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Efficiency of a multi-barrier household system for surface water treatment combining a household slow sand filter to a Mesita Azul® ultraviolet disinfection device

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Abstract

Low-cost household technologies for water treatment are crucial to improving drinking water quality and preventing health, social and economic impacts, mostly in middle- and low-income regions. This work assessed the removal efficiency of physical-chemical and bacteriological parameters from river water by a multi-barrier household water treatment system for 113 consecutive days. This system combines a pre-treatment step through a non-woven synthetic blanket, filtration by an intermittent household slow sand filter (HSSF) and a Mesita Azul® ultraviolet disinfection device. In general, the water quality was improved by the evaluated system. Turbidity was removed by an average of 73% (ranging from 33 – 94%), total coliforms (TC) of 3.88 $\log_{10}$ (ranging from 2.22 – 5.16 $\log_{10}$) and *E. coli* of 2.49 $\log_{10}$ (ranging from 1.81 – 3.30 $\log_{10}$). Filtration improvement was mostly correlated to HSSF biofilm development and influent water quality. Characterisation of HSSF s*chmutzdecke* demonstrated a predominance of organic content, and a higher presence of carbohydrates than proteins on the sand and the blanket. Ultraviolet disinfection with Mesita Azul® inactivated most of

the remaining bacteria after filtration and no regrowth was observed after 15 days of disinfection. In conclusion, the multi-barrier household water treatment system was efficient in treating river water, reducing risks of microbial contamination to achieve safe drinking water.

Keywords: slow sand filter, turbidity, bacterial removal, s*chmutzdecke*, UV disinfection

1. Introduction

The quality of drinking water is a worldwide concern, mostly in low- and middle-income countries and communities. Nowadays, more than 2 billion people use drinking water contaminated with faeces. Moreover, 785 million people do not have access to basic drinking water services, and among them, 579 million people consume untreated surface water or from unprotected wells and springs (WHO, 2019a). Consumption of polluted water causes health, social and economic impacts. It is estimated that 485,000 deaths from diarrhoea per year are related to contaminated drinking water, mostly among children under five years old. Furthermore, illness caused by untreated water reduces the work productivity and school attendance, and increases medical costs for treatment (WHO, 2019a).

In this context, individual point-of-use, or household low-cost technologies are alternatives to deliver better drinking water quality in regions and communities that still consume directly from contaminated natural sources. An example is the use of a household slow sand filter (HSSF), which is a home scale of the slow sand filter. The HSSF can present different designs through which water is treated by a combination of processes inside the filter (Kubara and Haarhoff, 2010; CAWST, 2012; Jadhav et al. 2015; Freitas et al., 2022). The biological process occurs mainly on the top of the sand media, in the *schmutzdecke* (biofilm layer), while the physical-chemical processes take place along the sand media (CAWST, 2012; Jadhav et al. 2015; Ranjan and Prem, 2018). This combination is responsible for removing physical-chemical and biological contaminants from water sources,

improving water quality (CAWST, 2012; Verma et al.; 2017; Freitas et al., 2022). It is important to note that the *schmutzdecke* can take up to 6 weeks to fully develop, impacting the filtration and disinfection potential of the HSSF (Ranjan and Prem, 2018).

Several reports have indicated the efficiency of HSSF and application on field studies (Freitas et al., 2022). Briefly, these filters can remove more than 75% of turbidity (Napotnik et al., 2017; Andreoli and Sabogal-Paz, 2020; Maciel and Sabogal-Paz, 2020; Souza Freitas and Sabogal-Paz, 2020; Freitas et al., 2022; Terin and Sabogal-Paz, 2021), among other parameters, such as organic matter (Napotnik et al., 2017; Andreoli and Sabogal-Paz, 2020; Souza Freitas and Sabogal-Paz, 2020; Freitas et al., 2022), metals (Mwabi et al., 2011; Freitas et al., 2022) and nitrogen compounds (Mwabi et al., 2011, Freitas et al., 2022) . Bacterial removal can vary among the studies, but most of them report a removal range between 1 and 2 $\log_{10}$ (Young-Rojanschi and Madramootoo, 2014; Elliott et al., 2015; Terin and Sabogal-Paz, 2019; Maciel and Sabogal-Paz, 2020; Medeiros et al., 2020; Napotnik et al., 2020; Souza Freitas and Sabogal-Paz, 2020; Freitas et al., 2021). Moreover, HSSF can also remove other microorganisms, such as protozoa (Andreoli and Sabogal-Paz, 2020; Napotnik et al., 2020; Freitas et al., 2021), viruses (Jenkins et al., 2011; Elliott et al., 2015; Napotnik et al., 2020) and cyanobacteria (Terin and Sabogal-Paz, 2019).

Since HSSF optimal operation and efficiency depends on the raw water quality, a pre-treatment stage may be necessary (CAWST, 2012; Freitas et al., 2021). A pre-treatment with a non-woven blanket has been demonstrated as a cost-effective method to reduce the influent water turbidity and improve the HSSF operation with easy installation and maintenance (Freitas et al., 2021; Terin et al., 2021). Furthermore, since the bacterial removal usually does not achieve more than 2 $\log_{10}$, a disinfection step is essential to ensure safe drinking water, particularly while the *schmutzdecke* develops. The application of a pre-treatment step, followed by filtration and disinfection stages, characterises a multi-barrier treatment approach for water treatment (CAWST, 2012; WHO, 2017).

The use of ultraviolet (UV) light for water disinfection is widely known and technology advances have led to the development of low-cost UV equipment. One example is the Mesita Azul® ("little blue table" in Spanish), which is an innovative household UV disinfection system (Reygadas et al. 2015) developed by Cantaro Azul and the University of California, Berkeley. Mesita Azul® is a flow-through device designed to disinfect water in low-income settings with a low-pressure UVC lamp (254 nm) above a water layer of approximately 4 cm, which can operate at a flow rate up to 5 L/min, consuming 15 W of electricity and applying a dose of up to approximately 120 mJ/cm² (Figure S1 – Supplementary Material). The cost of installing a Mesita Azul® is around USD$ 80 (Reygadas et al. 2015). The efficiency of Mesita Azul® is reported in field studies, and the WHO International Scheme to Evaluate Household Water Treatment Technologies classified this technology as one-star, providing targeted protection against bacteria and protozoa in non-turbid water (Reygadas et al., 2015; 2018; WHO, 2019b).

Although the efficiency of HSSF is well documented (Freitas et al., 2022), as well the disinfection by UV, a real scale wholesome system involving a pre-treatment and the combination of HSSF with a post-treatment have not been widely studied in real conditions and with continuous operation. Few works evaluated the use of chlorine as post-treatment, which can alter the water taste and odour, reducing acceptability by consumers (Crider et al., 2018). Only one previous study, assessing the effectiveness of HSSF as a pre-treatment to UV disinfection, was able to fully disinfect and reduce the risk associated to drinking water (Frank et al., 2014). However, further research that explores the combination of HSSF and new UV technologies as post-treatment should emerge.

In this context, the main objective of this study was to evaluate, in real operation conditions, the removal efficiency of physical-chemical and bacteriological parameters by a household multi-barrier water treatment system and evaluate the effectiveness of Mesita Azul® as an HSSF post-treatment.

This system combines a pre-filtration step through a non-woven synthetic blanket and filtration by an intermittent HSSF followed by the Mesita Azul® UV disinfection device.

2. Methodology

2.1 The intermittent household slow sand filter (HSSF)

2.1.1 Structure

An HSSF was constructed as described by Terin et al. (2021) using PVC pipes and fittings with a cross-section area of 0.053 m², and with a 20 L bucket equipped with a float valve. This valve allowed limiting the water level above the filtration layer in approximately 15 cm, controlling the maximum filtration rate (Terin et al., 2021).

Gravel and sand used to fill the HSSF were acquired from local hardware and gardening stores, and it was previously washed, sun-dried and sieved. The support layer consisted of 7.5 cm of coarse gravel at the bottom, followed by 5 cm of fine gravel and 5 cm of coarse sand. Above that, the filtration media layer consisted of 50 cm of fine sand with an effective size (d_{10}) of 0.18 mm and uniformity coefficient (UC) of 1.56 mm (Table S1 - Supplementary material). Before introducing the filter media, clean well water was added to the filter to prevent the formation of air pockets.

A layer of a non-woven synthetic blanket was placed above the filtration media using a PVC ring. This blanket can retain particles and ease maintenance, and also act as a support for the *schmutzdecke*.

