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Household slow sand filters in intermittent and continuous flows to treat water containing low mineral ion concentrations and Bisphenol A

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Household slow sand filter (HSSF) has been used as an alternative to drinking water treatment in rural communities worldwide; however, its performance to treat influent water with quality similar to rainwater still needs further studies. Rainwater presents low pH and slight mineral ion concentrations, an aspect that can modify the filter media and consequently the HSSF efficiency. Furthermore, house roofs used in rainwater harvesting can be made of plastic. Therefore, it can introduce chemicals such as Bisphenol A (BPA) in the water. In this context, two pilot-scale HSSFs operated in continuous and intermittent flows were evaluated to treat water containing BPA and low mineral ion concentrations in order to assess the filter performance. Filter media leaching was noticed in the trials; thus, filter media and construction material selection must be carefully evaluated to eliminate risks of pollutant occurrence in drinking water. Operational differences between continuous and intermittent flows influenced the HSSF efficiency for BPA and DOC removals; even so, the filters' performance was low probably due to the slow *schmutzdecke* development. According to tracer test results, HSSF can be classified as a plug flow reactor and strategies to improve its hydraulic performance are not required.

Keywords: biosand filter; decentralised treatment; drinking water; endocrine disruptor; rainwater

1. Introduction

Access to drinking water in rural communities is a problem because they usually have a regional diffuse distribution that limits technically and/or economically the interconnection with water supply networks. Therefore, they need a decentralised supply solution. Research for efficient, easy-to-implement, operate and maintain low-cost technological solutions are essential to the success of water projects in these overlooked communities. According to WHO (2012) until reliable, safe, and piped water is accessible to every household, temporary actions, such as household water treatment and safe storage (HWTS) are needed to reduce waterborne diseases. In this context, household slow sand filter (HSSF) has acquired importance worldwide due to its efficiency and simplicity (Cawst, 2012; Sobsey et al., 2008). Real-scale HSSF has been reported in 69 countries and there are more than 300,000 units in operation worldwide (Cawst, 2012).

1.1. HSSF basic concepts

The worldwide requirement for a low-cost HWTS, which is simple to maintain and has safe water production, led to the development of the household slow sand filter (HSSF) in the 1990s. HSSF is a small filter that can work in intermittent or continuous flows, making it appropriate for homes (Cawst, 2012; Terin and Sabogal-Paz, 2019; Young-Rojanschi and Madramootoo, 2014). HSSF is made of concrete or plastic and it is filled with layers of sand and gravel that are carefully prepared (Cawst, 2012). The development of the biological layer (*schmutzdecke*) on top of the fine sand is required to obtain the highest efficiency. HSSF has similar limitations to SSF when removing solids and organic compounds. High concentration of suspended material in the influent water obstructs the intergranular voids causing a reduction in the filter run and an increase in the frequency of cleaning (Souza Freitas and Sabogal-Paz, 2019). However, solids and organic compound removals are easily enhanced by using pre-treatment (e.g. coagulant dosage or sedimentation) and/or post-treatment (e.g. adsorption). Influent water quality and efficiency reported by some authors are shown in Table 1.

[Table 1 near here]

The maximum turbidity for HSSF is up to 50 NTU, according to Cawst (2012); however, for countries with more restrictive drinking water standards, this value must be reduced to 10 NTU.

1.2. HSSF in intermittent and continuous flows

HSSF is a modified SSF which works with a higher filtration rate (up to 29 times) and a smaller sand layer (up to 50% less) than the conventional filter. HSSF cleaning processes do not require removing the top of the filter media (Cawst, 2012) and it has reduced the scale, compatible with a household water treatment (WHO, 2016). A single user can build an HSSF with easily accessible materials (Faria Maciel and Sabogal-Paz, 2018) and it can operate with intermittent flow, an operational aspect not possible in conventional SSF. Furthermore, HSSF can improve its performance by installing a non-woven synthetic fabric on the top of the filter media (Faria Maciel and Sabogal-Paz, 2018), which can be easily positioned and fixed because the filter has a small superficial area, usually, up to 0.1 m^2.

