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Microbial communities contribute to the elimination of As, Fe, Mn, and NH_4^+ from groundwater in household sand filters

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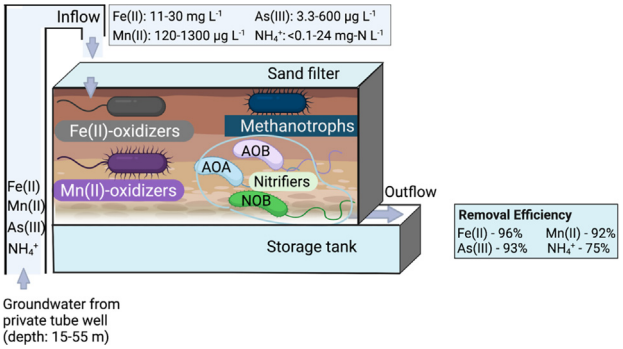
HIGHLIGHTS

- Performance and microbial communities of sand filters (SFs) were analyzed.
- Groundwater composition is responsible for differences in microbial communities.
- Fe- and Mn- oxidizers contribute to high As, Fe, Mn removal in sand filters.
- NH_4^+-oxidizing archaea followed by NO_2^--oxidizing bacteria transform NH_4^+ to NO_3^-.
- Microbial methane oxidation might take place in household SFs.

GRAPHICAL ABSTRACT



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ABSTRACT

Household sand filters (SFs) are widely applied to remove iron (Fe), manganese (Mn), arsenic (As), and ammonium (NH_4^+) from groundwater in the Red River delta, Vietnam. Processes in the filters probably include a combination of biotic and abiotic reactions. However, there is limited information on the microbial communities treating varied groundwater compositions and on whether biological oxidation of Fe(II), Mn(II), As(III), and NH_4^+ contributes to the overall performance of SFs. We therefore analyzed the removal efficiencies, as well as the microbial communities and their potential activities, of SFs fed by groundwater with varying compositions from low (3.3 µg L^{-1}) to high (600 µg L^{-1}) As concentrations. The results revealed that Fe(II)-, Mn(II)-, NH_4^+-, and NO_2^--oxidizing microorganisms were prevalent and contributed to the performance of SFs. Additionally, groundwater composition was responsible for the differences among the present microbial communities. We found i) microaerophilic Fe(II) oxidation by *Sideroxydans* in all SFs, with the highest abundance in SFs fed by low-As and high-Fe groundwater, ii) *Hyphomicropbiaceae* as the main Mn(II)-oxidizers in all SFs, iii) As sequestration on formed Fe and Mn (oxyhydr) oxide minerals, iv) nitrification by ammonium-oxidizing archaea (AOA) followed by nitrite-oxidizing bacteria (NOB), and v) unexpectedly, the presence of a substantial amount of methane monooxygenase genes (*pmoA*), suggesting microbial methane oxidation taking place in SFs. Overall, our study revealed diverse microbial communities in SFs used for purifying arsenic-contaminated groundwater, and our data indicate an important contribution of microbial activities to the key functional processes in SFs.

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1. Introduction

Household filters are recommended by the WHO as a solution to increase access to safe water for drinking purpose (WHO, 2011). Household filter technologies are modified depending on the sources and the identity of water contaminants, with the aim to eliminate contaminants such as pathogens, inorganic/organic compounds, and toxic metals (Freitas et al., 2022). Besides slow household sand filters (SHSFs) applied to remove particles and pathogens (Andreoli and Sabogal-Paz, 2020; Elliott et al., 2008; Medeiros et al., 2020), household filters to remove As have been widely implemented in developing countries such as Nepal (Kanchan filter) (Mueller et al., 2021; Ngai et al., 2007), Bangladesh (SONO filter) (Hussam and Munir, 2007; Neumann et al., 2013), and Vietnam (Berg et al., 2006).

In Vietnam, since the early 1990s until today, household sand filters (SFs) represent a conventional and popular treatment method to eliminate Fe and As from groundwater (Berg et al., 2006). Groundwater in Vietnam typically contains <0.01–48 mg L^{-1} Fe(II), 0.05–3.3 mg L^{-1} Mn(II), 10–382 μg L^{-1} As(III), and < 0.25–96 mg-N L^{-1} NH_4^+ (Winkel et al., 2011). The groundwater is pumped intermittently into a top compartment meanwhile being mixed with atmospheric oxygen (O_2), bringing the dissolved oxygen(DO) of groundwater before entering the sand layer to 7–8 mg L^{-1} (VanLe et al., unpublished data). The aerated groundwater then trickles through the filter driven by gravity. The filtered water is drained out completely between each filtration, allowing O_2 to re-occupy into pore spaces in the sand layer. Outflow water is collected in a lower basin and used for drinking and cleaning purposes (Fig. 1A) (Berg et al., 2006; Luzi et al., 2004; Nitzsche et al., 2015a). The As removal efficiency is dependent on the Fe:As:P ratios in the groundwater. Accounting for the competition of P and As for sorption on Fe mineral surface, the mass ratio of (Fe-1.8P)/As has to be higher than 50 to ensure efficient As removal (Berg et al., 2006; Hug et al., 2008).

Fe(II), Mn(II), As(III), and NH_4^+ removal mechanisms in household SFs are governed by a complex interplay of biogeochemical processes. Abiotic Fe(II) oxidation by atmospheric O_2 forms poorly crystalline Fe(III) (oxyhydr)oxide minerals coated on sand grains (Berg et al., 2006; Voegelin et al., 2014). This process also mediates co-oxidation of arsenite to arsenate, which strongly adsorbs on Fe(III) (oxyhydr)oxide minerals (Hug and Leupin, 2003; Voegelin et al., 2014). Arsenic elimination in SFs predicted based on simple co-precipitation of As(III) with Fe(III) (oxyhydr)oxides was lower than observed for real household filters (Berg et al., 2006). Hence, it was suggested that As(III) oxidation might either be coupled to abiotic Mn(IV) reduction or is also catalyzed by bacteria. However, until now, there is only one study reporting the distribution of microbial communities in one household SF (Nitzsche et al., 2015a, 2015b). In this study, a black layer of MnO_2 coated on clean sand was observed in the bottom layer and an enrichment culture of manganese-oxidizing bacteria (MnOB) was obtained, suggesting the occurrence of biotic Mn oxidation in SFs (Nitzsche et al., 2015b; Voegelin et al., 2014). Additionally, microbial ammonium oxidation forming nitrate (NO_3^-) was demonstrated in the same SF (Nitzsche et al., 2015b).