The charging volume of 16 L was determined by the void volume inside the filter (Table S2 - Supplementary material). A schematic of HSSF is shown in Figure 1.

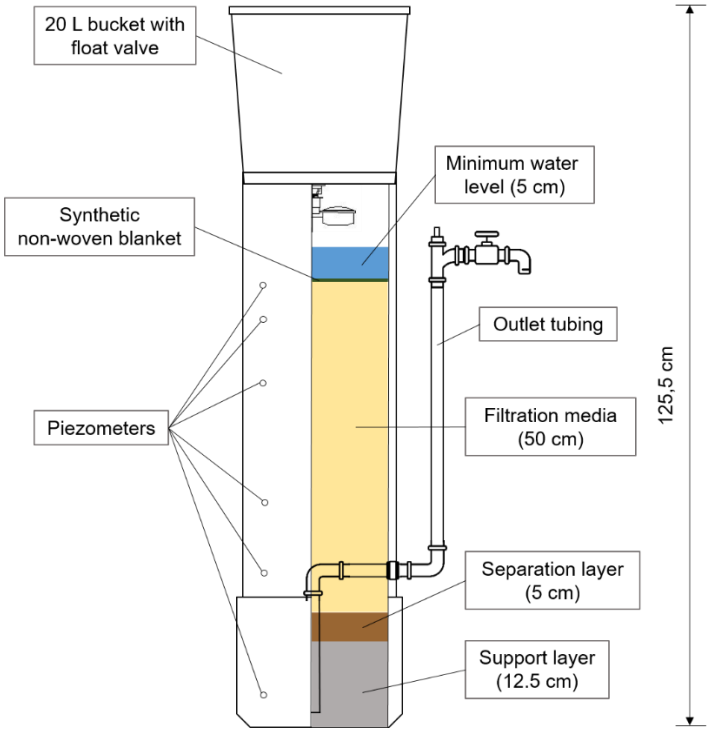

Figure 1. Intermittent household slow sand filter with a float valve (adapted from Terin et al. 2021).

2.1.2 Filtration rate and tracer tests

In order to characterise the total filtration time, the maximum filtration rate (MFR), and the time to reach MFR, the filter was fed with 16 L of clean well water and the filtered volume in the outlet tube was quantified every minute until the filtered volume was below 1 mL/min. This test was performed in triplicate.

The tracer test was performed to characterise the flow pattern of the filter, as described by Terin et al. (2021). Briefly, 200 mg/L of NaCl solution was prepared and two 16 L feeds were added into the filter, followed by feeds with clean well water, sufficient to remove all the NaCl solution from the filters. The interval between feeds was the total filtration time, which was previously determined (130 min). The conductivity was measured every minute with a probe (with *Go!Link* interface) positioned at the filter outlet tube and registered by Logger Lite software (Vernier Software & Technology, EUA). The tracer test was performed in triplicate.

Morrill Dispersion Index (MDI) and modified MDI (mMDI) were calculated according to Lynn et al. (2013) to determine flow behaviour (complete mixing or plug flow). While MDI can vary between 1.00 (ideal plug flow) and 22.00 (ideal complete mixing), the mMDI take into account the reactor volume and the number of feeds with the tracer solution. In this case, the range of mMDI was from 2.33 (ideal plug flow) and 22.00 (ideal complete mixing).

2.2 Operational settings

2.2.1 System functioning

In alternate days, 450 L of water from the Monjolinho River (São Carlos/Brazil) was pumped to a 500 L elevated reservoir and settled for 24h for sedimentation. The next day, approximately 150-200 L of water was pre-treated by filtration through a two-layered non-woven synthetic blanket and stored in a 200 L intermediary reservoir. Immediately, the water was transferred to a second 200 L intermediary reservoir by pumping until reaching full capacity. The remaining water from the elevated reservoir and the first intermediary reservoir was discharged to receive new river and pre-treated water, respectively.

The water in the second intermediary reservoir was used to supply the HSSF for two consecutive days before being filled again, except at weekends, which supplied the HSSF for three days. Thus, 16 L of water was pumped into an HSSF bucket three times a day (every 8 h). An electronic timer was programmed to turn the pump on and off for all feedings, thus not needing manual input. The filtered water was stored, and every day 48 L was disinfected by the Mesita Azul® UV device.

Prior to disinfection, Mesita Azul® was turned on for 5 min to allow lamp heating and then the 48L of filtered water was pumped into the device with an average flow of 1.5 L/min. The disinfected water was stored in a 60 L reservoir. A scheme of the complete system is presented in Figure 2.

The household water treatment system was operated for 113 consecutive days (from June 8th to September 29th, 2021). At the end of the operation, the treated and disinfected water was maintained for 15 days in the final reservoir to assess potential microbial regrowth daily.

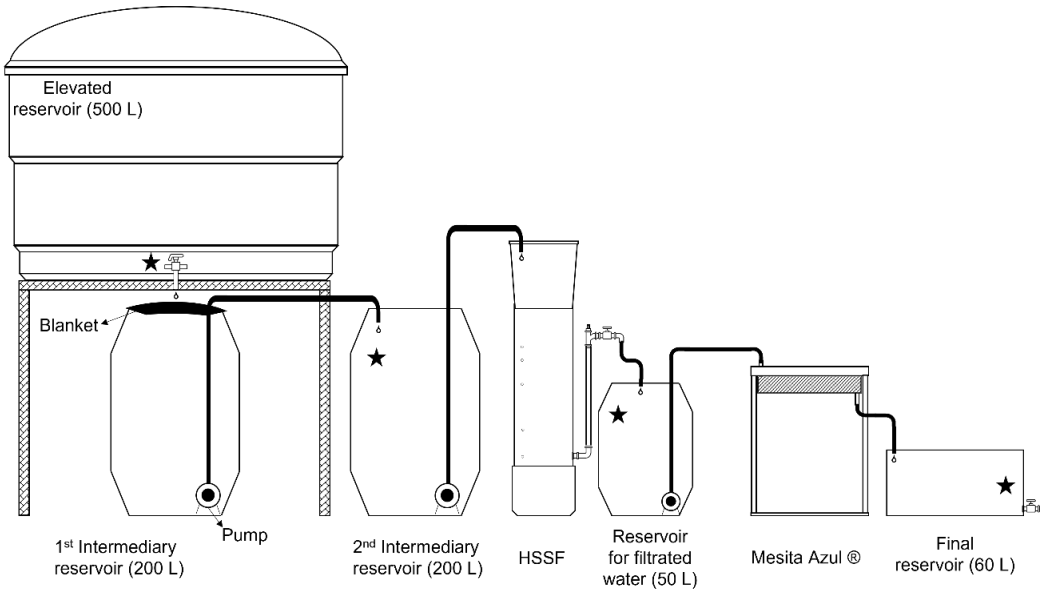

Figure 2. Complete household system for river water treatment combining a pre-treatment, filtration through HSSF and disinfection by the Mesita Azul® UV device. Stars indicate water sampling points.

2.2.2 Maintenance

The accumulation of particles and the growth of the biofilm layer reduce the HSSF filtration rate. The HSSF was unable to produce 48 L/day each time, which consisted of removing the non-woven synthetic blanket above the filtration layer, scraping and washing it with clean well water. Moreover, the first centimetres of the sand were cleaned by gently stirring and adding well water until the water above the sand layer appeared to be clean. The elevated reservoir was cleaned every two weeks by brushing the walls and washing it with clean well water.

2.3 Sample collection and analyses

Raw river water, pre-treated water, filtered water and disinfected water were collected and analysed during the morning on weekdays. Parameters, frequency of analyses and the methodology applied are presented in Table 1. Standard Methods (APHA, 2012) were followed to evaluate the parameters. MFR and head loss were measured daily in the HSSF, 20 min after the first feed of the day.

