

Role of In-situ Reducing Bacteria on Arsenic Mobility in Aquitard Groundwater

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Abstract—In southwestern Taiwan, groundwater arsenic (As) originated primarily in aquitard formations of shallow marine sequences. The most plausible mechanism of the As release to groundwater is the reductive dissolution of As-rich Fe hydroxides, which is driven by reducing bacteria-mediated geochemical reactions. The purpose of this study is to evaluate the contribution of FeRB to the As mobility and transformation. An experiment that involved As-contained synthetic amorphous Fe(III) hydroxide (HFO) and the inoculation of in-situ Fe-reducing bacteria (FeRB) was performed. The results of respective HFO and As(V) reduction experiments showed that FeRB contains varied Fe- and As-reducing bacteria and promotes both reduction reactions. The rates and extents of HFO reduction are different with various organic substrates. In the experiment on As-containing HFO reduction, the competition of organic carbon for sorption sites plays an important role on As mobilization. The results suggest that Fe- and As-reducing bacteria are important to the As mobilization in the presence of acetate. However, the increase in the As(III) concentrations with time for various organic substrates in the As-contained HFO-reducing experiment differ from the rates of As(V) reduction with various organic substrates in the As(V)-reducing experiment. The aqueous As(V) acts as an electron acceptor and reduced to As(III) after the solid phase of Fe(III) reductively dissolved.

Keywords—Arsenic; Groundwater; Reducing bacteria; Iron hydroxides; Mobilization

I. INTRODUCTION

In 1961, elevated concentrations of arsenic (As) and blackfoot disease were first found in southwestern Taiwan [1]. This disease was associated with the direct ingestion of deep well water with high As contents [2]. From 1999 to date, the Water Resources Agency of Taiwan undertook a comprehensive groundwater quality survey at monitoring wells in the southern Choushui river alluvial fan, in the southwest part of Taiwan. The As concentrations of groundwater in this area were 0.12 ± 0.14 mg/L and around 70% of monitoring wells exceeded the World Health Organization (WHO) guideline of 0.01 mg/L [3]. High groundwater As concentrations occurred in shallow (20-70 m) aquifers and the origin is from the uppermost aquitards formed in the Holocene transgression, whose geologic ages are in the range of 3000 to 9000 yr [4]. Nowadays, most inhabitants of this region do not use groundwater for drinking, but it is still

extensively served for aquacultural needs, posing a potential health risk to humans [5].

Reduction condition is the critical factor for As mobilization in shallow groundwater of southern Choushui river alluvial fan. High total organic carbon (TOC) and inorganic carbon (bicarbonate) concentrations were accompanied with the elevated groundwater As concentration [6]. Arsenic mobility is plausibly related to the recent inflow of carbon through displacement by carbonate or the organic carbon-driven reduction of Fe oxide [7]. Based on the determination by X-ray photoelectron spectroscopy (XPS), goethite, hematite and magnetite were the primary Fe minerals of core samples in the distal-fan area [8]. Sequential extraction data and the XRF analysis indicated that Fe oxyhydroxides and sulfides were likely to be the major sinks of As. Generally, the reductive dissolution of Fe oxyhydroxides, which accompanied high aqueous TOC, HCO_3^- and NH_4^+ concentrations, was likely the principal release mechanism of As into groundwater in this area [6], which was similar to that in the West Bengal and Bangladesh [9].

Microbial degradation of sedimentary organic matter is regarded as a biogeochemical factor of the reductive dissolution of As-rich Fe (hydr)oxides in anaerobic groundwater [10]. Many As- and Fe-reducing bacteria and Achaeta have been identified from groundwater and hot spring water [11]. In the Ganges Delta, Fe(III)-reducing bacteria (FeRB), mostly of the Geobacter species, stimulated the reduction of Fe(III), followed by the reduction of As(V) and the release of As(III) [12]. Furthermore, the oxidation state of As at the onset of Fe reduction is significant in As mobilization, because As(V) and As(III) sorption differ at environmentally relevant pH values [13]. Mineralogy and the accumulation of Fe(II) reaction products, adsorbed or precipitated Fe(II) on the mineral surface, affect the rates of reductive dissolution of Fe (hydr)oxides [14]. These reactions affect the transformation and/or mobilization of iron and arsenic products. In the southern Choushui river alluvial fan of Taiwan, bacterial strains of arsenate reducer and arsenite oxidizer have been isolated from in-situ groundwater sample for the determination of As resistance and the development of bioremediation [15][16]. However, the isolation of FeRB strains and the associated geochemical processes of As mobility were less considered. To investigate the influences of FeRB on As mobility and

transformation, batch experiments involved As-contained synthetic amorphous Fe(III) oxide (HFO) mixed with the inoculation of FeRB cultured from field groundwater were conducted in this study. Concentration changes of total As, As(III), As(V) and Fe(II) were measured throughout the experiment. Results of this study can provide a foundation for future biogeochemical investigations.

