Figure captions

Figure 1. Profiles of sediment and groundwater at the study site in Araihazar, Bangladesh: (a) difference in diffuse spectral reflectance at 530 and 520 nm measured on-site for cuttings recovered during drilling; (b) proportion of Fe(II) in the HCl-leachable Fe fraction of sediment samples; (c) dissolved As concentrations in groundwater; (d) concentration of P-extractable As in the sediment. With the exception of the cuttings used for reflectance measurements in January 2005, all sediment and groundwater samples were obtained with the needle-sampler in January (open circles) and April (black triangles) 2005. The grey shading indicates the extent of a thick clay layer encountered at the site.

Figure 2. Variations in dissolved Fe and As concentrations over the course of the 11-month long incubations of sediment and groundwater slurries collected in Bangladesh at three different depths in April 2005. Note the logarithmic scale for dissolved Fe. Disconnected symbols indicate samples exhibiting a large drop in dissolved Fe at the completion of the incubations.

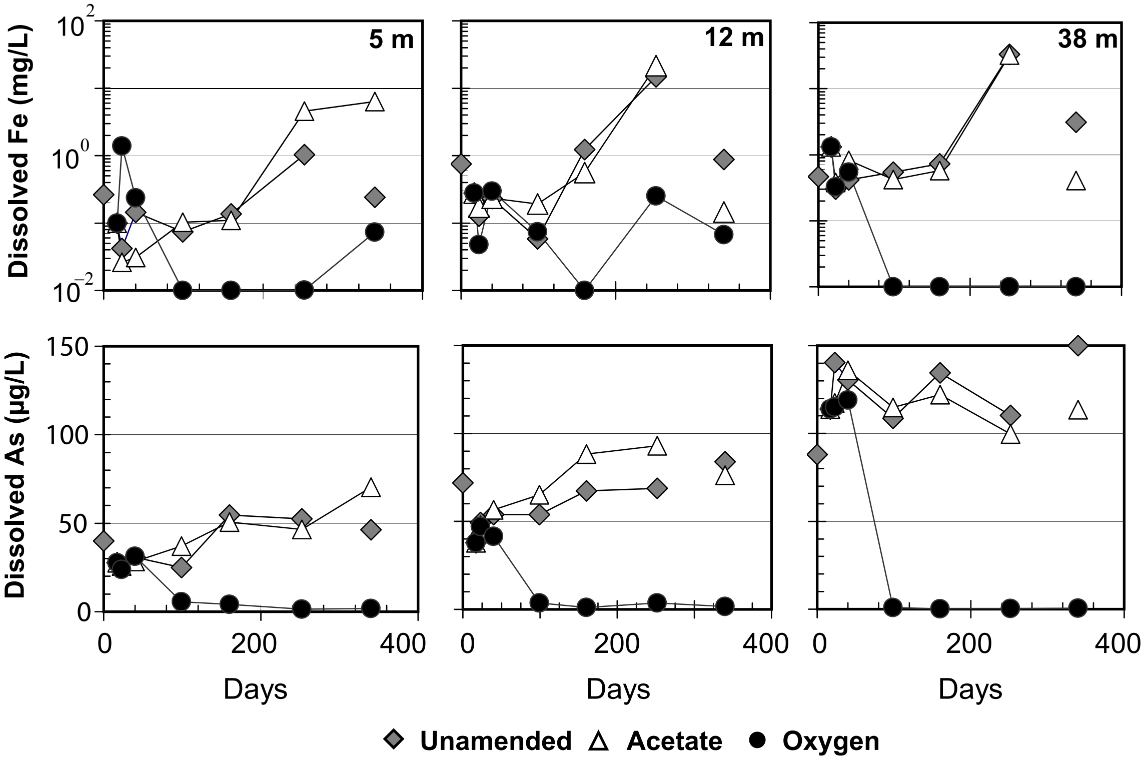
Figure 3. Amount of As released to groundwater for anoxic incubations normalized to the quantity of sediment in each slurry of Bangladesh aquifer material. The rates of release were calculated by least-squares regression fit over the time span, as indicated by the regression lines. The regression for the 5 m depth combined the unamended and acetate incubations, while the regressions were calculated separately at 12 m and 38 m.

Figure 1



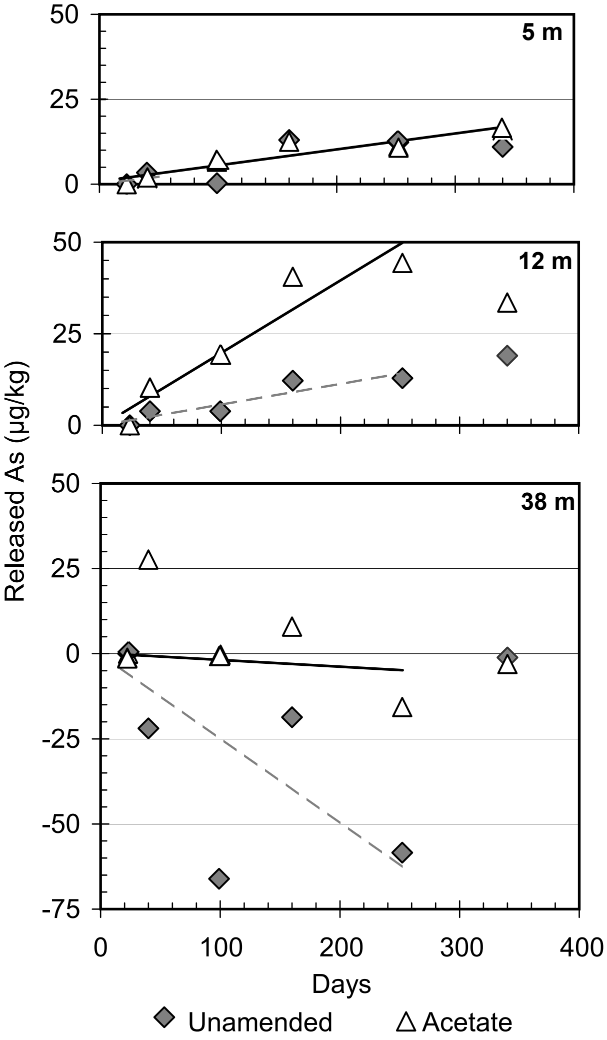
Reflectance Diff. (%)
Leachable Fe(II)/Fe
Dissolved As (μg/L)
P-extr. As (mg/kg)
1.0
0.5
0.0
0.0
0.5
1.0
0
50
100
150
0
1
2
3
0
10
20
30
40
50
Depth (m)
○ Jan '05
▲ Apr '05

Figure 2



5 m
12 m
38 m
Dissolved Fe (mg/L)
10^2
10^0
10^{-2}
Dissolved As (μg/L)
150
100
50
0
0
200
400
Days
◆ Unamended △ Acetate ● Oxygen

Figure 3


5 m
12 m
38 m
50
25
0
-25
-50
-75
0
100
200
300
400
Days
Released As (µg/kg)
◆ Unamended
△ Acetate