Table 1. Parameters, frequency of analyses and methodology applied to measure the water quality

Parameter	Frequency	Methodology/equipment
Temperature	Daily[c]	Mercury thermometer
Dissolved Oxygen	Daily[d]	Oximeter DM-4P (Digimed, Brazil)
pH	Daily	pH meter DM20 (Digimed, Brazil)
Conductivity	Daily	Electrical conductivity meter DM32 (Digimed, Brazil)
Turbidity	Daily	Turbidimeter 2100N (Hach, USA)
Apparent colour	Daily	Colorimeter DM-COR (Digimed, Brazil)
Absorbance at 254 nm[a]	Daily	Nanocolor spectrophotometer (MN, Germany)
Total coliforms and *E. coli*	Twice a week	Membrane filtration (APHA, 2012 - 9222)
Absorbance at 254 nm[b]	Weekly	Nanocolor spectrophotometer (MN, Germany)
True colour	Weekly	Colorimeter DM-COR (Digimed, Brazil)
Particle size distribution	Weekly	Zetameter Zetasizer Nano Series ZS90 (Malvern Panalytical, UK)
Nitrite	Weekly	DR5000 spectrophotometer (Hach, USA)
Nitrate	Weekly	DR5000 spectrophotometer (Hach, USA)

[a] Absorbance at 254 nm was measured daily without the 0.45 µm membrane filtration step.

[b] Absorbance at 254 nm was measured weekly with the 0.45 µm membrane filtration step.

[c] Daily analyses were not performed on weekends and holidays.

[d] DO was measured daily until day 43 (first maintenance)

To analyse the extracellular polymeric substance from the s*chmutzdecke,* six samples of approximately 0.5 mL from the random zones of non-woven blanket surface and the first 5 mm of top sand bed were collected weekly using a cut-off syringe and stored at -20 °C for further analyses

of protein and carbohydrates content. A method described by Lubarsky et al. (2010) was used to extract these substances from sand samples. The protein and carbohydrate quantification were performed with the method described by Raunkjaer et al. (1994), and Dubois et al. (1956), respectively. Detailed methodology for extraction and quantification are presented in the Supplementary material.

At the end of the operation, *schmutzdecke* samples from the non-woven blanket and the first centimetres of sand were collected and analysed for suspended solids, according to Standard Methods (APHA, 2012) and Freitas et al. (2021). These samples were also concentrated by triple centrifugation (1000 ×*g*) and a drop of the concentrated sample was used for morphological identification of organisms by bright field microscopy (Olympus ® BX60, Japan).

2.4 Statistical analyses

For statistical analyses, the data normality was evaluated using the Shapiro-Wilk test. The Paired Student's *t* test was performed for normal dependent datasets and the Wilcoxon test was used for non-parametric dependent datasets. A 5% significance level (p-value < 0.05) was considered in both tests. Spearman's bivariate analyses were performed to correlate the performance of parameters.

3. Results and Discussion

3.1 HSSF filtration rate and tracer tests

The total filtration time in the constructed HSSF was 130 min and the total idle time was around 6h with the clean filter. According to the HSSF manual, the idle time should be between 1 and 48h, allowing the processes to act for bacterial removal (CAWST, 2012).

The average MFR was 323 mL/min (8.78 m^3/m^2.day) reached 20 min after the feeds. The tracer test showed an MDI of 2.40 and mMDI of 2.51, presenting a flow pattern closer to the ideal plug flow.

The tracer started coming out the filter after one feed, reaching a maximum concentration between the second and the third feed volumes, and the tracer concentration reaches zero between the fourth and fifth feed volumes (Figure S2 – Supplementary material). The steep increase and decrease observed in the tracer concentration during filtration are characteristic of a plug flow reactor (Figure S2).

3.2 Efficiency of the multi-barrier household system

3.2.1 Operational parameters

The multi-barrier household system was operated for 113 consecutive days, mostly during the winter and dry season in Brazil. The average water sample temperature was approximately 19 °C, with a variation between 11 to 28 °C. The accumulated precipitation volume, gathered from the Brazilian National Institute of Meteorology (INMET) throughout all days of operation was 60.6 mm (of which 43.4 mm was on the third day of operation).

Regarding the HSSF, the average MFR observed was 199 mL/min (5.41 $m^3/m^2.d$), with a maximum of 260 and minimum of 70 mL/min (7.06 – 1.90 $m^3/m^2.d$, respectively). HSSF maintenance was performed three times during operation after 41, 76 and 103 days.

3.2.2 System efficiency

The multi-barrier household system evaluated in this work included a pre-treatment, followed by an intermittent HSSF and a UV disinfection device. Overall, we observed an improvement of the water quality through the treatment process, according to the WHO Guidelines for drinking water (WHO, 2017). The data summary of each parameter evaluated is presented in Table 2.

3.2.2.1 Turbidity

The mean turbidity of the raw water was 12.36 NTU with only a few values higher than 20 NTU. The low turbidity levels on river water can be related to low precipitation during the operation. The average turbidity removal from the whole system was 73.3%, ranging from 32.9 to 94.4%. Figure 3 shows the turbidity values in raw and treated water and the percentage of removal in each stage during the operation.

The pre-treatment with the non-woven blanket was essential to retain major particles and reduce the turbidity to achieve recommended values for HSSF influent water for more restrictive drinking water standards (10 NTU) (Sabogal-Paz, 2020). This process was able to remove an average of 39.9% of turbidity, increasing efficiency during the days of operation due to biofilm development on the blanket. Only four samples (6%) of pre-treated water were above 10 NTU. This data was similar to previous studies that observed near 45% of turbidity removal by pre-treatment (Freitas et al., 2021; Terin et al., 2021).

The filtration through HSSF was the main step in which higher turbidity removal was achieved, reaching an average of 55.9%, ranging from 22.5 to 90.3%. Reported turbidity removal by HSSF has a high range of efficiency. In general, filtration can retain 75%-90% of the turbidity (Freitas et al., 2022; Jenkins et al., 2011; Murphy et al., 2010), but efficiency lower than 50% can be observed when the influent water has lower turbidity, such as groundwater and pre-treated water. (Freitas et al., 2022; Andreoli and Sabogal-Paz, 2020; Medeiros et al., 2020; Adeyemo et al., 2015).

After 16 days, turbidity in filtered water reached values below 5 NTU, as recommended by WHO for household water treatment (WHO, 2017), but values under 1 NTU, ideally suggested by the guideline, were rarely observed on the days before the HSSF maintenance (Figure 3). As expected, disturbance of the biofilm layer after maintenance slightly reduced the values of turbidity removal,

recovering after a few days, as also seen by other authors (Terin et al., 2021; Maciel and Sabogal-Paz, 2020; Singer et al., 2017; Ghebremichael et al., 2012).

Table 2. Mean (and standard deviation), minimum and maximum values of physical-chemical and microbiological parameters for each water sample in the multi-barrier treatment system, and the total removal observed during the operation.

