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Microbial community composition of a household sand filter used for arsenic, iron, and manganese removal from groundwater in Vietnam

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HIGHLIGHTS

- We studied the microbial community composition of a household sand filter for arsenic removal from groundwater.
- Mn-oxidizing bacteria were enriched in a distinct filter layer.
- The microbial community on the filter was dominated by nitrifying microorganisms.
- Abiotic iron oxidation prevailed over biotic Fe(II) oxidation.
- The formation of Mn oxides contributed to abiotic As oxidation and immobilization by sorption to Fe oxides.

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ABSTRACT

Household sand filters are used in rural areas of Vietnam to remove As, Fe, and Mn from groundwater for drinking water purposes. Currently, it is unknown what role microbial processes play in mineral oxide formation and As removal during water filtration. We performed most probable number counts to quantify the abundance of physiological groups of microorganisms capable of catalyzing Fe- and Mn-redox transformation processes in a household sand filter. We found up to 10^4 cells g^{-1} dry sand of nitrate-reducing Fe(II)-oxidizing bacteria and Fe(III)-reducing bacteria, and no microaerophilic Fe(II)-oxidizing bacteria, but up to 10^6 cells g^{-1} dry sand Mn-oxidizing bacteria. 16S rRNA gene amplicon sequencing confirmed MPN counts insofar as only low abundances of known taxa capable of performing Fe- and Mn-redox transformations were detected. Instead the microbial community on the sand filter was dominated by nitrifying microorganisms, e.g. *Nitrospira*, *Nitrosomonadales*, and an archaeal OTU affiliated to *Candidatus* Nitrososphaera. Quantitative PCR for *Nitrospira* and ammonia monooxygenase genes agreed with DNA sequencing results underlining the numerical importance of nitrifiers in the sand filter. Based on our analysis of the microbial community composition and previous studies on the solid phase chemistry of sand filters we conclude that abiotic Fe(II) oxidation processes prevail over biotic Fe(II) oxidation on the filter. Yet, Mn-oxidizing bacteria play an important role for Mn(II) oxidation and Mn(III/IV) oxide precipitation in a distinct layer of the sand filter. The formation of Mn(III/IV) oxides contributes to abiotic As(III) oxidation and immobilization of As(V) by sorption to Fe(III) (oxyhydr)oxides.

1. Introduction

In many regions of South and South-East Asia, groundwater aquifers contain high concentrations of arsenic (As) (Mandal and Suzuki, 2002; Muehe and Kappler, 2014). Due to the chronic health effects associated with prolonged exposure to As-rich drinking water, the world health organization set the drinking water limit to 10 μg L^{-1} in 1993. Groundwater aquifers in South and South-East Asia often also contain high concentrations of dissolved iron (Fe) and manganese (Mn), which affect the taste and visual nature of the drinking water (Buschmann et al., 2007; Hug et al., 2008; Winkel et al., 2011). In rural areas of Vietnam, household sand filters have been established as cost-efficient and effective solution for the removal of As, Fe, and Mn from groundwater (Fig. 1). Previous studies have shown that at groundwater Fe/As ratios above 250 (weight based), sand filters can effectively reduce As concentrations in the filtered water to levels below 10 μg L^{-1} (Berg et al., 2006; Nitzsche et al., 2015).

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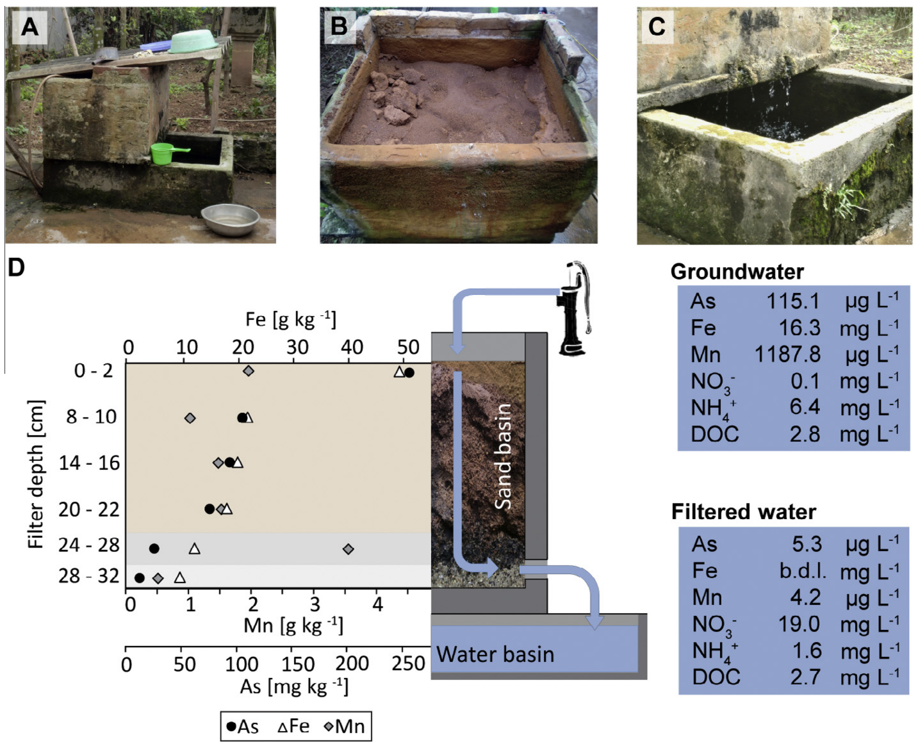