However, microbial communities of sand filters can differ when fed by groundwater with varying geochemical composition. Hence the contribution of biotic oxidation of Fe(II), Mn(II), As(III), and NH_4^+ in household SFs with varying groundwater geochemistry is still unrevealed. Additionally, other factors such as filter dimensions, height of the sand layer, and irregularities during operation (Fig. S3) can influence microbial activities in SFs and control the removal efficiency of Fe(II), Mn(II), As(III), and NH_4^+. In order to evaluate the microbial composition of SFs when being fed by varying groundwater composition, and how the microorganisms contribute to the performance of the household SFs, a detailed study on microbial communities in household SFs running under different geochemical conditions is required.

Therefore, we investigated a series of household SFs in high to low-As areas in the Red River delta in Vietnam. The main objectives of this study were (i) to analyze operational conditions and contaminant removal efficiency of 20 household SFs running under varying geochemical conditions, (ii) to identify the microbial community composition focusing on potential Fe(II)-, Mn(II)-, As(III)- and NH_4^+-oxidizers among 7 households SFs (selected based on differences in groundwater composition and depth of groundwater abstraction), and (iii) to elucidate the influence of groundwater geochemistry on the microbial community structures in household SFs.

2. Materials and methods

2.1. Study areas and sample collection

To select suitable field sites to study the effect of varying groundwater composition on microbial communities in household sand filters, we created a GIS-based searchable map based on the Red River's groundwater database from Winkel et al.(Winkel et al., 2011) (ArcGIS Map, Esri, USA) (details are described in SI). The selected sampling sites are in 3 different villages, i.e., Van Phuc (20° 55′ 08.63" N, 105° 53′ 47.61″ E) called site A; Tu Nhien (20° 51′ 30.05" N, 105° 55′ 33.34″ E) called site B; and Dung Tien (20° 49′ 01.21" N, 105° 51′ 13.59″ E) called site C (Fig. S1). Sand filter ID, coordinates, and the location at each field site are listed in Table S1.

All samples were collected during sampling campaigns in 2018 and 2020. Groundwater before flowing through the filter (termed inflow water), and

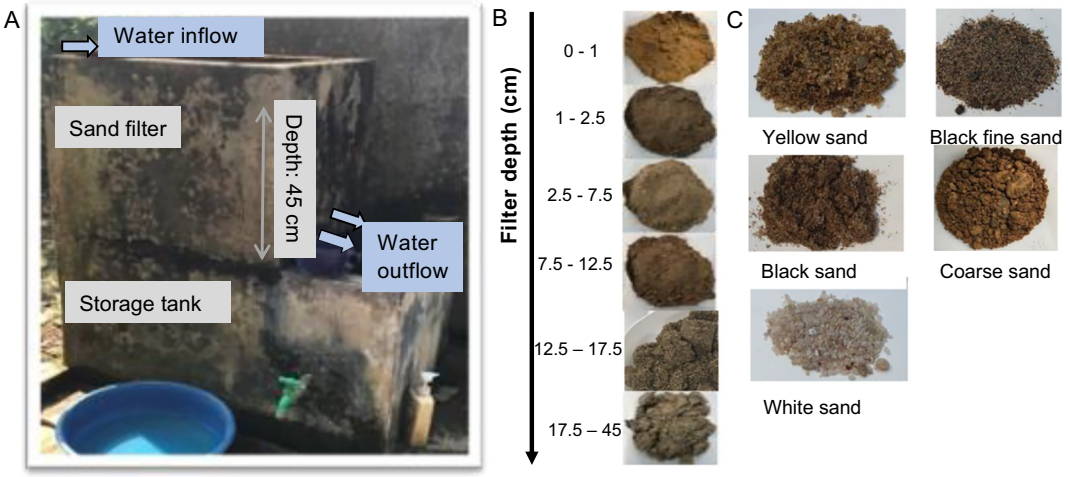

Fig. 1. Example of a household SF (SF B5) applied to remove As and Fe from groundwater in the Red River delta (A), heterogeneous stratification of sand layers with depth of the sand filter (top compartment of the filter) (B), and different types of sands commonly used as filter materials in household SFs (C).

water after filtration (termed outflow water) were filtered through 0.45 μm membrane filters and acidified on-site for later total element analysis with inductively coupled plasma mass spectrometry (ICP-MS, XSeries 2, Thermo-Fisher) and DOC analysis (High TOC II, Elementar). Non-acidified samples were used for NH_4^+, NO_3^-, NO_2^- quantification by continuous flow analysis (Seal Analytical AA3, Norderstedt, Germany). Water samples were collected in triplicate for each analysis, immediately cooled on ice during transport, and stored at 4 °C.

Water samples for DNA extraction were collected in sterile 5 L PE bottles. The water was filtered through 0.22 μm pore size, sterile membrane filters (EMD Millipore) using a suction-type filter holder (Sartorius 16,510) connected to a vacuum pump (Microsart®). Sand filter material was collected by a sterile spatula in different depth layers (every 1–5 cm) in SFs and stored in sterile Falcon tubes and zip bags. All sediment samples were collected in triplicates and stored on dry ice during transport. Sand for physical and chemical characterization was stored at 4 °C. Samples for microbial community analysis were stored at −20 °C.

2.2. DNA extraction

DNA of sand and water samples (collected biomass on membrane filters with 0.22 μm pore size) was extracted in triplicate by PowerWater® and PowerSoil® DNA Isolation Kits, respectively. 0.25 g of sediment or ¼ of a membrane filter was used for extraction based on the manufacturer's protocols. DNA concentrations were quantified by a Qubit® 2.0 Fluorometer with DNA HS kits (Life Technologies, Carlsbad, CA, USA).