Parameters evaluated	WHO limits[c]	Raw		Pre-treated		HSSF Filtered		UV disinfected		Total System Removal	
		Mean (SD)	Min Max	Mean (SD)	Min Max	Mean (SD)	Min Max	Mean (SD)	Min Max	Mean (SD)	Min Max
Turbidity (NTU)	< 5.0 (Ideally <1.0)	12.36 (7.95)	5.02 51.70	6.41 (2.14)	4.27 14.20	2.70 (2.14)	0.66 10.70	2.67 (2.14)	0.74 10.60	73.7 % (17.8)	32.9 % 94.4 %
Total Coliforms (CFU/100mL)	0	26,400 (36,400)	660 145,000	10,700 (14,500)	2,200 61,000	190 (246)	8 880	2 (1.74)	0 5	3.88 LRV[e] (0.60)	2.22 LRV 5.16 LRV
E. coli (CFU/100mL)	0	457 (425)	100 2,000	181 (107)	30 485	13.2 (20.8)	0 90	0.14 (0.45)	0 2	2.49 LRV (0.38)	1.81 LRV 3.30 LRV
Temperature (°C)	-	18.7 (4.20)	24.5 13.0	19.5 (3.80)	11.5 28.0	19.5 (3.90)	11.0 28.0	19.3 (3.8)	11.0 27.0	ND	ND
Dissolved oxygen (mg/L)	-	6.2 (1.1)	2.6 7.6	7.5 (0.6)	6.0 8.4	7.8 (0.6)	6.8 9.0	8.0 (0.7)	6.9 9.3	- 39.5 % (46.5)	5.3 % - 200.0 %
pH	6.5 – 8.5	6.64 (0.07)	6.38 6.74	6.85 (0.05)	6.73 6.95	6.97 (0.05)	6.85 7.08	7.06 (0.05)	6.97 7.16	- 6.6 % (1.3)	- 4.8 % - 11.1 %
Conductivity (µS/cm²)	-	45.72 (4.09)	38.76 58.19	45.41 (3.35)	39.58 54.34	45.89 (3.51)	39.75 53.51	45.64 (3.22)	39.71 51.75	- 0.3 % (5.7)	- 20.0 % 11.5 %
Abs 254nm[a]	-	0.142 (0.041)	0.067 0.243	0.108 (0.025)	0.038 0.170	0.060 (0.024)	0.036 0.142	0.059 (0.024)	0.037 0.139	56.6 % (15.4)	22.1 % 78.7 %
Abs 254nm[b]	-	0.055 (0.020)	0.021 0.094	0.053 (0.020)	0.013 0.085	0.044 (0.017)	0.008 0.080	0.042 (0.016)	0.009 0.081	22.6 % (20.0)	- 14.7 % 57.1 %
Nitrite (mg/L)	3.0	0.018 (0.005)	0.013 0.030	0.019 (0.004)	0.015 0.028	0.016 (0.003)	0.012 0.021	0.017 (0.003)	0.014 0.024	2.2 % (20.0)	- 42.9 % 30.4 %
Nitrate (mg/L)	50.0	1.3 (0.6)	0.7 2.6	1.6 (0.3)	1.0 2.2	1.7 (0.4)	1.1 2.6	1.8 (0.4)	1.1 2.4	- 61.1 % (67.2)	- 30.8 % 214.3 %
Apparent colour (mg Pt Co/L)	-	53.4 (18.6)	28.5 123.0	36.2 (8.4)	23.2 58.2	16.5 (8.9)	5.9 46.5	16.6 (8.9)	6.2 46.6	65.8 % (16.9)	26.8 % 91.2 %
True colour (mg Pt Co/L)	15.0	12.6 (4.3)	4.5 20.6	12.1 (4.3)	5.3 20.5	9.2 (2.9)	5.4 17.4	9.3 (2.9)	5.7 17.3	18.6 % (26.7)	- 37.8 % 59.7 %
Particle size (nm)	-	358.5 (73.7)	273.8 564.3	326.0 (33.6)	265.1 368.3	265.0 (25.5)	224.4 306.6	263.2 (25.6)	216.0 299.1	ND	ND
Transmittance (%)	-	ND[d]	ND	ND	ND	87.5 (3.9)	72.9 92.6	87.5 (3.9)	73.1 92.2	ND	ND

[a] Absorbance measured without filtration;
[b] Absorbance measured after filtration;
[c] WHO Guidelines for drinking water (WHO, 2017);
[d] Not determined (ND);
[e] Log Removal Value (LRV)

Table 3. Spearman's bivariate analysis between turbidity and microbiological parameters of the filtered water, and HSSF operational settings.

Parameters		Turbidity		Total Coliforms (TC)		*E. coli*	
		Effluent (NTU)	Removal (%)	Effluent (CFU/100mL)	Removal (%)	Effluent (CFU/100mL)	Removal (%)
Time	p-value	**< 0.01**	**< 0.01**	0.205	0.224	**< 0.01**	**< 0.01**
(days)	|r|	**0.799**	**0.825**	0.247	0.237	**0.512**	**0.559**
Ripening time	p-value	**< 0.01**	**0.034**	**< 0.01**	**0.048**	0.203	0.177
(days)	|r|	**0.295**	**0.287**	**0.585**	**0.376**	0.248	0.263
MFR	p-value	**0.016**	**0.041**	0.206	0.904	0.594	0.645
($m^3/m^2.d$)	|r|	**0.397**	**0.342**	0.394	0.039	0.171	0.149
$\Delta h\ L^{-1}$ bed top	p-value	0.059	0.101	**0.043**	0.137	0.568	0.843
(0-2cm)	|r|	0.309	0.270	**0.567**	0.439	0.175	0.061
$\Delta h\ L^{-1}$ whole bed	p-value	**0.013**	**< 0.01**	0.105	0.070	0.067	0.323
(0-52cm)	|r|	**0.400**	**0.488**	0.470	0.518	0.522	0.298
Influent turbidity	p-value	**< 0.01**	**0.035**	0.077	0.111	**0.021**	0.120
(NTU)	|r|	**0.461**	**0.286**	0.339	0.308	**0.436**	0.300
Influent TC	p-value	**< 0.01**	**< 0.01**	0.539	0.323	0.717	0.202
(CFU/100mL)	|r|	**0.495**	**0.527**	0.121	0.194	0.072	0.249
Influent *E. coli*	p-value	0.106	0.172	**< 0.01**	**0.012**	**< 0.01**	0.563
(CFU/100mL)	|r|	0.312	0.266	**0.510**	**0.470**	**0.618**	0.114

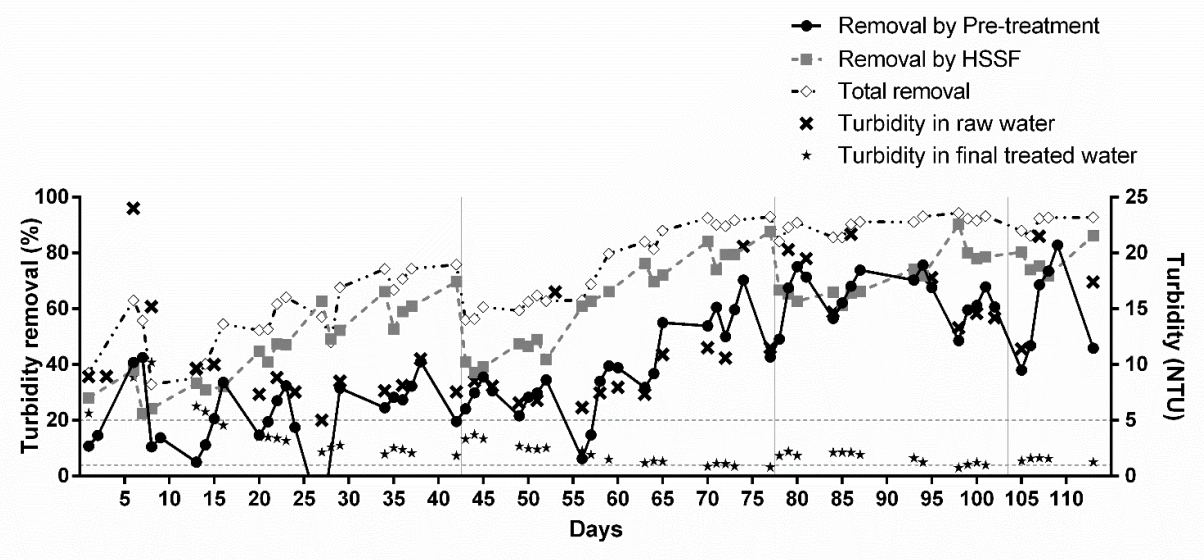

Figure 3. Percentage of total turbidity removal by the whole system and by the pre-treatment and the HSSF filtration, represented on the left axis. Turbidity in raw water and remaining after treatment, represented on the right axis. Vertical lines mark the days of HSSF maintenance. Horizontal dotted lines indicate the recommended (5 NTU) and ideal (1 NTU) turbidity values for household water treatment, according to the WHO (2017).

Spearman's bivariate analysis (Table 3) showed that the remaining turbidity and percentage of removal by HSSF were strongly correlated to the days of operation. Likewise, they correlate with the HSSF ripening time and the MFR, indicating that the *schmutzdecke* development and the reduction of filtration rate during HSSF operation, due to clogging and reduced pore size, are able to retain more particles, improving the turbidity removal, decreasing the residual turbidity. This fact was also observed by several authors, suggesting that the biofilm development can increase the turbidity removal, without presenting a direct relation to the influent water turbidity (Terin et al., 2021; Maciel and Sabogal-Paz, 2020; Adeyemo et al., 2015; Nair et al., 2014; Elliott et al., 2008).

In the present work, influent water quality also affects the turbidity removal (Table 3). When higher values of turbidity and total coliforms are present in the pre-treated water, a greater percentage of turbidity removal in HSSF was observed. Although some authors did not observe this correlation (Maciel and Sabogal-Paz, 2020; Napotnik et al., 2017), others have reported the importance of the influent water quality to the *schmutzdecke* development, and consequently to turbidity removal (Freitas et al., 2021; Terin et al., 2021; Napotnik et al., 2020; Kennedy et al., 2013).

Moreover, the correlation with head loss in the whole filtration bed suggests that the removal of particles can occur along the HSSF, and not only on the top layer (Table 3). Maciel and Sabogal-Paz (2020) showed a similar correlation, however they also detected a relation with the turbidity removal and the head loss in the top bed (2 cm), which was not observed in the current study.