Fig. 1. Household sand filters used for arsenic, iron, and manganese removal from groundwater in Vietnam. (A) Shows a typical sand filter widely used in the Red River Delta in Vietnam. The sand filter consists of two basins stacked on top of each other. The upper basin is filled with sand and the lower basin serves as water storage container. The groundwater is pumped from a shallow tube well onto the filter, trickles though the sand column, and is collected in the bottom tank. (B) Top view on the sand filled upper basin and (C) water trickling into the storage container via holes in the bottom of the upper sand filled basin. (D) Schematic cross section of the sand filter showing a depth profile of the filter sand which is colored due to the precipitation and coating of sand particles with red-brown Fe(III) (oxyhydr)oxides (depth of 0–23 cm) and black Mn(IV) oxides (depth of 23–28 cm). The bottom sand layer (depth of 28–32 cm) shows the initial color of the filter sand. Diagram shows a depth profile of accumulated As, Fe, and Mn in the different colored sand layers quantified by XRF (Nitzsche et al., 2015). Blue boxes show concentrations of As, Fe, Mn, NO_3^-, NH_4^+ and DOC in the groundwater before and after filtration (b.d.l. = below detection limit) (Nitzsche et al., 2015).

The geochemical processes for the removal of As, Fe, and Mn from anoxic groundwater in aerated sand filters are well known: Dissolved Fe(II) is oxidized which leads to the formation of poorly soluble Fe(III) (oxyhydr)oxide minerals on the sand particles (Jessen et al., 2005; Voegelin et al., 2014). Fe(II) oxidation at neutral pH can result from homogeneous oxidation of dissolved Fe(II) by oxygen, heterogeneous oxidation of Fe(II) driven by catalytic surfaces reactions, and under microaerophilic oxygen concentrations by microbial Fe(II) oxidation (van Beek et al., 2012; Melton et al., 2014b). The abiotic Fe oxidation also causes co-oxidation of As(III) to As(V) which equally well or even more strongly adsorb to the newly formed Fe(III) (oxyhydr)oxide precipitates at pH >8 than As(III) (Dixit and Hering, 2003). Homogeneous Mn oxidation by oxygen at neutral pH is relatively slow, so that the formation of soluble Mn(III/IV) oxides occurs mainly as result of heterogeneous and microbial Mn oxidation (Tebo et al., 2004; Learman et al., 2011; Luan et al., 2012). Both Mn(III) and Mn(IV) oxides can function as oxidants for Fe(II) (Postma, 1985; Villinski et al., 2001) and As(III) (Tournassat et al., 2002; Ying et al., 2012). The formation of Mn oxides on the sand filter will therefore improve As and Fe removal from the filtered water (Nitzsche et al., 2015). A black Mn oxide layer developed when most of the reduced Fe and As has been removed from the groundwater (Nitzsche et al., 2015). The Mn oxide layer is therefore indicative for a transition zone below which Fe and As concentrations in the filtered water are considerably reduced.

Although much is known about the abiotic processes contributing to the performance of sand filters and their As, Fe, and Mn removal efficiency (Voegelin et al., 2014; Nitzsche et al., 2015), the composition of the microbial community and the potential contribution of different physiological groups of microorganisms to the removal of As, Fe, and Mn have not been investigated for household sand filters in Vietnam to date. Anoxic groundwater becomes aerated during filtration and oxic/anoxic interfaces at the surface of sand particles might allow Fe(II)-oxidizing bacteria to successfully compete with slower abiotic Fe(II) oxidation rates at reduced oxygen concentrations (Emerson and Moyer, 1997). Microbial As(III) respiration and/or detoxification mechanisms could affect As redox speciation on the filter and simultaneously sustain and secure the survival of a metabolically diverse microbial community (Oremland and Stolz, 2003). In addition, the slow kinetics of purely abiotic surface-catalyzed Mn(II) oxidation suggests that Mn(II)-oxidizing bacteria might be contributing to Mn removal in sand filters (Tebo et al., 2004; Voegelin et al., 2014). Previous studies have mostly relied on conventional molecular techniques (e.g. DGGE, T-RFLP) to describe the diversity and distribution of microorganisms in lab or full-scale biofilters for the simultaneous removal of As, Fe, Mn or ammonium (NH_4^+) from groundwater (Li et al., 2013; Yang et al., 2014), but no studies on sand filters used in actual households are available.

The aim of the present study therefore was a comprehensive and quantitative analysis of the composition and depth distribution of the microbial community in a sand filter, used by a rural household in Vietnam. More specifically, we performed most probable number (MPN) counts to quantify the number of microaerophilic and nitrate-reducing Fe(II)-oxidizing bacteria, Fe(III)-reducing bacteria, as well as Mn(II)-oxidizing, and Mn(IV)-reducing bacteria in both water samples and different