2.3. 16S rRNA gene amplicon sequence analysis

Bacterial and archaeal 16S rRNA genes were amplified using universal primers 515f: GTGYCAGCMGCCGCGGTAA (Parada et al., 2016) and 806r: GGACTACNVGGGTWTCTAAT (Apprill et al., 2015) fused to Illumina adapters. Details of the PCR assay are mentioned in the SI.

Library preparation steps (Nextera, Illumina) and 250 bp paired-end sequencing with MiSeq (Illumina, San Diego, CA, USA) using v2 chemistry were performed by Microsynth AG (Switzerland), and between 19,219 and 259,470 read pairs were obtained for each sample. Raw sequencing data were analyzed with nf-core/ampliseq v1.2.0 (Straub et al., 2020) as detailed in the SI. Raw sequencing data can be found at the NCBI Sequence Read Archive (SRA); accession number PRJNA757982 (https://www.ncbi.nlm.nih.gov/bioproject/PRJNA757982).

2.4. Statical analysis

Spearman's rank correlations and *p*-values were calculated using the Hmisc packages version 4.6–0 (Harell Jr., 2021). Multiple testing corrected p-values (false discovery rate; fdr) were generated using the Benjamini-Hochberg method (Benjamini and Hochberg, 1995) in R version 4.1.2 (Team, 2021) (Table S2).

Bray Curtis dissimilarity metrics, Principal Coordinates Analysis (PCoA) plot was generated with https://github.com/qiime2/q2-diversity in QIIME2 v2019.10 within the nf-core/ampliseq pipeline.

The ADONIS (Permutational Multivariate Analysis of Variance Using Distance Matrices) test (Anderson, 2001) was performed with QIIME2 v2019.10 on the previously calculated Bray Curtis dissimilarity metrics. ADONIS computes an R2 value (effect size) which shows the percentage of variation explained by a condition, as well as a *p*-value to determine the statistical significance. To determine the effect size of a specific condition, the variance of other factors was removed (statistically controlled for), see Table S3 for formula and test results.

2.5. Quantitative polymerase chain reaction (qPCR)

Quantitative PCR (qPCR) was used to quantify general 16S rRNA genes of bacteria and archaea, 16S rRNA genes of anaerobic ammonium-oxidizers (anammox), as well as functional genes, i.e., targeting the ammonium monooxidase (*amoA*) of bacterial and archaeal ammonia-oxidizers (AOB, AOA respectively), the nitrite oxidoreductase (*nxrB*) of bacterial nitrite-oxidizers (NOB), and methane monooxygenase genes (*pmoA*) of methanotrophs. The qPCR protocol was performed using an iQ5 real-time PCR system (iQ5 optical system software, version 2.0, Bio-Rad). qPCR primer sequences, gene-specific plasmid standards, and details of the thermal programs are given in the SI (Table S5).

3. Results and discussion

3.1. Characterization and performance evaluation of household SFs

We first examined the design, performance, and solid-phase composition of 20 household SFs treating low (3.3 μg L^{-1}) to high (600 μg L^{-1}) As-containing groundwater in 3 villages located in the hotspots of As groundwater contamination in the Red River delta based on previous predictions (Fig. S1) (Winkel et al., 2011).

3.1.1. SF design parameters and operational conditions

The sand filters are built as rectangular-shaped concrete boxes (open at the top), filled with sand (the height of the sand column ranged between 35 and 100 cm with a sand particle size of ca. 0.05–2.0 mm), and filter bed volumes varied from 0.2 to 1 m^3 depending on the demand of purified water (Table 1). The groundwater originated from an aquifer depth of ca. 15–55 m (Table 1) and was pumped intermittently on top of the sand surface (Fig. 1). The water inflow and outflow pH values were 7.0 ± 0.4 and 7.3 ± 0.3, respectively, and the pH of the SF material was approximately 7.5. The filtration rate recorded in the field was between 1 and 4 $m^3 m^{-2} h^{-1}$ with an estimated hydraulic retention time of ca. 30 min

Table 1
Design parameters and operational conditions of selected household sand filters in the Red River delta in Vietnam. Data were collected in sampling campaigns in 2018 and 2020. The sand filters were collected at 3 field sites (A: Van Phuc, B: Tu Nhien and C: Dung Tien villages) along the Red River delta, coordinates of each filter ID and the location of the 3 field sites are listed in Table S1.

Filter ID	Surface area (m^2)	Sand layer volume (m^3)	Filtration rate ($m^3/m^2.h$)	Tube well depth (m)	Age of SF (years)	Design/ operational conditions		
						Filtration	Pre-aeration	Frequency of changing the sand (months)
(Voegelin et al., 2014)	0.47	0.2	2.5	45	8	1 step	NO	12
A1	0.28	0.2	4.2	35–45	1	2 steps	NO	6–7
	0.73	0.53	1.64					
B3	0.78	0.33	1.5	45–50	5	1 step	NO	12
B4	0.3	0.15	4	15–20	10	1 step	NO	24
B5	0.4	0.2	3	25–30	<1	1 step	NO	3–6
B8	1.15	0.7	1	35–40	>5	1 step	YES	6–12
C17	0.78	0.6	1.5	40–55	>5	1 step	YES	2–3
C18	0.65	0.52	1.8	40–50	<3	1 step	YES	6