3.2.2.2 Organic matter

The presence of organic content in water can be inferred by the measurement of absorbance at 254 nm and colour, and a slight decrease of these parameters (Table 2) was observed, indicating organic matter retention, mostly during HSSF filtration. In general, HSSF has reduced the capacity to retain dissolved organic contents, such as humic substances, presenting low removal rate of true colour and UV-absorbance values (Freitas et al., 2022). The present work observed a reduction by HSSF of 5% and 15%, respectively. Although a higher reduction of these parameters by HSSF can be found (Freitas, et al., 2021; Terin et al., 2021; Lynn et al., 2013), the authors usually do not find a greater reduction of total organic content. Reports reveal that the total organic content removal by HSSF can vary between 2 and 30% (Freitas et al., 2022; Andreoli and Sabogal-Paz, 2020; Lynn et al., 2013).

3.2.2.3 Other physical-chemical parameters

A significant increase of dissolved oxygen was detected through pre-treatment and UV disinfection (Table 2). It is speculated that the water pumping between reservoirs and through the Mesita Azul® may cause this increment in dissolved oxygen. Other works that used similar pre-treatment did not observe this variation (Freitas et al., 2021; Terin et al., 2021).

In general, pH slightly raised during all the filtration steps. This could be explained by the ions leaching from the filter media. The pH increase in HSSF was also reported by several authors (Freitas et al., 2021; Baig et al., 2011; Murphy et al., 2010; Ngai et al., 2007). Further investigations could be performed to identify better relations between filtration and the rise in pH.

Regarding the nitrate concentration, although a rise was observed, mainly in the pre-treatment stage (Table 2), the values are still far below the maximum recommended by the WHO (50 mg/L) (WHO, 2017). It is known that the nitrification process can occur in biofilm layers and that the increase and decrease of nitrate concentrations are reported in the HSSF (Freitas et al., 2022).

Particle removal by HSSF also improved the water UV transmittance, which is essential for better disinfection. The average transmittance of filtered water was 87.5, with a minimum of 72.9 at the beginning of the operation, increasing throughout the days, reaching a maximum value of 92.6.

3.2.2.4 Microbiological parameters

Total coliforms (TC) and *E. coli* were quantified twice a week from all stages along the system operation. Average, maximum and minimum values of bacterial concentration are presented in Table 2. Figure 4 shows the concentration of these microorganisms in raw and treated water and the removal in each treatment stage. The detection limit of the applied methodology was 1 CFU/100mL.

The whole system achieved a mean removal of 3.88 and 2.49 $\log_{10}$ of TC and *E. coli*, respectively (Table 2). Higher removal rates in the system were obtained when higher concentrations of bacteria were quantified in the raw water. Therefore, higher removal rates could be observed here if river water presented higher bacteria concentration. Although it is not the main purpose, pre-treatment was able to remove an average of 0.32 $\log_{10}$ of both microorganisms, which were probably adsorbed to major particles. Similar results for pre-treatment were observed by Freitas et al. (2021) and Terin et al. (2021).

A higher removal rate was generally observed after HSSF, with means of 1.88 $\log_{10}$ for TC and 1.40 $\log_{10}$ for *E. coli*. Mostly studies with HSSF showed bacterial removal between 1 and 2 $\log_{10}$, although higher values were also reported. Differences could be explained by the filter configuration, such as sand size, flow rate and influent water quality (Freitas et al., 2022).

Spearman's bivariate analysis (Table 3) demonstrated that the days of operation were strongly correlated to the concentration and removal of *E. coli*, but not for TC. Nonetheless, the ripening time

and the head loss on the top filter bed (first 2 cm) were correlated to the effluent TC concentration, but not for *E. coli*. This suggests that the *schmutzdecke* development may be responsible for retaining bacteria, mostly in the top layer of HSSF.

Several reports have shown that removal was improved after the ripening period, retaining more bacteria when the *schmutzdecke* was established (Maciel and Sabogal-Paz, 2020; Nair et al., 2014; Kennedy et al., 2013; Baig et al., 2011). Moreover, some studies have demonstrated that higher bacteria removal occurs on the first layer (5-10 cm) of HSSF, mainly when the biofilm is fully developed (Freitas et al., 2021; Young-Rojanschi and Madramootoo, 2014; Nair et al., 2014).

As expected, after HSSF maintenance, the values of bacterial removal were somewhat lower, recovering after a few days. There are some studies with similar results, indicating lower bacterial reduction after cleaning (Souza Freitas and Sabogal-Paz, 2020; Kennedy et al., 2013). Singer et al. (2017) demonstrated a decline of 0.16 to 0.83 $\log_{10}$ of removal after cleaning. A correlation of TC removal and ripening time (Table 3) shows an increase in the removal rate after some days of maintenance. A similar correlation was observed between the time after cleaning and *E. coli* removal (Maciel and Sabogal-Paz, 2020).

Similarly, as seen for turbidity, the influent water quality impacts the bacterial concentration in the effluent water. When higher values of turbidity and *E. coli* were present in the pre-treated water, higher concentrations of both bacteria were found after HSSF. This could be explained by the bacteria adsorption to colloidal particles remaining after filtration. However, usually higher values of water turbidity increase the bacterial removal (Jenkins et al., 2011).

Furthermore, the water residence time in HSSF can also be a factor on bacteria removal, which allows predation by other organisms and natural die-off (Freitas et al., 2022). The initial idle time in

the present work was 6 h, reducing throughout the operation time. Although longer idle time (> 15 h) could improve bacterial removal (Ghebremichael et al., 2012; Jenkins et al., 2011), the tested period was considered optimal for practical issues for a family that uses 48 L of water per day.

UV disinfection through Mesita Azul® mostly inactivated the remaining bacteria after HSSF. The average removal of TC and *E. coli* were 1.67 and 0.77 $\log_{10}$, respectively. The mean concentration of TC at the end of treatment was 2.0 CFU/100 mL, and a maximum of 5.0 CFU/100 mL. *E. coli* was detected in 10.7% of samples (3/28) at the final reservoir always below 2.0 CFU/100 mL.

As mentioned, higher bacterial reduction could be expected with higher contamination in river water. Correlation analysis demonstrated that when higher concentrations of TC and *E. coli* were in filtered water, higher disinfection values were observed. Interestingly, when lower removal rates were detected in HSSF, higher removal was observed in UV disinfection (Figure 4), which proves that the Mesita Azul® post-treatment can ensure higher security for drinking water even on vulnerable occasions of the operating system. Moreover, although better disinfection rates were obtained with higher transmittance, which was expected, a positive relation was observed between turbidity in filtered water and bacterial disinfection. This is probably explained by the higher presence of bacteria when influent turbidity is higher.

According to WHO, Mesita Azul® is effective against bacteria and protozoa in non-turbidity water, achieving > 5 $\log_{10}$ of *E. coli* (WHO, 2019b). This device was able to reduce the risk of contamination by *E. coli* in low-income communities (Reygadas et al., 2015; 2018). Although in the present work the average inactivation rate was inferior than that presented by WHO, because of lower bacterial concentration on influent water, the remaining *E. coli* from HSSF was totally inactivated in most samples, reducing the contamination risk.

After passing through Mesita Azul, ® few samples were detected with *E. coli* and in most samples TC was identified at low concentrations (< 5.0 CFU/100mL). The remaining bacteria after UV disinfection could be explained by some factors: (a) inefficacy of UV inactivation due to some residual water turbidity and colour, although the UV-transmittance was high and the Mesita Azul® proved to be effective for bacteria; (b) contamination in the reservoir, since the final reservoir was not cleaned after discharging the treated water; (c) and more probably due to the tailing effect. The final stage of a UV inactivation curve is the tailing phase, in which higher UV resistance is observed due to photoreactivation and the presence of more protective components, such as suspended particles, demanding higher doses to full inactivation (Sastry et al., 2011).

Aiming to ensure full inactivation of bacteria at the end of the treatment, a higher UV dose needs to be applied. This could be achieved by increasing the lamp efficacy above 15W or reducing the flow rate. A combination of UV and chlorine in low concentration could also be an alternative to inactivate any remaining microorganism. Furthermore, new reactors with LED lamps may be a more suitable option in the future, increasing durability, with adjustable effectiveness and reducing toxic waste.

