



Speciation change and redistribution of arsenic in soil under anaerobic microbial activities



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HIGHLIGHTS

- The soil As was released, immobilized, redissolved and resequenced by anaerobes.
- Reduction of iron and arsenate was responsible for arsenic release.
- Sulfide controlled the arsenic immobilization, redissolution and resequencing.

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ABSTRACT

Arsenic speciation and behavior in soil are strongly affected by redox conditions. This work investigated speciation transformation and redistribution of arsenic in soil under anaerobic conditions. The effect of microbial sulfidogenesis on these processes was examined by addition of sulfate to the incubation systems. As(III) was found to be the dominant arsenic species in solution during the process of anaerobic incubation. The change of dissolved As concentration with incubation time showed “M” shaped profiles, e.g. the curves displaying two peaks at approximately 24 h and 240 h for the system with added sulfate. Arsenic was released and reduced to As(III) in the early stage of the incubation, and then resequenced into the solid phase. After excess sulfide was generated, the resequenced arsenic was released again (probably due to the dissolution of arsenic sulfide by dissolved sulfide ions) via the formation of thioarsenite. At the end of the incubation process, most of the dissolved arsenic was removed again from solution. The findings may have important implications to the fate of arsenic in flooded sulfur-rich soils.

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1. Introduction

Arsenic is a toxic metalloid and widely exists in soil, groundwater and surface water throughout the world [35]. Due to its adverse impact on human health, there is increasing concern over arsenic pollution in soil, especially in paddy soil where elevated levels of arsenic in rice grains were observed [45,36,31]. The behavior and bioavailability of arsenic in soil are strongly influenced by its speciation. Improved understanding of As speciation and behaviors in soil is required for predicting its fate and for developing effective technologies for the remediation of arsenic pollution [23,28,27,17,51].

The common oxidation state of As in soil is pentavalent and trivalent, i.e. As(V) and As(III). In oxidizing environment, arsenic is usually associated with various oxide or clay minerals in the

form of adsorbed and co-precipitated arsenate or occurs as arsenate minerals (e.g. ferric arsenate) [16,4], whereas in reducing environment arsenic is present in the form of adsorbed/coprecipitated arsenite and/or arsenic sulfide etc [10,5]. Redox condition is one of the most important factors controlling arsenic mobility in the environment [51]. When redox condition changes, arsenic may be transformed from As(V) to As(III) or the other way round, leading to its mobilization or sequestration. In subsurface environment, the redox condition is generally controlled by anaerobic microorganisms, which can reduce arsenic and/or iron by using organic carbons as electron donors. As a result, arsenic may be mobilized and released from arsenic-bearing minerals into aqueous phase due to reductive dissolution of iron oxides and/or weaker retention of arsenite compared to arsenate by various minerals [1,23].

When the environment becomes more reducing, the influence of microbial sulfidogenesis dominates the fate of arsenic in several ways [38,39,41,44,6,7]. Sulfide ions are involved in Fe(III) reduction and consequent precipitation of amorphous FeS, leading to

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enhanced arsenic mobilization because FeS adsorbs much less arsenic than ferric oxyhydroxides [30,50]. However, transformation of FeS to pyrite can remove arsenic from aqueous phase by incorporation into the pyrite structure [30]. The presence of sulfide ions in solution may also cause arsenic mobilization by the formation of thioarsenic species and/or replacement of adsorbed arsenic [6]. Besides, arsenic may also be immobilized as arsenic sulfide when aqueous arsenite and sulfide ions accumulate to the level exceeding arsenic sulfide solubility [34,25].

The behavior of arsenic in soil and sediment is very complicated due to the complex nature of soil and variation of redox conditions. Anoxic-sulfidic environment will be developed in flooded soil (e.g. the submerged paddy field soil) because of anaerobic microbial activities. Detailed processes of arsenic speciation transformation and the role of sulfate in arsenic redistribution in soil under the impact of anaerobic microorganism have not been fully understood by far. In a previous study on the behavior of arsenic in the polluted offshore sediment ($\sim 735 \text{ mg kg}^{-1}$ As) under anaerobic microbial activities, it was found that arsenic was released in the early stage of the incubation due to reductive solubilization of arsenic and fol-

lowed by resequstration into the solid phase mainly as amorphous As_2S_3 during the later stage of the process [50]. However, arsenic sulfide can further be solubilized in the presence excess aqueous sulfide [41].