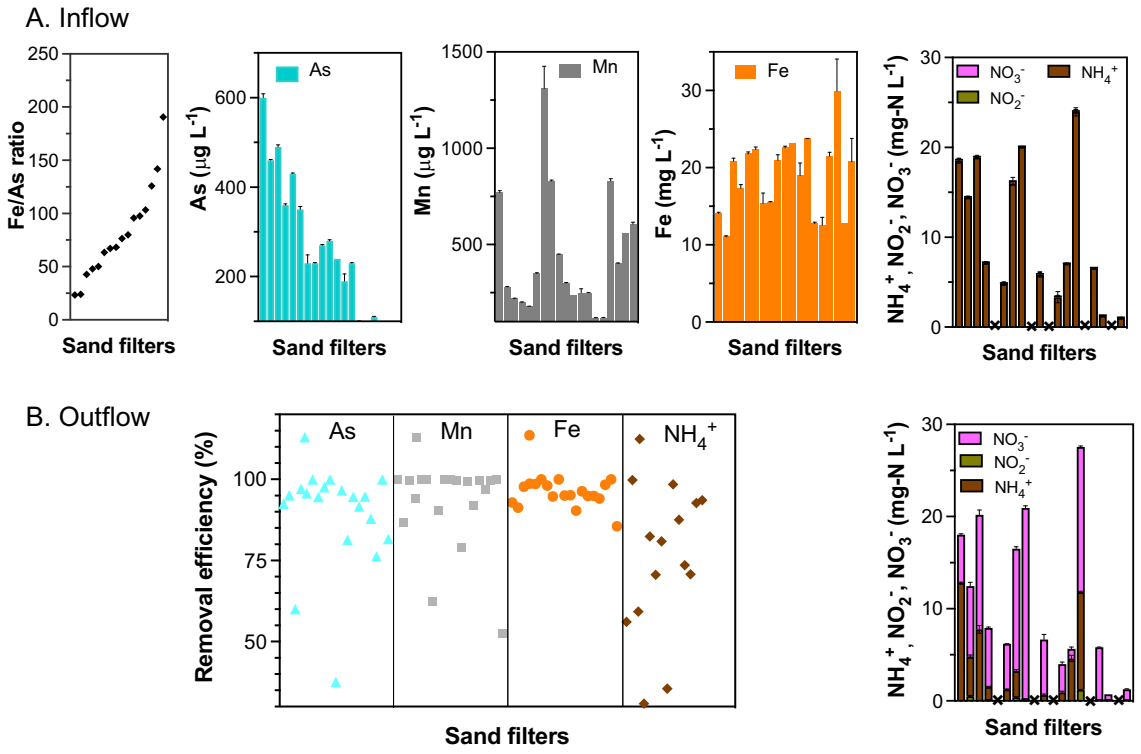

Fig. 2. Groundwater composition of 20 studied SFs plotted in the order of increasing Fe/As ratios. From the left to the right: Fe/As ratio, As, Mn, Fe, NH_4^+, NO_2^-, and NO_3^- concentrations (indicated as inflow, panel A). Panel B shows the removal efficiency of As, Mn, Fe, and NH_4^+ of 20 studied SFs (B, left) and concentrations of NH_4^+, NO_2^-, and NO_3^- after filtration (B, right). All samples were analyzed in triplicate, and error bars indicate standard deviation.

(Table 1). These parameters were confirmed in previous studies (Berg et al., 2006; Nitzsche et al., 2015a; Voegelin et al., 2014).

3.1.2. Performance of household SFs (Fe, Mn, As, and NH_4^+ removal efficiency)

The Fe, Mn, As, and NH_4^+ removal efficiency of SFs running with different groundwater compositions showed that independent from the filter design, size, and frequency of operation, SFs efficiently removed Fe, Mn, As, and partly oxidized NH_4^+ to NO_3^-. The groundwater pumped into the 20 studied SFs contained 11–30 mg L^{-1} Fe, 120–1300 μg L^{-1} Mn, 3.3–600 μg L^{-1} As, and < 0.1 to 24 mg-N L^{-1} NH_4^+ with Fe/As ratios ranging from 23.4 to 190 (Fig. 2A). After filtration, the removal efficiency of Fe, Mn, and As reached (on average) 96 ± 3.8 %, 92.2 ± 13.9 %, and 92.8 ± 7.2 %, respectively, and NH_4^+ was transformed to a large extent, but not completely, to NO_3^- (74.7 ± 29.8 %) (Fig. 2B, S5). However, in some filters, the remaining As concentration after filtration (averaging 17 ± 21 μg L^{-1}) was still higher than 10 μg L^{-1} (the drinking water standard provided by the WHO), thus requiring further treatment for being used as safe drinking water. Indeed, a Spearman correlation analysis revealed that the As concentration in the outflow was significantly positively correlated with As concentrations of the water inflow(correlation = 0.6; fdr = 0.02), but negatively correlated with $\frac{Fe}{As}$ ratios (correlation = −0.75, fdr = 0.004) and $\frac{Fe - 1.8P}{As}$ ratios (correlation = −0.8; fdr = 0.0001) (Fig. 3, Table S2). Additionally, the formation of NO_2^- and NO_3^- in the outflow was positively correlated to the amount of NH_4^+ in the inflow, suggesting nitrification during the filtration process (Fig. 3, Table S2).

3.1.3. Comparison of operational conditions and performance of household sand filters in Vietnam with other household SFs

In this context, we compared the designs and performances of sand filters applied to remove As in Vietnam with i) original household slow sand filters (HSSFs), ii) SFs employing zero-valent iron (SONO, Kanchan, and NIS) and iii) delayed aeration sand filters. The relevant comparison parameters are shown in Table 2.