The use of UV as a HSSF post-treatment has been rarely investigated. Frank et al. (2014) observed *E. coli* inactivation higher than 2.00 $\log_{10}$. The authors placed the HSSF filtered water in bottles under handheld UV lamps in room temperature to achieve a dose of approximately 180 mJ/cm^2. To the best of our knowledge, the present study is the first to evaluate UV technology directly combined with the HSSF, assessing the effectiveness continuously.

After the operation, microbial regrowth was assessed for 15 days in the final reservoir. However, an increase in TC or *E. coli* concentration during these days was not observed, indicating a lack of recontamination or cell multiplication after treatment and disinfection.

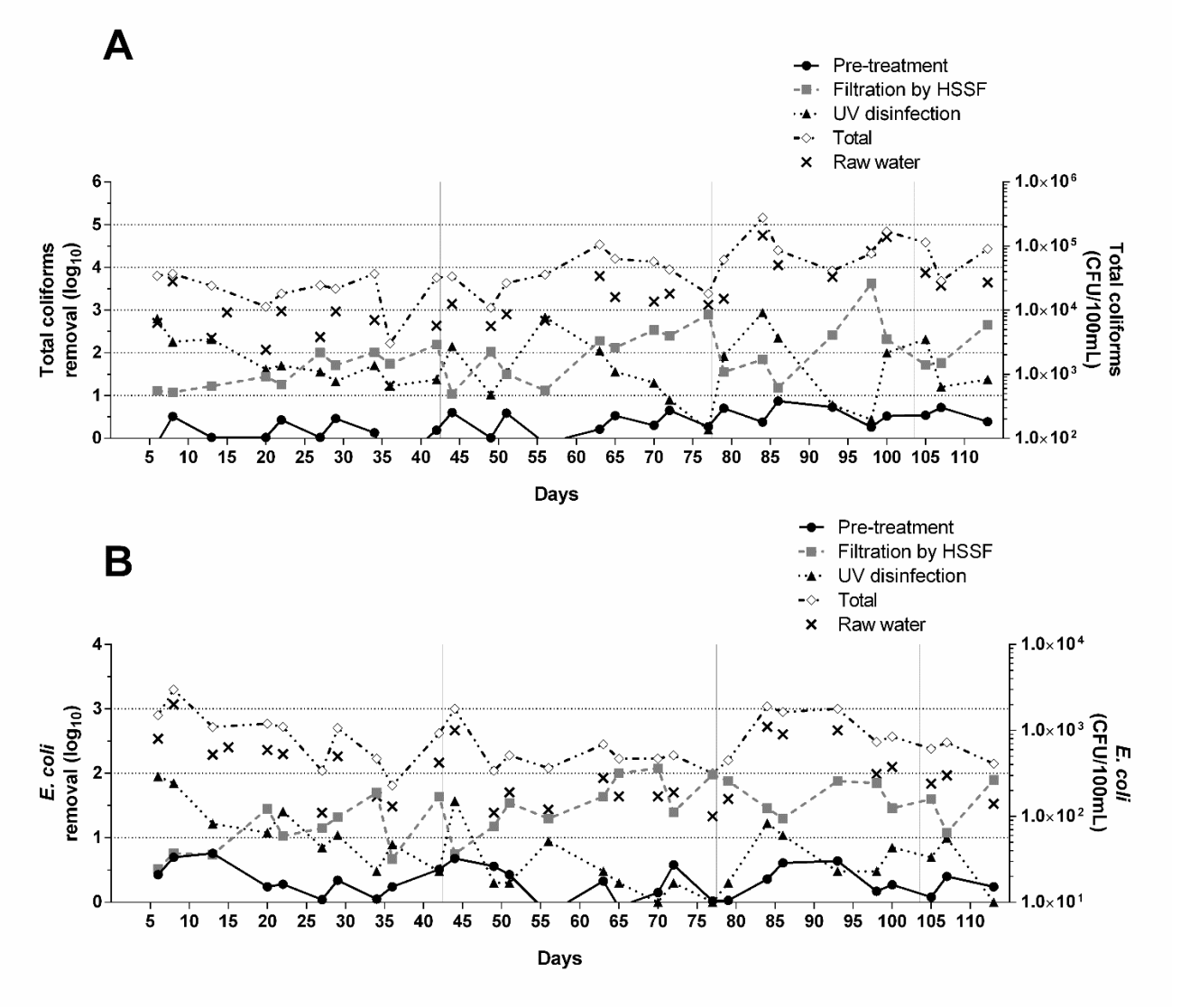

Figure 4. Log_{10} removal of total coliforms (A) and *E. coli* (B) by the whole system and by the pre-treatment, the HSSF filtration and UV disinfection, represented on the left axis. Initial concentration of microorganisms in raw water, represented on the right axis. Vertical lines mark the days of HSSF maintenance.

3.2.3 *Schmutzdecke* characterisation

The characterisation of the HSSF biofilm layer was performed weekly by quantification of protein and carbohydrate from the blanket and the first centimetres of top sand. Moreover, at the end of operation, the total solids and total suspended solids were assessed and a microscopical analysis was conducted.

The total solids analysis revealed a higher concentration of volatile solids on both the blanket (0.15mg/mL) and sand (0.14 mg/mL), representing 73% and 90% of the total solids, respectively. However, when assessing the suspended solids, greater inorganic material was observed on the blanket (62%), but not on the sand (37%), where the volatile suspend solids was higher. These results are presented in Table S3 (Supplementary material). Some authors presented different results, ranging from mostly containing inorganic material in the sand and in the blanket, but also with a higher concentration of organic material in the blanket compared to the sand, depending on some operational parameters. The higher prevalence of volatile solids indicates the predominance of organic content on the biological layer, developed during HSSF operation (Pompei et al., 2017).

Regarding extracellular polymeric substances, the average carbohydrate concentration on the blanket ranged from 16 to 52 mg/mL, slightly greater than on sand, which ranged from 11 to 44 mg/g. For protein, a higher concentration was observed on the sand, with an average ranging from 8 to 39 mg/g, while on the blanket the values were very low (0 – 9 mg/mL). These results are shown in Table S4 and Figure S3 (Supplementary material). In general, an increase in protein and carbohydrate concentration was observed over time and a reduction was observed in the week after HSSF maintenance.

Although the protein in biofilm may be predominantly associated with bacteria secretion (Flemming and Wingender, 2001) and carbohydrates related to algae presence (Underwood and Paterson, 2003), this relation is not exclusive and a discrepancy can be found depending on species variation and other organisms present in the s*chmutzdecke* (Xiao and Zheng, 2016; Lubarsky et al., 2010). The present work showed a higher accumulation of carbohydrates over protein, however since the system operated in the dry season it was not expected to find a high incidence of algae. Regarding the protein concentration, the pore size of blanket may allow higher retaining of algae while bacteria

can pass through and be trapped on the sand, which can explain the higher concentration of proteins on the sand.

In the microscopical examination, a low number and diversity of organisms were observed in the samples, but higher on the blanket than on the sand (Table S5 - Supplementary material). The low number and diversity could be explained by the low precipitation during operation and the HSSF maintenance performed only 2 weeks before the sample collection. Among the organisms examined, algae were the predominant group on the sand and blanket, but protozoa, rotifers and microcrustacean were also observed. Authors that used similar water sources also detected similar biological communities in the s*chmutzdecke* (Freitas et al., 2021; Terin et al., 2021; Andreoli and Sabogal-Paz, 2020; Medeiros et al., 2020).

Although not observable in microscopy, bacteria should be the main organism composing the biofilm layer (Pompei et al., 2017). The presence of other organisms has an important role on the s*chmutzdecke* development (Freitas et al., 2022). Algae assist on the colonization of the microenvironment and, as a source of the food chain (Nakamoto et al., 2014), can actively contribute to bacterial reduction by excreting substances that are toxic (Ribalet et al., 2008; Wichard et al., 2005). Moreover, some protozoa and rotifers can prey bacteria and other microorganisms (Freitas et al., 2022; Guchi, 2015).

The *schmutzdecke* characterisation showed its growth over the weeks, both in the blanket and the sand mostly with the rise of organic content and the presence of carbohydrates and proteins. Therefore, better particles and bacteria retaining in HSSF may be related to the development of the biofilm layer. The non-woven blanket in the HSSF acted as a support medium for *schmutzdecke*, offering a more robust biofilm layer, which seems to impact the filtration efficiency (Freitas et al.,

2021). Moreover, the biological community observed could be responsible for other factors that improve water quality, such as bacteria predation.

