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Assessing the mechanisms controlling the mobilization of arsenic in the arsenic contaminated shallow alluvial aquifer in the blackfoot disease endemic area

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ABSTRACT

High levels of arsenic in groundwater and drinking water represent a major health problem worldwide. Drinking arsenic-contaminated groundwater is a likely cause of blackfoot disease (BFD) in Taiwan, but mechanisms controlling the mobilization of arsenic present at elevated concentrations within aquifers remain understudied. Microcosm experiments using sediments from arsenic contaminated shallow alluvial aquifers in the blackfoot disease endemic area showed simultaneous microbial reduction of Fe(III) and As(V). Significant soluble Fe(II) (0.23 ± 0.03 mM) in pore waters and mobilization of As(III) (206.7 ± 21.2 nM) occurred during the first week. Aqueous Fe(II) and As(III) respectively reached concentrations of 0.27 ± 0.01 mM and 571.4 ± 63.3 nM after 8 weeks. We also showed that the addition of acetate caused a further increase in aqueous Fe(II) but the dissolved arsenic did not increase. We further isolated an As(V)-reducing bacterium native to aquifer sediments which showed that the direct enzymatic reduction of As(V) to the potentially more-soluble As(III) in pore water is possible in this aquifer. Our results provide evidence that microorganisms can mediate the release of sedimentary arsenic to groundwater in this region and the capacity for arsenic release was not limited by the availability of electron donors in the sediments.

1. Introduction

Arsenic (As), a known human carcinogen, is widely distributed in food, water, soils, and air [1,2]. It is a ubiquitous element of both natural and anthropogenic origin and is often responsible for contaminating water supplies [3]. In most cases, arsenic contamination of groundwater is derived naturally from arsenic-rich aquifer sediments [4]. Natural arsenic in groundwater used as a source for drinking, household, and agricultural purposes represents a major health problem for humans in many places around the world, and particularly in South Asia (West Bengal, Bangladesh) where tens of millions of people in the Bengal Delta are at risk from drinking arsenic-contaminated water [4–6]. Numerous other locations worldwide have been reported, and some of these, such as those in Taiwan, have been recognized for several decades [4].

In Taiwan, arsenic-contaminated aquifers typically exist in the coastal plain sediments where groundwater is reducing [7–10]. The southwestern coast area is historically associated with endemic cases of blackfoot disease (BFD), a peripheral vascular disease caused by long-term ingestion of arsenic-contaminated groundwater [11]. In addition to skin pigmentation, a very high incidence of keratosis, and skin, lung, bladder and other cancers were found among people who lived in southwestern coast area and drank arsenic-contaminated well water prior to the 1990s [11,12]. At present, although well water is not directly ingested by most residents in this region, it is still extensively provided to meet domestic, irrigation, aquacultural, and industrial needs. High bioaccumulated arsenic concentrations were found in farmed fish that were associated with the arsenic found in pond water in this area [12]. However, there is still intense debate on the etiology of BFD as well as the sources of arsenic. Despite considerable research over the past 30 years into the occurrence and distribution of arsenic in the region's groundwater, few studies have been conducted to investigate the geochemical and microbiological processes that influence the element's speciation and mobility in the aquifers of the BFD endemic area. To this end, it is important to understand all relevant abiotic and biotic factors that contribute to arsenic mobilization and sequestration in such environments.

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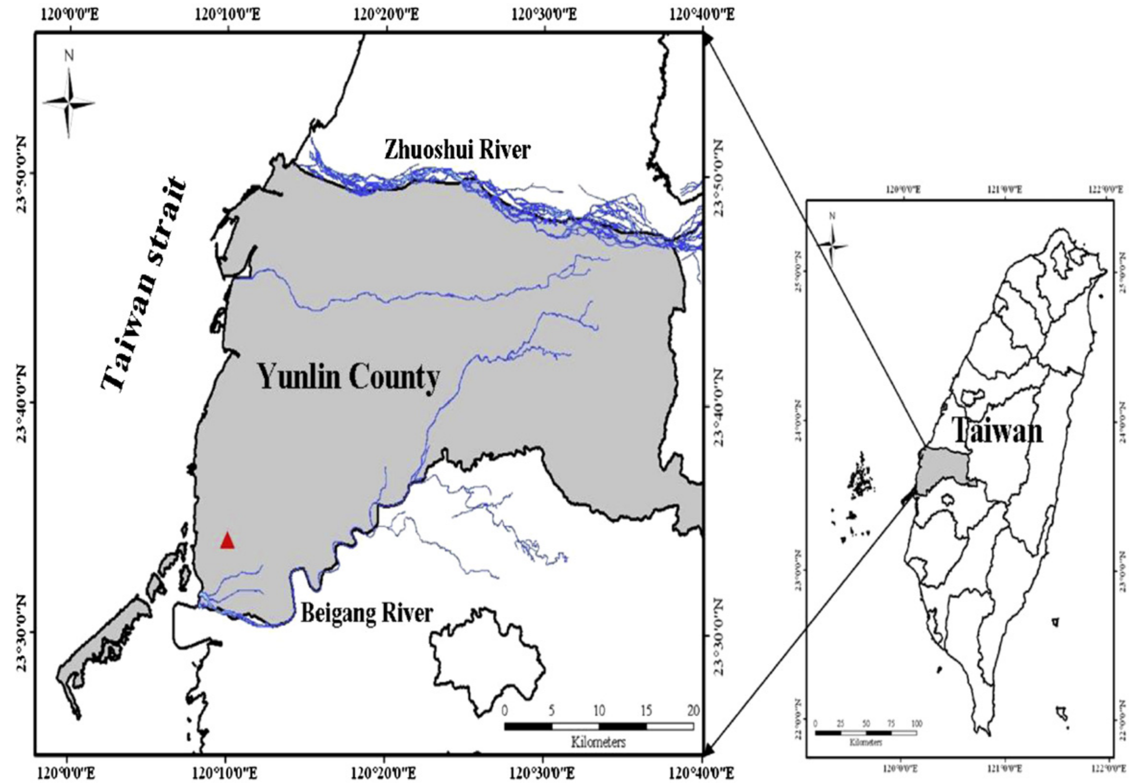