Overall, the designs, flow regimes, and maintenances of household SFs were similar in all filters. However, differences in performance were quite substantial. Firstly, the pathogen removal efficiency of As filters including the ones used in Vietnam, the elemental iron-based filters and the delayed

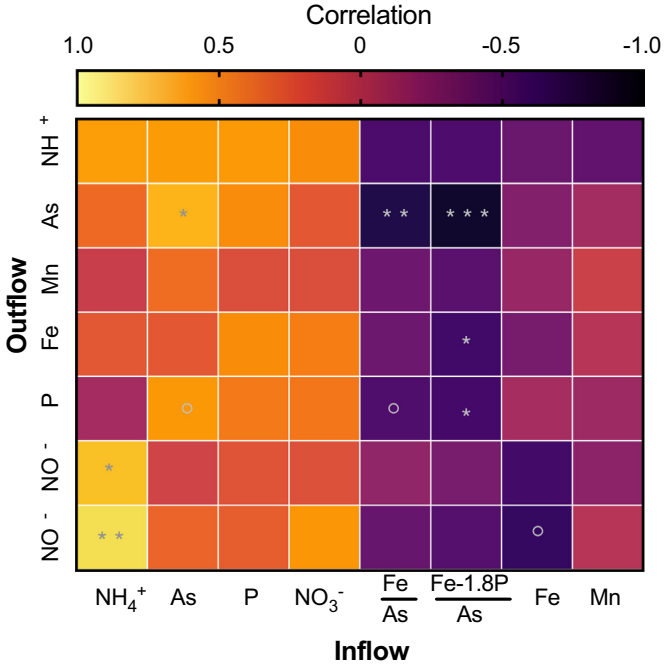

Fig. 3. Relationship of water inflow and outflow based on Spearman correlation (°: 0.05 < fdr > 0.1, *: fdr < 0.05, **: fdr < 0.01, ***: fdr < 0.001).

Table 2
Comparison of operational conditions and performance of household sand filters in Vietnam with others household sand filters.

	SFs in Vietnam	Elemental iron-based filters			Delayed aeration	HSSFs
		SONO	Kanchan	NIS		
Aim	Remove As and Fe and Mn	Remove As, Mn and Fe	Remove As	Remove As	Remove As	Remove turbidity and pathogens
Area/ Country	Vietnam (Red River delta)	Bangladesh Burkina Faso	Nepal Cambodia	China	Bangladesh	
Flow regime	Intermittent	Intermittent	Intermittent	Intermittent	Intermittent	Intermittent
Flow rate (Lh^{-1})	n.a	20–30 8–12	10–15	n.a	9	1
Filtration rate ($m^3m^{-2}h^{-1}$)	1–4	0.1–0.15	n.a	n.a	1	0.06
Maintenance (Require when clogging	Remove top sand layers (5–10 cm)	Remove top sand layers (2.5 cm)	Stirring 5 cm	Remove the top layer and slit up the nails	Backwashing	
Feeding volume (L)	30–100	40–80	18	80	n.a	48
Schmutzdecke	NO	NO	YES	YES		YES
Re.Eff. Fe (%)	96 ± 3.8	> 90	90–95	98	97	n.a
Re.Eff. Mn (%)	92.2 ± 13.9	n.a	n.a	–800[c]	n.a	n.a
Re.Eff. As (%)	92.8 ± 7.2[a]	94–99 60–80[a]	85–90 39–75[a]	86–95	92	n.a
Re.Eff. NH_4^+ (%)	74.7 ± 29.8[b]	n.a	n.a	n.a	58[b]	n.a
Re.Eff. Turbidity (%)	n.a	n.a	80–95	−44[c]	n.a	64 ± 9
Re.Eff. *E. coli* (log)	n.a	n.a	n.a	n.a	n.a	1.5–1.8
Re.Eff. Coliform	-10^3 (times)[c]	n.a	60–99 (%) 15–99 (%)	n.a	n.a	n.a
References	This study (Nitzsche et al., 2015a) (Berg et al., 2006)	(Bretzler et al., 2020) (Hussam and Munir, 2007) (Neumann et al., 2013)	(Chiew et al., 2009) (Ngai et al., 2007) (Mueller et al., 2021)	(Smith et al., 2017)	(Annaduzzaman et al., 2021)	(Freitas et al., 2022; Medeiros et al., 2020)

[a] As effluent was higher than drinking standard (10 μg L^{-1})
[b] Incomplete nitrification.
[c] Effluent value was higher than the influent.

aeration filters were insufficient compared to HSSFs, probably due to the absence of the "Schmutzdecke" as a biofilter layer. Secondly, the performance of iron-based filters applied to remove As, including SONO filters containing a layer of composite iron matrix (CIM) under a coarse sand layer (Hussam and Munir, 2007; Neumann et al., 2013), Kanchan filters containing an iron nail compartment above a sand layer (Mueller et al., 2021; Ngai et al., 2007), and NIS filters containing a layer of iron nails between two sand layers (Smith et al., 2017), were compared. Kanchan, and NIS did not effectively remove As when the groundwater simultaneously contained high P and low Fe (Bretzler et al., 2020). So far, the SONO (Neumann et al., 2013) and NIS filters (Smith et al., 2017) have shown the best As removal efficiency (As outflow <50 μg L^{-1}, average As removal rate 92 %). Lastly, filters that are based on delayed aeration, which means anoxic groundwater was stored in an anoxic container before aeration and a 2-steps sand filtration, were suggested as a promising solution for As elimination. However, the maintenance of these filters required backwashing, and their costs are slightly higher than for the other household SFs. Thus, delayed aeration filters might be more suitable for a small community rather than one family (Annaduzzaman et al., 2021).

3.2. Microbial community composition and shared core taxa in household SFs

To understand how microbial communities of SFs differ when running with varying groundwater composition, the microbial community composition from 7 selected households was analyzed and taxa that were identified among all SFs were defined as the core microbial community. Fe, Mn, As and NH_4^+ concentrations of groundwater feeding 7 selected filters ranged from 12.5 to 30 mg L^{-1} Fe, 120–830 μg L^{-1} Mn, 3.3–490 μg L^{-1} As, and 1.0–24 mg-N L^{-1} NH_4^+ (Fig. S7). Microbial community analysis revealed the predominance of taxa potentially involved in Fe(II), Mn(II), NH_4^+ and, unexpectedly, CH_4 oxidation, suggesting their contribution to the excellent performances of the different SFs.