II. MATERIALS AND METHODS

A. Groundwater Sampling and Enrichment

The southern Choushui river alluvial fan is located in the southwestern part of Taiwan. Based on subsurface hydrogeological analysis to a depth of around 300 m, the formation is divided into three marine sequences and three non-marine sequences in the distal-fan and the mid-fan areas. The non-marine sequences of the formation, with coarse sediment, ranging from medium sand to highly permeable gravel, are typically regarded as aquifers, while the marine sequences of the formation with fine sediments, are regarded as aquitards. More detailed hydrogeological characteristics of this area have been described by [4]. According to previous surveys from 1992 to 2005, the groundwater As concentrations was highest at monitoring well YL7 with an averaged As concentration of $450.4 \pm 314.3 \mu\text{g/L}$. The screen of YL7 was located in the uppermost aquitard at depths of 18-22m. In this study, the groundwater sample from this monitoring well was collected for the culture of FeRB.

The field sampling methods are consistent with the NIEA code W103.50B of the Taiwan Environment Protection Agency. The enrichment experiments targeted Fe(III)-reducers: 1 ml of groundwater on site was injected into autoclaved and sealed Balch tubes, which contained 10 ml FeRB growth medium. This growth medium was modified from [17]. During the preparation of fresh medium, the pH was adjusted to 7.2 by adding concentrated NaOH or HCl. To purify the community of FeRB, the FeRB was enriched in an anaerobic chamber at room temperature and resubcultured in fresh medium at least four times. The mix-cultured FeRB before isolation was adopted in the following experiments to comprehend the complicated geochemical reactions.

B. Preparation of HFO and As-Adsorbed HFO

Fresh HFO was synthesized for each experiment using the following procedure [18]. A solution of 0.5M NaOH was added dropwise with stirring to 100ml of 0.05M $\text{Fe}(\text{NO}_3)_3$ until the solution stabilized at pH 7.5-8. The suspension was equilibrated for 30 minutes with constant stirring, and the pH drift was compensated for by adding 0.5 M NaOH. The HFO was not autoclaved following synthesis, to prevent mineralogical change. For As-contained HFO, 40 mg of HFO was resuspended in 50mL of 0.334 mM Na_2HAsO_4 (Sigma) at pH 7.2 to ensure that all available surface sites for As sorption were saturated with As(V) at the given pH. In all cases, the HFO and As-contained HFO was washed at least four times after preparation using sterile water and centrifuged at 4,000 rpm for 20 min to remove excess

particles and other cations that had not been sorbed. The pH was adjusted to 7.2 for all incubation experiments by adding 0.5 M NaOH prior to inoculation.

C. Incubation Experiments and Analysis

Three reduction experiments were conducted with different electron acceptors to determine the reductive reaction driven by cultured FeRB, including (1) fresh HFO slurry without pre-adsorbed of As, (2) aqueous As(V), and (3) As-contained HFO slurry. All analytical procedures of experiments were in duplicate. For fresh HFO reducing experiment, 40mg HFO slurry was resuspended directly to 200ml sterile water and adjusted to pH 7.2 in acid-washed and autoclaved plastic bottles with screw-cap lids. For As(V) reducing experiment, 200ml of 0.334 mM As(V) solution without HFO slurry was prepared in bottles. For As-contained HFO reducing experiment, 40mg pre-adsorbed HFO slurry was resuspended to 200 ml sterile water in bottles. Each experiment contained four subsets, including biotic and abiotic subsets with 0.5 mM of citrate or acetate, used as the organic substrates for cell growth. The abiotic subsets were conducted as the controlled experiment to ensure that no contamination was introduced during the synthesis, washing, distribution, and sampling of the HFO. The biotic subsets were inoculated in the anaerobic chamber (80% N_2 , 10% CO_2 , 10% H_2) and then incubated at 27°C for the rest of the experiment. All sealed bottles were transferred to an anaerobic chamber before experiment and equilibrated for 24 hrs to allow passive gas exchange before inoculation. The bottles were not stirred during incubation but were shaken vigorously before each sampling.

