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Research Article

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synthetic minerals

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box containing N_2 gas. Core samples were dark in color, and consisted of silty sand, mud, and clay. e C-14 dating data show that the sequence formation is in the Holocene age [22]. Only the sediments retrieved from depths of 19.5-20.0 m were used for enrichment. Solid As ranged from 4.02 to 12.77 mg kg^{-1} with a mean of 7.21 mg kg^{-1}, whereas the Fe contents varied from 2.5 to 4.6%, with a mean of 3.5%. e amounts of organic and inorganic carbon in the collected core sediments were comparable and generally less than 0.5% [4]. Groundwater and core sediment samples were stored at 4°C in polyethylene bags to minimize microbial activity. Further groundwater and sediment manipulation were performed only under strict anoxic condition.

Bacterial strains genus identi cation

Before conducting the microcosm experiments, identi cation of microbial population in the groundwater (YL7) and core sediment (YL7-1 at depth of 20 m) were rstly conducted. To enrich bacterial populations, groundwater sample of YL7 was inoculated into a chemically de ned medium (CDM) (8.12 mM $MgSO_4$, 18.7 mM NH_4Cl, 7 mM Na_2SO_4, 0.0574 mM K_2HPO_4, 0.457 mM $CaCl_2$, 44.6 mM Na-lactate, 0.012 mM $FeSO_4$, and 9.5 mM $NaHCO_3$ with the pH adjusted to 7.2) [26] amended with or without 2 mM of As(III) or 10 mM of As(V) under aerobic and anaerobic incubation, respectively. Samples were then incubated at 25°C for 24 h. A pure culture bacterial genomic DNA was prepa6(h b)-5(n <</MCID 39-5(l)-3(a)19(t)-5(io)121683 482.03yS(i)-3(t)0.598.12 mM 93b)12y Wa7]riAnt1(ur)13(e)0.6(c)-7(u)-5(l)1

of groundwater (YL7), unamended core sediment (YL7-1) and core sediment (YL7-1) amended with acetate are reported in Table 2. Seven genera including *Pseudomonas* sp., *Psychrobacter* sp., *Vib*ren4368 Tw 9 MC /Span <</MCID 903 >>BDC 4T /T1_1 1 Tf 9 0 0 9 1 8

report [35]. Liao et al. [29] conducted the microcosm experiment using the indigenous sediments amended with or without acetate. According to the results, the congruent reduction of Fe and As led to the major release of As. Further, addition of organic carbon limited the As mobilization. Note that the combined results of this study and Liao et al. [29] indicated the distinct microbial activities occurred between groundwater and sediment, which were also proved by the analysis of bacterial microorganisms (Table 2). Adding acetate altered the terminal electron acceptors as observed herein indicating the shi of microbial community as reported by Islam et al. [3].

In assays of RB-W mixed with sterile groundwater, the trends of As and Fe species were comparable to that inoculated with the same population but amending with arti cial groundwater (Figures 2b,2e and 3c). e similar levels of dissolved As in the abiotic sets, with and without acetate, revealed that competitive desorption by organic carbon, if it occurred, was insigni cant under the experimental condition (Figures 3d,3e).

Overall, only biotic sets with and without addition of acetate showed noticeable increase in dissolved As and Fe; this nding supports the role of microorganisms in mediating the cycle of As and Fe under reducing conditions [3,11,39] (Figures 2 and 3). e discrepancy between the RB-W and RB-S experimental results is due to the contrasting bacterial communities cultured separately from groundwater and core sediment (Table 2) [28-29]. Di erent microorganism can utilize sedimentary As via various reaction pathways and in uence the groundwater As concentration. In this study, we observed that RB-W population directly mediated the coupled reducing reaction of As and Fe in the sediment leading to a systematic increase of As (III) and Fe (II) concentrations. On the other hand, RB-S population exhibited strong reducing ability toward Fe minerals, resulting in dramatic increase of Fe(II) and formation of secondary As-bearing iron oxyhydroxides. It is noteworthy that the decouple reduction of As and Fe via the chelating solubilization of Fe minerals observed in this study provided another potential mechanism for the mobility of sedimentary As.

By combining the data of microcosm experiments with RB enrichment cultures, the discrepancy results of As and Fe between the experimental sets were attributed to the contrasting bacterial community between groundwater and sediment. (Figure 2 and Table 2). Reducing bacteria from groundwater prefer to release both Fe(II) and As(III) to aqueous phase whereas reducing bacteria from sediment prefer to only reduce Fe(II) and sequestrate As to solid phase. Further, the dominant dissolved Fe as Fe(II) or Fe(III) in this study revealed various reactive pathways toward Fe minerals [17,19,40-42]. Nonetheless, either through chelating solubilization or reducing dissolution of Fe minerals was regarded as one kind of reductive pathways [17].

Microbial e ects on the As and Fe cycling

ree possible arsenic mobilization mechanisms mediated by metal-respiring bacteria have been proposed [39]. ese microbial reactions are driven by degraded organic carbon using As(V), Fe(III), or both as terminal electron acceptors. For the RB-W inoculated treatments, microbial reduction of sedimentary Fe and As seemed

Figure 4: Normalized As XANES spectra of the sediments after the experiments ZLWK ,5% SRSXODWLRQV 7KH QXPEHU UHSUHVHQWV WKH GHSWK RI VHGLPHQW XVHG WKH capital letters W and S represent the inoculation of RB-W and RB-S, and C denotes the controls.

amount of dissolved Fe(II) measured in sets with RB-S was double that of RB-W treatments, only minor levels of As(III) were detected regardless of the sediments type (Figure 2). Based on these results, the metabolic pathway of sediment microbial community showed strong a nity for Fe, and the potential formation of secondary Fe minerals seemed to sequestrate As [43]. us, we concluded that the exertion of bacterial communities of these two RB enrichments expresses di erent metabolic pathways toward terminal electron acceptors on the incubation experiments. In short, the reducing bacteria cultured from groundwater prefer to mediate and release of both As (III) and Fe (II) whereas the reducing bacteria from sediment prefer to only reduce Fe (III) to F (II) and form As-bearing iron oxyhydroxides minerals.

For the sets suspended with in-situ groundwater, an unusual release mechanism of As that converted sedimentary arsenic and iron into solubilized As(III) and Fe(III) were observed (Figures 3a and 3b). Most studies focused on the reducing dissolution of As-bearing Fe oxyhydroxides in As-contaminated area but overlooked the other possible release pathway such as the chelating solubilization of Fe minerals [44]. Moreover, the mobility of As and Fe was decoupled processes, especially with the aid of reactive carbon source. Decoupling release of As and Fe in aqueous phase generally re ected adaption of the respiratory pathways in the sediments, through dynamic changes in species which were metabolically active in the microbial community, or through altered expression of the relev8/T1_-h(i)-3(a)-rb8b4(e rTm [7(h)2k)7 151e punity,

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