3.2.1. Microbial Fe, Mn, and As oxidation

In contrast to previous studies that suggested a minor contribution of Fe (II)-oxidizing bacteria in the filters (Berg et al., 2006; Nitzsche et al., 2015b; Voegelin et al., 2014), we found the well-known Fe(II)-oxidizing genus *Sideroxydans* with a relative abundance of 0.1–13.2 % predominantly in the top layer of all investigated SFs (Fig. 4A). Additionally, since NO_3^- formed due to the nitrification processes, it became a dominant electron acceptor in the deeper layers of the filters where O_2 is depleted. We also suggest that *Gallionellaceae* (3.0 ± 3.0 %) belonged to the core microbial community that was shared in all studied SFs (Fig. 4B) to be involved in nitrate reduction coupled to Fe(II) oxidation (NRFeOx) because recently several novel species of NRFeOx bacteria (*Candidatus* Ferrigenium) (Huang et al., 2022) belonging to the *Gallionellacea* family were enriched under similar conditions as present in the sand filters (poor in organic carbon and enriched in NO_3^- and Fe) (Huang et al., 2021; Jakus et al., 2021). Therefore, the high Fe removal efficiency in the SFs (on average 96 ± 3.8 %) might be achieved by a combination of heterogeneous abiotic Fe(II) oxidation and microbial Fe(II) oxidation. The formation of extra Fe(III) phases via microbial Fe(II) oxidation then provides additional minerals with binding sites for As removal, ultimately enhancing As removal efficiency of the SF.

The genera *Hyphomicrobium* and *Pedomicrobium,* belonging to the core microbial community affiliating with *Hyphomicrobiaceae* (2.8 ± 1.8 %) (Fig. 4B) were likely the key MnOB in the SFs. They contain the Mn(II)-oxidizing gene *moxA* and were used as model organisms for studying Mn (II) oxidation (Larsen et al., 1999; Ridge et al., 2007). Microbial Mn(II) oxidation is considered a predominant process over abiotic Mn(II) oxidation in SFs, due to the much slower abiotic Mn(II) oxidation rate (half-live times of Mn(II) controlled by abiotic oxidation on mineral surfaces are between 5 and 2800 days) (Davies and Morgan, 1989; Diem and Stumm, 1984; Morgan, 2005). In comparison to abiotic Mn(II) oxidation, the presence of MnOB can accelerate the process up to several orders of magnitude (Hansel, 2017; Nealson, 2006; Tebo et al., 2005). The formation of Mn oxides via biotic Mn(II) oxidation provides a second possibility for As(III) oxidation in SFs (by the reactive Mn oxides). Therefore, SFs show a very stable As removal efficiency under different geochemical conditions.

In contrast to the evidence for microbial Fe(II)- and Mn(II)-oxidizers in the SFs, we did not see extensive evidence for biological As oxidation. This suggests that As(III) as predominant As species in the groundwater was abiotically oxidized to As(V) and immobilized in the filter bed (Voegelin et al., 2014). The As(III) oxidation processes are mainly governed by Fe