Water samples were filtered through 0.45 μm filters before analyses to determine the concentrations of As(III) and As(V). Arsenic species of filtered samples were separated by passing through an HPLC (Hitachi 7110, Naka, Japan) and an anion column (Machey-Nagel, Nucleosil, 10 μm , 250 \times 4.6 mm), which linked to an electro-thermal atomic absorption spectrometer (AAS, Perkin-Elmer AA100) and a hydride generation (HG) system (Perkin-Elmer FIAS-400) [19]. Samples were spiked with As species to determine the recovery rate in the laboratory procedure, which yielded recovery rates of As(III) and As(V) of 100.7 ± 3.8 and $97.2 \pm 4.0\%$, respectively. The coefficient of variation was used to test the reliability and was less than 5% in all experiments. Fe(II) concentrations were measured colorimetrically using the ferrozine method [20] by adding 200 μL sample to 1.5 mL of the ferrozine solution with the measuring absorbance of 562 nm.

III. RESULTS

A. HFO and As(V) Reduction Experiments

For fresh HFO slurry, iron-reducing bacteria drives the rapid reduction of Fe(III) under anaerobic condition (Fig. 1). The Fe(II) concentrations in FeRB and citrate involved experiment rapidly raised within 5 days whereas that in acetate involved experiment slightly increased and equilibrated after 9 days. The final concentrations of Fe(II) in

the citrate- and acetate- amended bottles are 204 and 68 mg/L, respectively. Some black grains precipitated in the bottles that were inoculated with FeRB, at the onset of Fe(III) reduction. The reductive dissolution of Fe(III) released Fe(II) to solution, and then induced the transformation of the less thermodynamically favorable phase, HFO, to more stable minerals, such as goethite and magnetite, with smaller available surface areas [21]. The rate of Fe(III) reduction with citrate-added exceeded that with acetate-added, indicating that the reductive dissolution rate of Fe(III) is governed by Fe mineralogy, the accumulation of Fe(II) reaction products, and the type of organic substrate [22].

For aqueous As(V) reduction experiment, the reduction was driven by FeRB and completed within 20 days, and the reduction rates were similar for two considered organic substrates (Fig. 2(a) and (c)). The concentrations of As(V) and As(III) in abiotic experiments maintained the constants (25 and 0 mg/L, respectively) with time. According to these results, the microbial-mediated reduction of As(V) is promoted by the As(V)-reducing bacterial, growing simultaneously at the onset of the enrichment experiments. Incorporation of As(V)-reducing bacteria, which can reduce As and Fe in batch column experiments, have demonstrated that the As mobilization processes are governed by these organisms [23]. Two known microbial pathways for reducing As(V) are the respiratory pathway (arrA pathway), which couples the oxidation of the organic substrate for cell growth, and the detoxification pathway (arsC pathway), where cell converts As(V) to As(III) and then actively transported out from the cell [24].

B. As-contained HFO Reduction Experiment

For As-contained HFO slurry, the reduction rates and equilibrated Fe(II) concentrations (Fig. 3) are similar to those in the fresh HFO slurry reduction experiments (Fig. 1). The Fe(II) concentration in FeRB and citrate involved experiment raised within 2 days and maintained a constant of ~205 mg/L after 15 days (Fig. 3(a)), while that in FeRB and acetate involved experiment slightly increased and equilibrated with ~45 mg/L after 10 days (Fig. 3(b)). Also, goethite- and magnetite-like precipitated in these two bottles at the onset of Fe(III) reduction. The As(V) concentrations in FeRB and citrate involved experiment rapidly raised within 1 day and then the As(III) concentration slightly increased (Fig. 3(a)). The As(V) concentrations in FeRB and acetate involved experiment mildly increased within 10 days and then the As(III) concentration increased on 12 days (Fig. 3(b)). The final concentrations of As(V) in the citrate- and acetate-amended bottles are ~3 and ~2 mg/L, respectively. Intuitively, all As should be released during the reductive dissolution of Fe(III) within the decrease of arsenic-contained HFO surface area. However, the equilibrated concentrations of total As are only about 2-8 mg/L in these four bottles, and are all lower than the expected final concentration (25 mg/L). These results show that the FeRB targeted primarily HFO in the presence of As(V), producing the same levels of Fe(II) with that in the absence of As(V). Although the surface area of amorphous Fe minerals was