Although *schmutzdecke* characterisation demonstrated some relationship with HSSF efficiency, further studies are needed to better understand the microbial community. Some studies with genetic sequencing were conducted with full- and lab-scale slow sand filters (Haig et al., 2014; 2015). The authors showed a higher predominance of *Proteobacteria* and *Bacteriodetes* groups, with temporal change in the bacterial community compositions during filter ageing. Moreover, *Acidovorax, Halomonas, Sphingobium*, and *Sphingomonas* were found to be key genera associated with filter performance (Haig et al., 2015). As microbial community can vary according to the influent raw water, a metagenomic study could be performed to provide more detailed data.

4. Conclusions

- The multi-barrier household water treatment evaluated was able to improve the surface water to become drinkable water. The turbidity was effectively reduced to standards established by the WHO, and the additional physical-chemical parameters were also in accordance with the guideline.
- In general, more than 99.9% of the total coliforms were removed/inactivated by the system, remaining at very low concentrations, probably due to the tailing effect of the UV inactivation curve. *E. coli* was practically totally removed, reducing possible contamination risks of drinking water.
- The pre-treatment was essential to retain major particles and ensures better influent water for HSSF. The slow sand filtration was crucial to remove most turbidity and bacteria. The remaining bacteria after HSSF was mostly inactivated by the Mesita Azul® UV device.
- *Schmutzdecke* development seems to play an important role in the improvement of particles and bacteria retention in HSSF.

- Although the evaluated treatment is a low-cost household system with a capacity to produce 48 L per day, the filter may be somewhat hard to operate due to the several steps and water pumping processes. Moreover, communities with a lack of electric energy were not eligible for this kind of system.

5. Supplementary Material

Filter media characteristics, tracer test results, protein and carbohydrates method and microorganisms found in the HSSF are in the supplementary material.

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SUPPLEMENTARY MATERIAL

Efficiency of a multi-barrier household system for surface water treatment combining a household slow sand filter to a Mesita Azul® ultraviolet disinfection device

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The Mesita Azul® is a flow-through UV device with a cylindrical stainless-steel chamber of 41x11 cm and capacity of 2 L, approximately, integrated with a low-pressure UV lamp (254 nm) of 15W. The UV irradiates over an area of 410 cm² and a water layer of 4 cm, approximately. Figure S1 shows the configuration of the UV reactor from Mesita Azul® device in the system and an operation scheme.

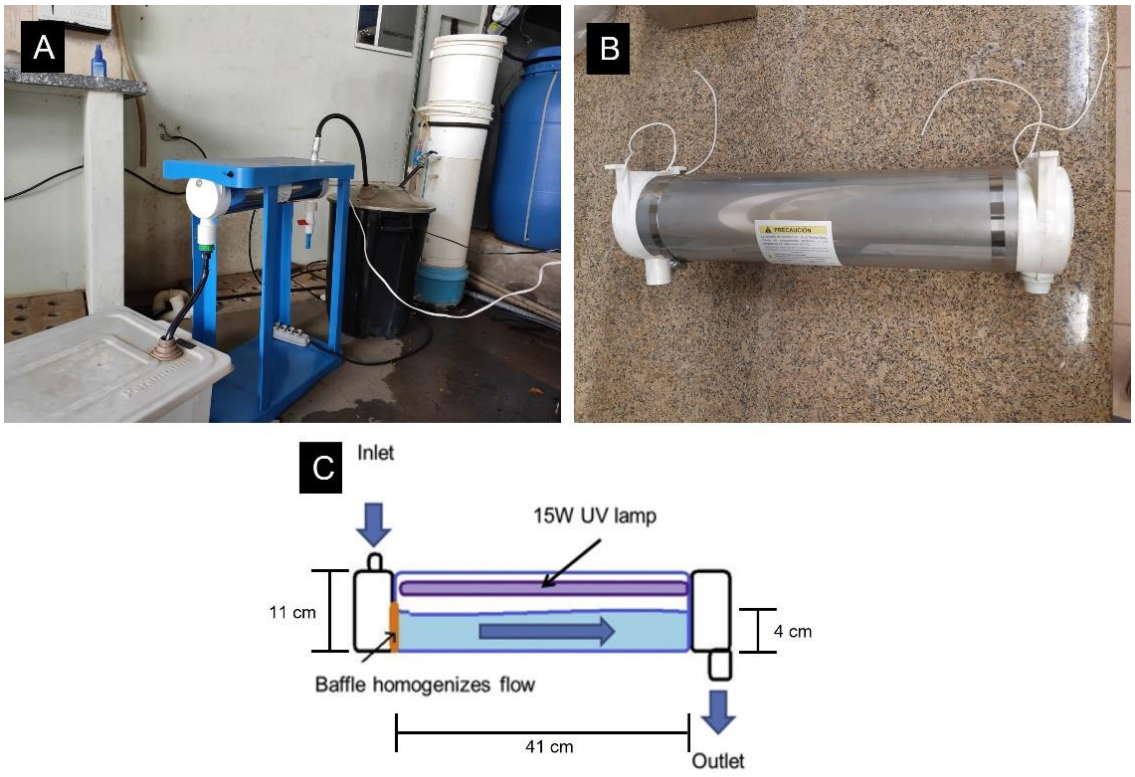

Figure S1. (A) Mesita Azul® configuration in the household system; (B) Mesita Azul® UV chamber; (C) scheme of UV reactor from Mesita Azul®, showing approximate dimensions, water inlet and outlet, and the UV low-pressure lamp of 15W above the water layer (Adapted from: Reygadas et al. 2015).

Table S1 shows the characterization of each sands and gravels used for the household slow sand filter construction, such as effective size, uniformity coefficient, specific mass, porosity and voids ratio.

Table S1. Characterization of sands and gravels used for HSSF construction

Material	Effective size (d_{10}) (mm)	Uniformity coefficient (UC)	Specific mass (g cm^3)	Voids ratio	Porosity (%)
Fine sand (filtration layer)	0.18	1.56	2.65	0.67	36.7
Coarse sand (separation layer)	1.00	1.52	2.63	0.77	41.3

Fine gravel (support layer)	4.76	0.8	2.65	-[a]	-
Coarse gravel (support layer)	6.43	1.62	2.64	-	-

[a] Gravels particle sizes were not suitable for voids ratio quantification by the used methodology.

Table S2 presents the characteristics of each layer from HSSF (filtration, separation and support layers) and the volume occupied by water, used for calculation of the charging volume of 16 L.

Table S2. Calculated HSSF charging volume

HSSF's parts	Area (m^2)	Thickness (m)	Volume (m^3)	Volume occupied by water (L)
Filtration layer	0.053	0.5	0.0265	9.73
Separation layer	0.053	0.05	0.00265	1.09
Support layer	0.053	0.125	0.00663	2.53
Stead water	0.053	0.05	0.00265	2.65
Inner exit tubing	0.00071	0.4	0.00028	0.28
Total				**15.72[a]**

[a] The charging volume was roundup to 16.0 L.

Methodology of protein and carbohydrates quantification

This section briefly describes the methodology used for quantification of protein and carbohydrates from the blanket surface and top sand of HSSF.

During the operation of the HSSF in the system, sampling was performed weekly. Six samples of approximately 0.5 mL from the random zones of non-woven blanket surfance and the first 5 mm of top sand bed were collected using a cut-off syringe and stored at -20 °C for

further analyses. For the analysis, three samples from the blanket and three from the sand of each week were randomly selected, and triplicates were performed for each one.

For sand samples extracellular polymeric substances were extracted using a methodology described by Lubarsky et al. (2010). The sand samples were placed in 2 mL tubes and mixed with distilled water for 90 min at 20 RPM. After mix and sedimentation, the supernatant containing the colloidal EPS fraction was collected for quantification of protein and carbohydrates. The remaining sand was dried overnight in an incubator at 37 °C then weighed it order to determine the mass of carbohydrate and protein per mass of dry sand (mg g^{-1}).

For the surface blanket samples, no extraction method was necessary. The concentration of carbohydrate and protein were given in terms of mass per volume of sample (mg mL^{-1}).

For carbohydrate quantification it was used the phenol methodology (Dubois et al., 1956). In 200 µL of samples supernatant were added 200 µL phenol (5%) and 1 mL sulphuric acid (98 %) then incubated 35 min at 30 °C. Following, the absorbance at 488 nm was measured in spectrophotometer (Nanocolor spectrophotometer, MN, Germany). A standard curve was prepared with D-glucose (Sigma-Aldrich) ranging from 0 to 200 mg/mL.