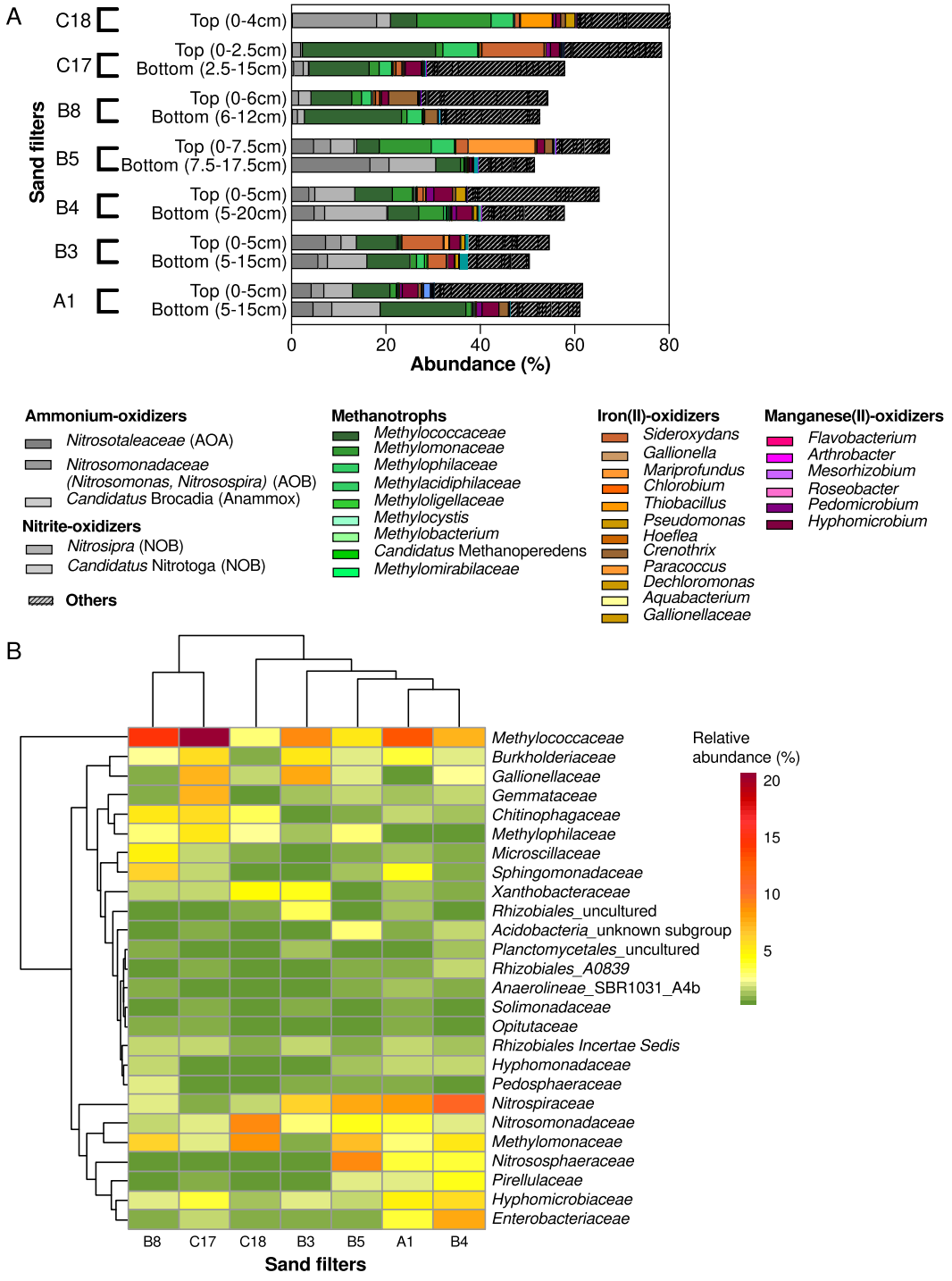

Fig. 4. Microbial composition in 7 selected household sand filters based on 16S rRNA gene amplicon sequencing. Relative abundance of selected taxa (including potential function) in two distinct layers in sand filters: the top layer is considered to be oxic and Fe(III)-rich, while the bottom layer is considered to be oxygen-poor (A). Relative abundances of core microbial communities (at the family level) that were shared across all selected sand filters (B).

(II,III)-mediated abiotic surface-catalyzed As(III) oxidation (Amstaetter et al., 2010; Hug and Leupin, 2003) or coupled to Mn(III, IV) reduction (Bai et al., 2016; Gude et al., 2017; Katsoyiannis et al., 2004).

3.2.2. Nitrification in sand filters

Geochemical data indicated NH_4^+ oxidation and NO_3^- formation during filtration processes with conversion rates (on average) of 74.7 ± 29.8 % (Fig. 2B). Additionally, we found sequences affiliating with typical nitrifiers, such as *Nitrosomonadaceae* (AOB), *Nitrososphaeraceae* (AOA), and *Nitrospiraceae* (NOB) accounting for 13.5 ± 8.4 % relative abundances of the microbial community (Fig. 4A). We thus conclude that nitrification likely plays a crucial role in nitrogen species transformation in the SFs.

We further investigated the vertical distribution of ammonium- and nitrite-oxidizers in different depths by qPCR. Bacterial and archaeal *amoA* genes (targeting AOA and AOB), the *nxrB* gene (targeting NOB), and the 16S rRNA gene targeting anammox taxa as well as total bacteria and total archaea were analyzed from selected SFs. These SFs were selected due to their highest (SF-A1, B5) and lowest (SF-B4) NH_4^+ removal efficiency (Fig. 5). Our results showed that AOA was predominant over AOB in 3 selected filters with an (qPCR-based) relative abundance of archaeal *amoA* genes ranging from

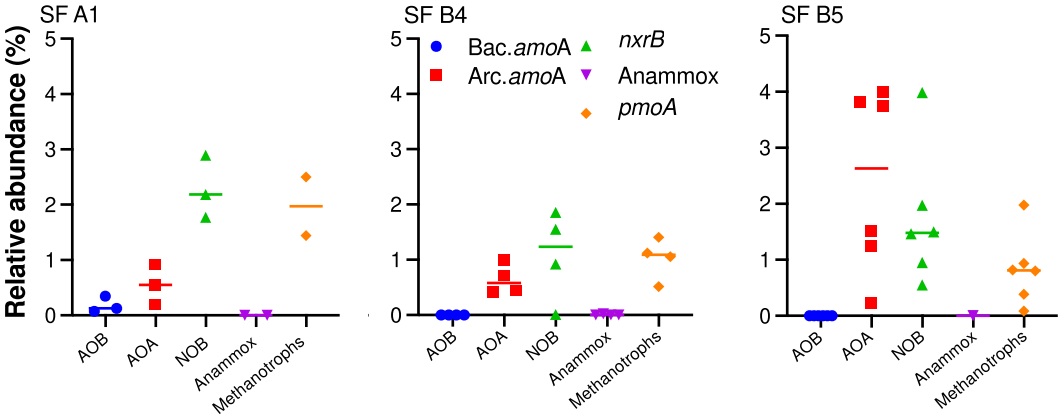

Fig. 5. Relative abundance of AOA, AOB, NOB, anammox, and methanotrophs was calculated from quantitative PCR results targeting 16S rRNA genes of total bacteria, archaea, anammox taxa, and functional genes such as archaeal and bacterial *amoA* affiliated to AOA and AOB, respectively, *nxrB* genes of NOB, and *pmoA* genes of methanotrophs. Samples were analyzed in three different filters, representing the highest (SF A1 and B5) and lowest (SF B4) performances of NH_4^+ oxidation. Error bars represent the standard deviation of triplicates.

0.5 to 2.5 %. In contrast, the bacterial *amoA* gene targeting AOB was <0.3 % (Fig. 5), and its copy number was reduced along the filter depths by the factor from 2 to 10 (Fig. S9). A relative abundance of *nxrB* genes of NOB was also detected between 2 and 4 % in selected filters (Fig. 5). The presence of AOB (*Nitrosomonas*) and NOB (*Nitrospira*) was commonly found in other full-scale water treatment SFs (Albers et al., 2015; Hu et al., 2020; Palomo et al., 2016; Poghosyan et al., 2020), but the remarkable occurrence of AOA is a unique finding for the household SFs of our study. Quantification of functional genes of AOA and NOB along the filter depths suggested their widespread abundance along the sand filters, even in deeper sand layer (below 15 cm) (Fig. S9) where we observed a depletion of AOB. Both AOA and *Nitrospira*-NOB were found to have a better tolerance to low DO level (Mehrani et al., 2020; You et al., 2009). Some studies showed that they can be active and enriched even at a DO concentration below 1.0 mg O_2/L (Erguder et al., 2009; Mehrani et al., 2020). Finally, a small fraction of anaerobic ammonium-oxidizing bacteria (anammox) was also detected below 5 cm depth in our sand filters (<0.3 %) (Fig. 5). However, their functional role needs to be confirmed in future studies.