decrease by HFO reduction, arsenic may be sequentially

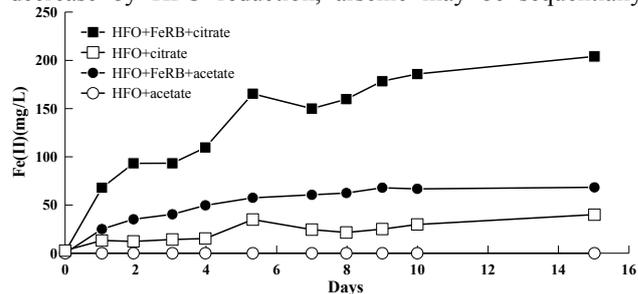


Figure 1. Variations of Fe(II) concentrations in fresh HFO reducing experiments, including biotic (FeRB inoculation) and abiotic (controlled) subsets with 0.5 mM citrate or acetate.

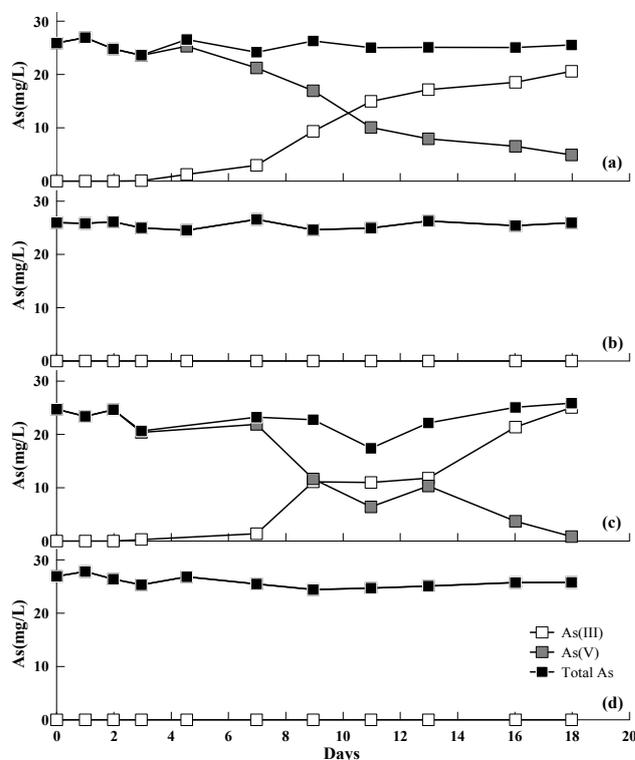


Figure 2. Concentrations of As(III), As(V), and total As in aqueous As(V) reducing experiments, including (a) biotic (FeRB inoculation), (b) abiotic (controlled) subsets with 0.5 mM citrate and (c)(d) those with 0.5 mM acetate. Total As denotes the sum of As(III) and As(V).

retained on surface of secondary minerals e.g. the co-precipitation of Fe(II)-As after the reduction of HFO [21][25]. Additionally, the patterns of As(V) concentration variations of abiotic experiments (Fig. 3(b) and (d)) are similar with those of biotic experiments (Fig. 3(a) and (c)). The As(V) concentrations in citrate and acetate involved experiment rapidly raised within 1 and 9 days and subsequently equilibrated at 6.02 and 1.86 mg/L, respectively. Though the equilibrated As concentrations are

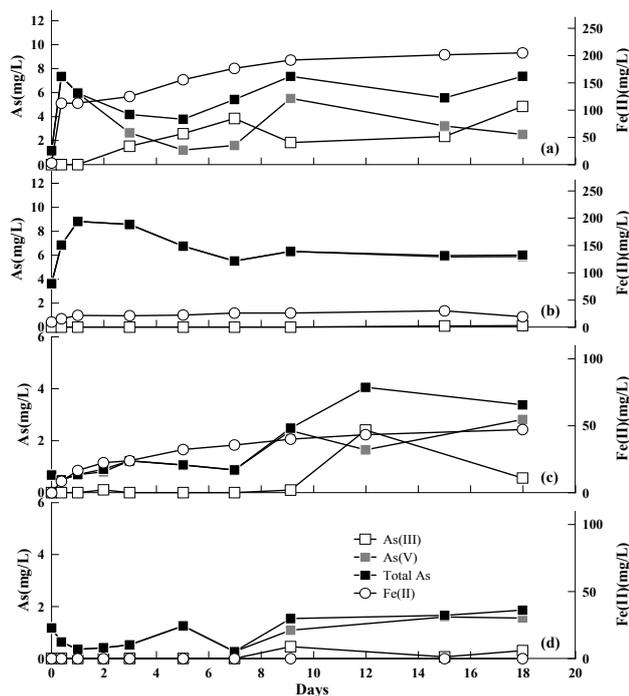


Figure 3. Concentrations of As(III), As(V), total As, and Fe(II) in aqueous As(V)-containing HFO reducing experiments, including (a) biotic (FeRB inoculation), (b) abiotic (controlled) subsets with 0.5 mM citrate and (c)(d) those with 0.5 mM acetate. Total As denotes the sum of As(III) and As(V).