The objectives of this work were to study anaerobic microbial mediated redistribution and the fate of arsenic in soil and to discuss the underlying mechanism in the process.

2. Materials and methods

2.1. Soil collection and treatment

The surface soil of 0–20 cm depth was collected from the paddy field in Zhangshi District, Shenyang City, Northeast China. The concentration of arsenic, iron and sulfate in the soil sample was $66.7 \mu\text{mol kg}^{-1}$ (5 mg kg^{-1}), $537.8 \text{ mmol kg}^{-1}$ and $67.9 \text{ mmol kg}^{-1}$, respectively. The soil was air dried prior to loading arsenic. Arsenic solution (6.6 mg L^{-1}) was prepared by dissolving sodium arsenate in de-ionized (DI) water and the pH was pre-adjusted to 7.0 using dilute HCl. The soil and arsenic solution was mixed at liq-

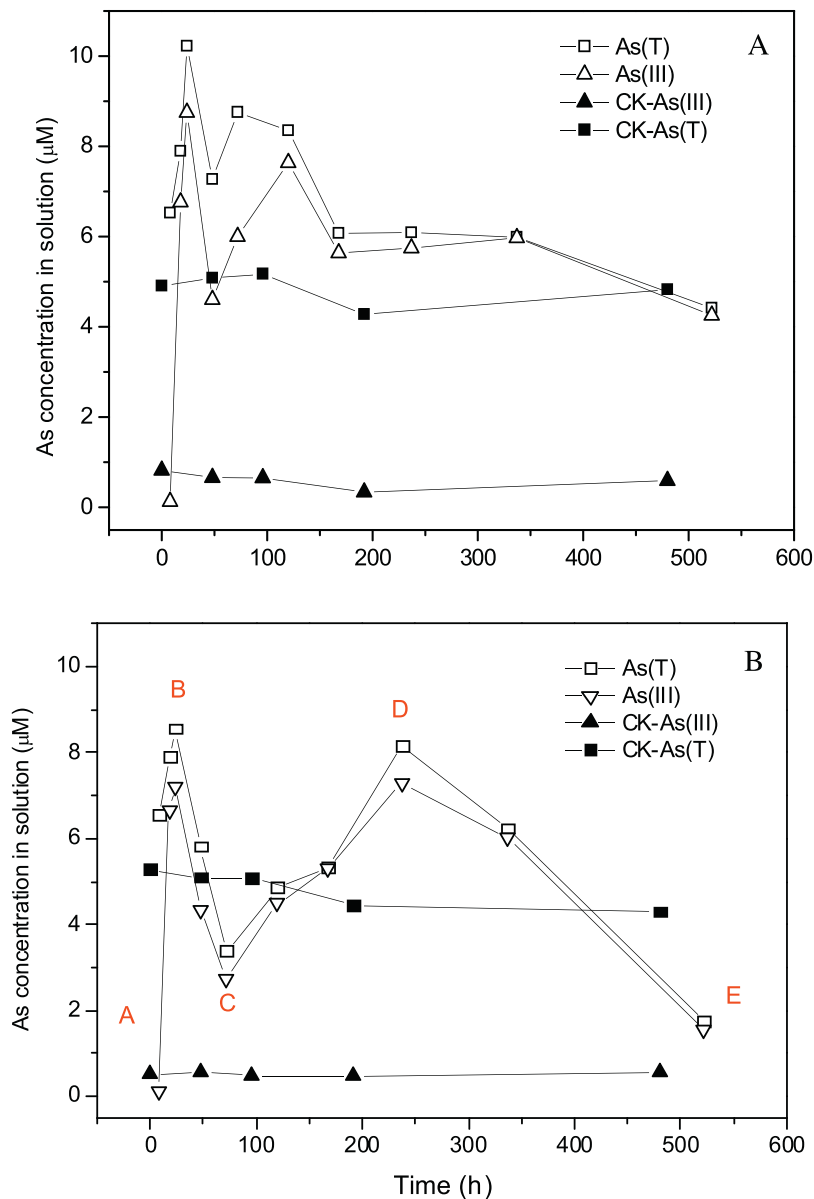


Fig. 1. Change of dissolved As concentration with incubation time; A: without added sulfate; B: with added sulfate. (CK: abiotic control; As(T): total arsenic; As(III): arsenite).

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