3.2.3. Potential methane oxidation in sand filters

Unexpectedly, a high relative abundance of putative methanotrophs was detected in all SFs, such as *Methylococcaceae* (10.3 ± 6.1 %) and *Methylomonaceae* (4.5 ± 2.8 %) (Fig. 4B). Since an elevated concentration of CH_4 (up to 45 mg L^{-1}) was detected in the groundwater at the same field sites (Glodowska et al., 2020; Stopelli et al., 2020), we speculated that the microbial communities in the household sand filters might be capable of oxidizing methane. We therefore performed qPCR targeting the particulate methane monooxygenase genes (*pmoA*) for 3 selected sand filters (SF A1, B4, B5) and the results are presented as (qPCR) relative abundance (proportion of this specific gene in relation to total bacterial and archaeal 16S rRNA genes) in Fig. 5. The results confirmed the presence of methanotrophs in all 3 sand filters, with the abundance of *pmoA* genes accounting up to 2.5 % (Fig. 5). Most of the recognized methanotrophs belonged to type I aerobic methanotrophic bacteria that oxidize methane to CO_2 using O_2 as electron acceptor (Stein et al., 2012). Recent studies about methane oxidation in the same As-contaminated aquifer revealed anaerobic methane oxidation coupled to the reduction of Fe(III) minerals (by *Candidatus* Methanoperedens) (Glodowska et al., 2020; Pienkowska et al., 2021), suggesting a similar process might take place in the O_2-depleted zone in household SFs.

3.3. Influence of groundwater geochemistry on the microbial community structures in household sand filters

The groundwater chemistry is the primary factor affecting the structure of microbial communities in sand filters (Hu et al., 2020). Indeed, the Bray Curtis dissimilarity analysis also indicated that microbial community composition differed considerably among filters running under various groundwater compositions, whereas minor differences were observed between microbial composition in the top (2.5–7.5 cm) and bottom layer (between 2.5 and 17.5 cm) from the same SF fed by the same groundwater (Fig. 6 A). Additionally, we used the ADONIS test to analyze the relative influence of groundwater composition on microbial communities. The results indicated that each factor of Fe, Mn, As, and NH_4^+ inflow concentrations can influence microbial communities in sand filters between 15 and 20 % (Fig. 6B, Table S3). This finding is also in line with Hu et al., 2020, which revealed that Fe, Mn, NH_4^+ and PO_4^{3-} were strongly influencing the microbial communtiy variation in rapid sand filters. We noticed that in SFs B3 and C17, fed by groundwater enriched with Fe(II) (Fe concentration of 21 and 30 mg L^{-1} respectively) and depleted in As(III) (As concentrations of 3.3 and 16.97 μg L^{-1}, respectively), *Sideroxydans* was present as the most dominant potential Fe(II)-oxidizer, with a relative abundance of up to 8.8 and 13.2 %, respectively (Fig. 4A). Additionally, we also found that SF B4 had the lowest NH_4^+ removal rate (12 %), probably due to the lower abundance of ammonium-oxidizers as indicated by the ca. 10–100 times lower

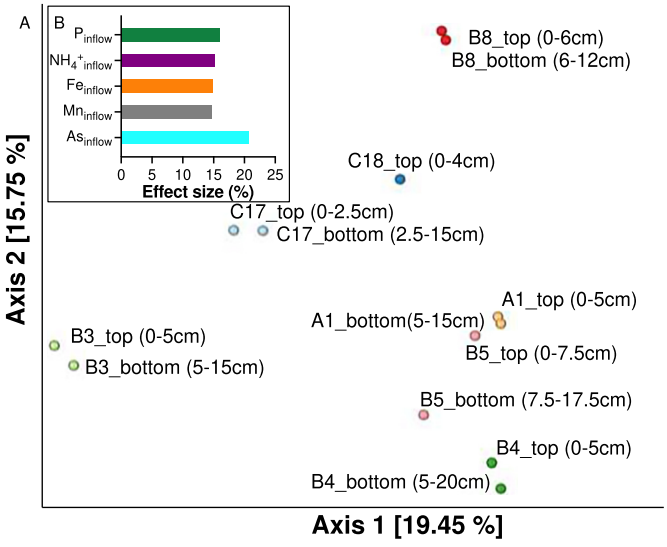

Fig. 6. Principal Coordinate Analysis (PCoA) plot based on Bray Curtis dissimilarity to compare the differences of microbial community composition based on 16S rRNA gene amplicon sequencing between samples (A). The effect of groundwater components (Fe, Mn, As, and NH_4^+ inflow) on microbial communities of SFs analyzed by ADONIS test, with Pr (>F) = 0.001 for all results (B).

amoA gene copy number in comparison to SFs which possessed a high NH_4^+ conversion rate such as SFs A1 and B5 (Fig. S8).

4. Conclusions

We conducted a comprehensive study on overall sand filter performance, microbial community composition, and the contribution of biological processes to the elimination of metal(loid)s (As, Mn, Fe) and nutrients (NH_4^+, NO_2^-, NO_3^-) from groundwater. Our findings suggest that Fe(II)- and Mn(II)-oxidizers probably complement abiotic oxidation in household SFs, thus contributing to the excellent Fe, Mn, and As removal efficiencies. Our data also indicated a role of AOA and NOB for NH_4^+ and NO_2^- oxidation and thus ammonium removal. Although, formed NO_3^- in filtered water did not exceed EU limits of 50 mg L^{-1}, it still raises a potential concern for the water quality when being used as drinking water. Finally, a high abundance of methanotrophs was detected in the sand filters, suggesting that SFs might also simultaneously oxidize CH_4 during the filtration processes. Overall, Fe(II)-, Mn(II)-, NH_4^+- and NO_2^--oxidizers belong to the core microbial communities in household SFs and their presence correlates with key functional processes in SFs.

CRediT authorship contribution statement

Anh Van Le: Conceptualization, Methodology, Investigation, Visualization, Writing – original draft. **Daniel Straub:** Formal analysis, Data curation, Writing – review & editing. **Britta Planer-Friedrich:** Resources, Writing – review & editing. **Stephan J. Hug:** Resources, Supervision, Writing – review & editing. **Sara Kleindienst:** Resources, Writing – review & editing. **Andreas Kappler:** Conceptualization, Resources, Supervision, Writing – review & editing, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.156496.

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