The influence of irrigation-induced water table fluctuation on iron redistribution and arsenic fate in unsaturated zone

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ABSTRACT: Using high arsenic (As) groundwater as the main source of irrigation water is a common practice in vast rural regions due to the water resource shortage of surface water. This study aims to describes the As and Fe redistribution during irrigation and consequent water table fluctuation through column experiment. Results clearly demonstrate the oxygen exchange between atmosphere and column pore during both water table decrease and recovery. The exchanged oxygen has resulted in the As and Fe redistribution. Moreover, the fixed As and Fe were accumulated mostly on the top of the column during percolation. However, topsoil is precisely the site where nutrients come from for crops. As a result, As uptake by crops from soil may continually occur. Thus, it is of great significance for food security to reduce the agricultural use of high-As groundwater.

1 INTRODUCTION

Recent studies have shown that consumption of As-contaminated foods became an alternative pathway for As exposed to human besides water drinking (Francisca *et al.*, 2015). Actually, As compounds in crops come mainly from soil and irrigation water. The As content in soil can reach up to $83 \mu\text{g} \cdot \text{g}^{-1}$ in some irrigated areas, which far exceeds the background level or safety level of agricultural soil (Williams *et al.*, 2006). This poses potentially considerable risk to public health. Given above reasons, it is imperative to comprehend the As and Fe redistribution behavior during irrigation. In order to achieve the goal, a column experiment was carried out in this study. Furthermore, water table fluctuation was set to the main artificially manageable process since it has been confirmed to be a most important factor influenced the redox reactions (Tong *et al.*, 2016).

2 METHODS/EXPERIMENTAL

The column experimental setup and procedure was shown in Figure 1. In this experiment, groundwater and soil was replaced by deoxidized As(III)-Fe(II) solution and acid washed quartz sand, respectively. External environment of this experiment is mainly maintained in anaerobic condition. The ventilation valve was open to atmosphere and the height of the splitter controlled the water table inside the column. The liquid samples were collected from T-value, and the flow cell was used for pH, ORP and DO measurements. The artificially manageable process is water table fluctuations, while the vertical infiltration recharge is uninterrupted. The sand inside the column was then sampled every 6 cm. The sampled sand was dried up under anaerobic atmosphere and then used for As and Fe sequential extraction.

Figure 1. (a) The column experiment setup. (1) Peristaltic pump, (2) Syringe-driven filter, (3) Water distributor, (4) Ventilation valve, (5) T valve, (6) Detector probe, (7) Small flow cell, (8) 3-way splitter, (9) Waste. (b) The experimental procedure.

3 RESULTS AND DISCUSSION

3.1 Water table fluctuation and oxygen transfer

As shown in Figure 2a, during the equilibrium phase (0–65 h), oxygen inside the column was washed away, and thus effluent dissolved oxygen content reached the lowest level at 65 h. Then, effluent DO rose rapidly when valve was opened and water table lowered down to 30 cm. This indicates that the decrease of water table can result in the infiltration of air into column pore. Effluent DO then decreased due to consumption of dissolved oxygen. Notably, the experimental results showed that oxygen exchange between atmosphere and column pore also happened during the period of water table recovery. The effluent DO showed a sharp rise when water table ascended, and the DO remained at about $6.7 \text{ mg} \cdot \text{L}^{-1}$ for a long time after recovery. This demonstrates that atmospheric oxygen was trapped by solution before the injected solution flowed into sand pores.

Figure 2. Effluent monitoring results.

3.2 The As(III) and Fe(II) oxidation

As shown in Figure 2c, it is a synchronous variation in DO and Fe(II) contents at the beginning of LWP, which illustrates that DO initiates Fe(II) oxidation. The effluent pH and Fe(II) content showed the same varying trend (Fig. 2b), which indicates the Fe(III) hydrolysis controlled the aqueous-phase pH in this experiment. However, the Fe(II) and DO content varying trends were not completely the opposite. Especially at the latter half of LWP, the effluent Fe(II) content and DO, solution pH consistently decreased. This may result from the self-catalyzed characteristic of Fe(II) oxidation in the presence of Fe(III) mineral.

Moreover, As(III) was observed to be partially oxidized during percolation (Fig. 2d). However, it has been confirmed that As(III) oxidation is quite slow in the absence of strong oxidant. The two processes that can accelerate As(III) oxidation under natural conditions are microbial mediation and hydroxyl radical-evolving function. Jia *et al.* has confirmed the production of hydroxyl radical when Fe(II) was oxidized by O₂ (Jia *et al.*, 2017). Thus, the mechanism promoting the As(III) oxidation was considered as the generation of hydroxyl radical when the Fe(II) was oxidized to Fe(III) during this column experiment.

3.3 As and Fe redistribution

The As and Fe extraction results are shown in Figure 3. It is obvious that both As and Fe contents were higher in the sands of top 9 cm, but decreased dramatically with depth increased. The As extraction results showed that the As in each sample mainly existed as the form of adsorbed As (especially strongly adsorbed As), accounting for 58.0%–88.0% with an average of 76.8% of total solid-phase As. Furthermore, the proportion of weakly adsorbed As was gradually increased with depth, while strongly adsorbed and non-adsorbed As decreased. This was likely due to the transformation from As-Fe surface complexation into As-Fe surface precipitation during the growth of Fe(III) mineral at the top of column.

3.4 Environmental implications

The As/Fe molar ratios ranged from 0.025 to 0.048 with an average of 0.035, which far exceeded the background level of soils. These fixed As were more likely to be released again due to the transformation

Figure 3. The results of As, Fe extraction.

of Fe(III) mineral or microbial actions after irrigation. This release could be very slow and last for a long time. Coincidentally, topsoil is precisely where nutrients come from for crops. Thus, irrigation and consequent water table fluctuation may pose a long-term potential risk to public health via dietary approach.

4 CONCLUSIONS

The column experiment results show that irrigation and consequent water table fluctuations (both decrease and increase) can result in the oxygen exchange between atmosphere and column pore. The exchanged oxygen was then initiated the Fe(II) and As(III) oxidation. As a results, As and Fe was accumulated on the sand column, especially top of column. These processes are highly likely to cause a long-term potential risk to public health via dietary approach. Thus, it is of great significance to reduce the agricultural use of high-As groundwater.

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