limited, the mild difference of released As concentrations between the FeRB-inoculated and abiotic subsets shows that the desorption from the solid surface of HFO is also the possible process of As mobilization.

IV. DISCUSSION

A. Effect of Different Organic Carbon on As Mobility

The organic substrates used herein may compete for the sorption sites with As(V), resulting in the desorption of As(V), especially for citrate. Competition of dissolved organic matter with As for sorption sites onto Fe hydroxides [26] cause the mobilization of As from the surface of Fe hydroxides [27] and is more prevalent in this study [23]. According to previous investigation, the correlation between the total organic carbon and As concentrations in the groundwater of southwestern Taiwan was significantly positive [15][28]. Moreover, the ratio of As desorption to the total released As added with citrate (82%, as calculated from the equilibrium As concentrations of FeRB-inoculated and abiotic subsets) is higher than that with acetate (55%), indicating that the desorption of As by the competition of large molecular organic carbon is apparent. The degraded organic carbon then acts as the electron donor for HFO reductive dissolution and As(V) reduction. Reduced As(III) by As-reducing bacteria is consistently more mobile than As(V) in solid phases [29]. Since that the FeRB used in this study is mixed strains and may contain both Fe- and As-reducing bacteria species, the interactions involved different

organic carbon interprets the contribution of Fe-reducing bacteria on the reductive dissolution of ferrihydrite and that of As-reducing bacteria on As mobility in groundwater [30].

B. Roles of Reducing Bacteria in As Speciation

Arsenite is produced by the partial reduction of aqueous As(V) after one day and seven days in the citrate- and acetate-amended bottles, respectively (Fig. 3(a) and (c)). The reduction of aqueous As(V) by reducing bacteria occurred after the desorption of As(V) and then the reduction of HFO. Arsenate acts as the electron acceptor after the equilibrium of aqueous Fe(II)/HFO. Three possible As mobilization mechanisms in anoxic groundwater, as mediated by metal-respiring bacteria, have been proposed [31]. First, Fe(III) was biochemically reduced to the soluble Fe(II) state by FeRB, causing the release of adsorbed As(V) into the aqueous phase. Second, adsorbed As is released from the surface of Fe(III) minerals by the reduction of As(V) to As(III), as mediated by dissimilarity arsenate-reducing prokaryotes (DARPs). Third, Fe(II) and As(III) are reductively released by iron-reducing DARPs. In this study, the mix-cultured FeRB may contain As-reducing bacteria and yields different contributions between Fe- and As-reducing in the experiment. The link between the Fe(III) reduction, driven by the FeRB, and the reduction of desorbed As(V) via DARPs, may be the possible mechanism of As mobilization and transformation. Notably, the reduction patterns of As(V) in the presence of various organic substrates are similar (Fig. 2(a) and (c)), while a significant difference in time lag exhibits in the formation of As(III) (Fig. 3(a) and (c)).

Although the FeRB-mediated reductive dissolution of Fe(III) in Fe (hydr)oxide minerals has been identified as the most plausible mechanism of As mobilization [32], the subsequent transformation of As species mentioned herein differs from those proposed elsewhere [31]. Biogeochemical processes for the release of As to groundwater are either direct enzymatic microbial reduction of As(V) by Fe(III)-reducing bacteria or indirect processes of which are associated with the reduction of As-rich Fe (hydr)oxides [12]. The most possible process is that the reduction of As(V) took place after Fe(III) reduction and As(V) desorption because some reducing bacteria species in cultured FeRB can reduce aqueous As(V) following the consumption of targeted Fe(III) mineral. The results of this study provide a foundation for future biogeochemical investigations. Researches of cultured FeRB are presently conducted by strains isolation and PCR reaction, and denaturing gradient gel electrophoresis (DGGE) analysis to assess the contribution of Fe- and As-reducing bacteria to the mobilization and transformation of As in groundwater.

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