For protein quantification, it was used a modified Lowry method (Gerbersdorf et al., 2008; Raunkjaer et al., 1994). In 250 µL of samples supernatant were added 250 µL of 2% sodium dodecyl sulphate salt and 700 µL of 'chemical reagent 4' and incubated for 15 min at 30 °C, and subsequent adding 100 µL of with Folin Ciocalteu reagent, and incubated for 45 min at 30 °C. The absorbance at 750 nm was measured in spectrophotometer (Nanocolor spectrophotometer, MN, Germany). A standard curve was prepared with bovine serum albumin (Sigma-Aldrich) ranging from 0 to 200 mg/mL.

Figure S2 shows the curves of the tracer test performed with the HSSF, showing the tracer concentration on the charged volumes. The steep increase and decreases observed in the tracer concentration during filtration are characteristic of a plug flow reactor.

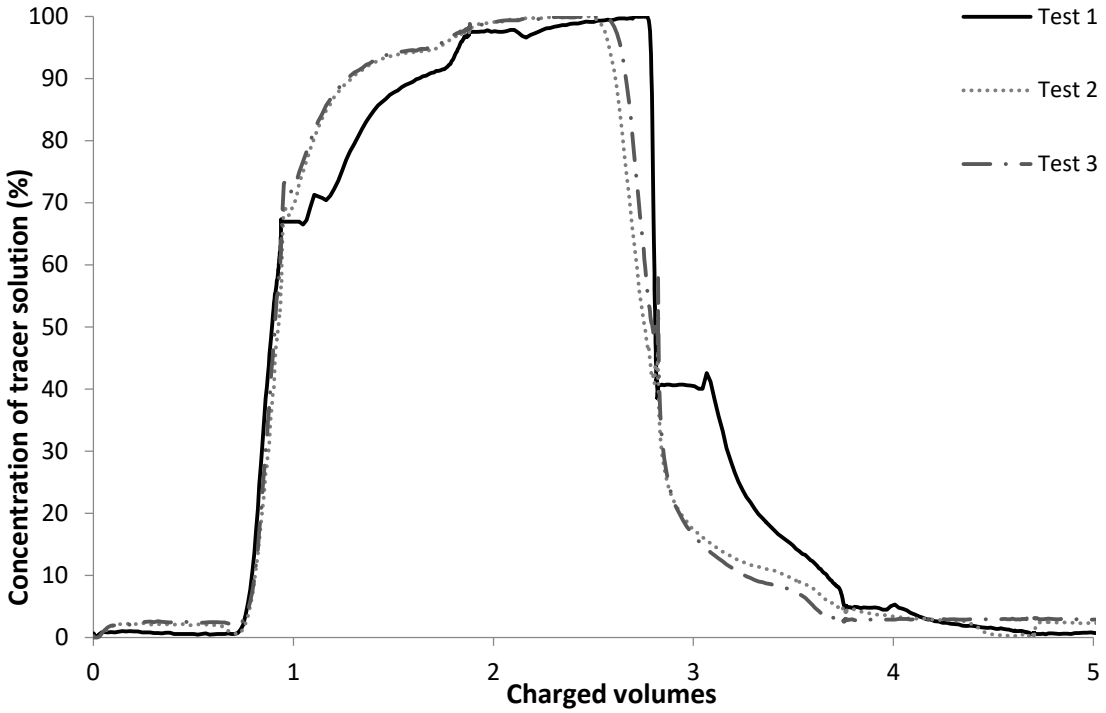

Figure S2. Curves from each test of the tracer concentration versus the charged volumes filtered.

Table S3 shows the values of total and suspended solids and the volatile solids content from samples of the blanket and sand obtained from HSSF after the end of system operation.

Table S3. Average concentration of total and suspended solids in the blanket and sand in HSSF (mg/mL)

Sample	Total solids	Total volatile solids	Total suspended solids	Volatile suspended solids
Blanket	0.2046	0.1508	0.1944	0.0744
Sand	0.1583	0.1433	0.1422	0.0900

Table S4 and Figure S2 shows the variation of protein and carbohydrate concentration in sand and blanket obtained weekly from HSSF during the system operation.

Table S4. Average concentrations of carbohydrates and proteins in sand and blanket of HSSF over the weeks of system operation. Coloured lines are measurements after filter maintenance.

Weeks	Carbohydrate		Protein	
	Sand (mg/g)	Blanket (mg/mL)	Sand (mg/g)	Blanket (mg/mL)
1	32.243	16.007	25.468	0.000
2	34.258	28.765	16.678	1.377
3	24.804	29.219	20.588	1.548
4	38.935	39.922	37.599	2.413
5	31.904	48.549	25.161	2.931
6	44.510	52.471	34.788	7.546
7	25.512	17.977	8.432	0.000
8	30.024	20.680	30.653	0.870
9	20.645	38.599	39.595	8.904
10	14.654	29.082	16.906	0.000

11	16.690	15.991	31.901	0.000
12	19.191	21.778	18.830	1.202
13	11.568	33.804	15.585	0.000
14	11.875	33.072	15.549	2.569
15	11.405	21.324	9.294	0.000
16	11.813	34.049	19.642	0.005

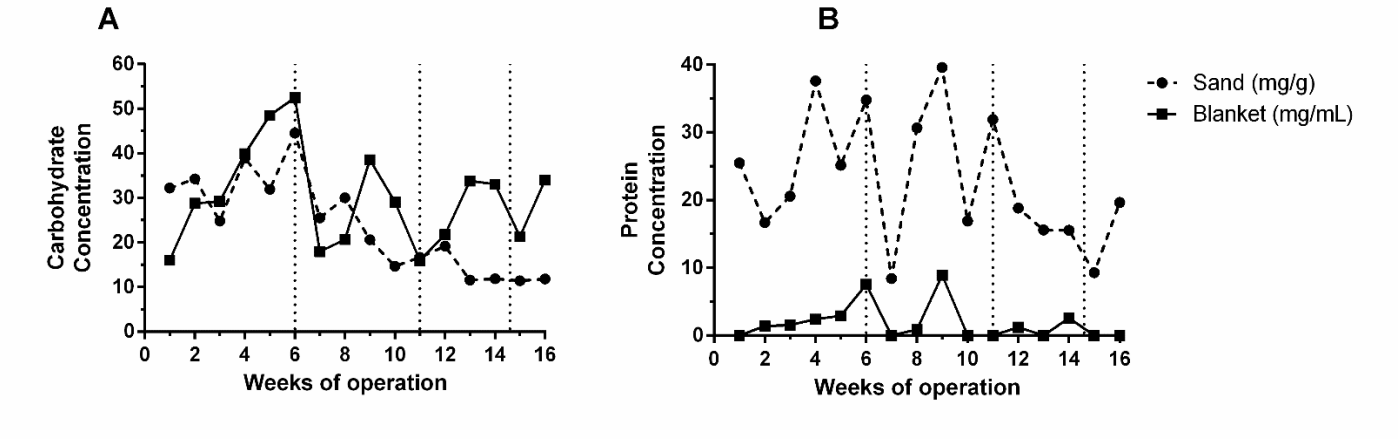

Figure S3. Average concentration of carbohydrate (A) and protein (B) in sand and blanket of HSSF over the weeks of system operation. Vertical lines mark the days of HSSF maintenance.

Table S5 shows the main group and some of the organisms identified in the microscopic analysis of the blanket and sand at the end of operation

Table S5. Groups and organisms identified on microscopic analysis of the blanket and sand of HSSF

Group	Organism	Blanket	Sand
Algae	*Aulacoseira* spp.	X	
	Chlorella spp.		X
	Chromulina spp.		X
	Cocconeis spp.		X
	Crucigeniella spp.		X
	Desmodesmus spp.	X	
	Diatoms	X	X
	Euglena spp.	X	
	Navicula spp.	X	X
	Nitzchia spp.		X
	Not identified	X	X
	Scenedesmus spp.	X	
	Selenastrum spp.	X	X
	Tetrastrum spp.	X	
	Trachelomonas spp.	X	X
	Westella spp.	X	X
Protozoa	*Balantidium coli*		X
	Colpidium spp.	X	X
	Not Identified	X	X
	Vorticela spp.		X
Animal	Gastrotricha	X	
	Rotifer	X	X

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