Fig. 1. Site map of the study area and approximate sediments sampling site (▲).

The processes controlling the release of arsenic into groundwater systems are complex and have been extensively studied over the past decade, but still remain a subject of intense debate. Various mobilization mechanisms have been proposed, including oxidation of arsenic-rich pyrite [13], reductive dissolution of iron oxyhydroxide phases [14,15], competition of solutes for sorption sites on iron oxides [16], mineral weathering [17,18], and microbial reduction of sorbed As(V) to the potentially more-mobile As(III) under anoxic conditions [19,20]. Among the proposed mechanisms, reductive dissolution of arsenic-bearing ferric oxyhydroxides has gained particular attention, with several studies showing that indigenous anaerobic metal-reducing bacteria can play a significant role in controlling arsenic mobilization [20–22]. However, the precise role of these anaerobic bacteria in arsenic mobilization remains largely uncharacterized [23]. Most studies that described metal-reducing bacteria in mediating arsenic mobilization have been based on molecular approaches with the sequences of 16S rRNA gene clone libraries [21,24–26]. Using a culture-dependent technique to identify and isolate the indigenous bacteria which is native to the aquifer sediment that might play a role in mediating the release of arsenic into groundwater systems has seldom been described.

In this study, we examine the role that indigenous microbial communities play in arsenic mobilization in an arsenic-contaminated aquifer in the BFD endemic area. Herein, we present data on the concentration and speciation of arsenic and iron associated with sediments from a well drilled in the BFD endemic area. In order to test whether arsenic-contaminated groundwater was attributable to microbiological processes in this aquifer, we conducted sediment incubation experiments under a range of biogeochemical conditions. A microcosm-based study was further carried out to elucidate the role of indigenous anaerobic bacteria in the biogeochemical cycling of arsenic and iron in aquifer sediments.

2. Materials and methods

2.1. Site description and sample collection

The sampling site was located in the southern Zhuoshui River alluvial fan of Taiwan (23°34′07″N, 120°10′04″E) (Fig. 1), which is close to southwestern coast in the blackfoot disease endemic area. The southern Zhuoshui River alluvial fan has an area of around 1000 km^2. It is bounded by two major rivers that flow through the area, the Zhuoshui River to the north and the Beigang River to the south, partitioned into the proximal-fan, the mid-fan, and the distal-fan areas [27].

A borehole was drilled about 10 m away from a well that has been routinely monitored for arsenic contamination by the Taiwan Industrial Development Bureau of the Ministry of Economic Affairs. The well had elevated levels of arsenic ranging from 0.30 to 0.78 mg/L [28]. To collect sediment samples, briefly, a borehole was drilled without using drilling mud. A drill rig and a split-tube sampler with PVC liner (50 mm outside diameter) which had been pre-cleaned were used to collect sediment cores. Cores were extruded and segmented by depth in an anoxic, sterilized glove box containing N_2 gas. Groundwater collected from wells nearby the drilled borehole with similar depth has been found to be reducing (−144 mV to −178 mV), indicating that the sediments were under reducing conditions. Therefore, after sampling, sediment samples were stored in sterile airtight polyethylene bags in an anaerobic tank and preserved at 4 °C in the dark to minimize microbial activity. Further sediment manipulations were performed only under strict anoxic conditions.

2.2. Microcosm sediment cultures

About 15 g of sediments (from 20 m in depth, dark gray fine sandy loam) was mixed with 30 mL of artificial