HSSF in intermittent flow (I-HSSF) can operate with filtration rates up to 29 $m^3.m^{-2}day^{-1}$ (1.2 m/h), depending on the hydraulic head (Elliott et al., 2006). Water to be treated has to rest in the pores of the filter media for a period of 1 to 48 h (i.e. pause period) between each batch operation (Cawst, 2012). This pause period is important to allow physico-chemical and biological processes to act on the *schmutzdecke* to treat water. The pause period is a design parameter directly related to the HSSF efficiency and its establishment (1 to 48 h) is not yet fully understood. The user feeds the I-HSSF manually with 15-20 L directly into the unit after the pause period. The treated volume corresponds to the water that is retained in the filter media; consequently, a unit can usually produce up to 80 L day^{-1} according to the pause period adopted (Schmidt and Cairncross, 2009). The I-HSSF area occupied inside the residence is around 0.1 m^2.

HSSF in continuous flow (C-HSSF) usually works with lower filtration rates, up to 9.6 $m^3.m^{-2}day^{-1}$ (Faria Maciel and Sabogal-Paz, 2018). The filter can produce up to 200 L day^{-1} of filtered water, depending on filter configuration. C-HSSF can be fed by gravity (using an elevated tank) or by direct pumping. This filter needs a filtration rate control and may require more area inside the home (±1.0 m^2) as it demands an external supply unit (i.e. elevated tank or pump).

1.3. HSSF hydraulic behaviour

HSSF flow characterisation is an important operational parameter (e.g. it can define the water sampling time) and few studies have considered this aspect. Bradley et al. (2011), Elliott et al. (2008) and Lynn et al. (2013) have evaluated I-HSSF hydraulic behaviour and classified it as a plug flow reactor. The C-HSSF has been also classified as a plug flow reactor by Faria Maciel and Sabogal-Paz (2018), Terin and Sabogal-Paz (2019), and Young-Rojanschi and Madramootoo (2015). However, relatively little attention has been given to the hydrodynamics of these filters.

1.4. HSSF versus emerging contaminants

Various studies have been conducted on the application of SSF and HSSF for the removal of pharmaceutical and personal care products (PPCPs) and endocrine-disrupting chemicals (EDCs) from water and wastewater (D'Alessio et al., 2015; Haig et al., 2016; Katayama-Hirayama et al., 2010; Li et al., 2018; Pompei et al., 2017). These authors evaluated filtration rates between 0.02 and 4.8 m^3 $m^{-2}day^{-1}$ and the mean removal efficiencies were between 11 to 92% for the target compounds. Nevertheless, there has been relatively little understanding of the fundamental mechanisms operating during SSF.

1.5. Bisphenol A, risk and detection

Bisphenol A (BPA, CAS n. 80-05-7) was synthesised in 1905 from phenol and acetone and it is mainly used to generate polycarbonate and epoxy (95% of the production) and the rest (5%) is transformed into resins, antioxidants, fungicides, paints and can coating (Huang et al., 2012).

BPA is an endocrine disruptor; hence, it is an exogenous agent that interferes with the synthesis, production, secretion, release, transport, binding, action or elimination of natural hormones responsible for homeostasis, reproduction, development and behaviour (Kavlock et al., 1996; Zoeller et al., 2012).

It has been detected in drinking water and food and has been banned from plastic containers in Europe and Canada (Rogers et al., 2013). BPA in drinking water may arise from its contact with polycarbonate plastics and epoxy resins (FAO and WHO, 2011) or contaminated raw water. According to Vom Saal and Hughes (2005), 115 *in vivo* studies were published regarding the effects of low BPA dosages and 94 indicated significant effects. In addition, in 31 publications on vertebrates and invertebrates, endocrine changes were found with apparently safe dosages (<50 $\mu g\ kg^{-1} day^{-1}$). An estrogenic effect was confirmed by *in vitro* tests with disruption of cell function (Beausoleil et al., 2018; Vom Saal and Hughes, 2005). Finally, the above authors reported that there is a need to consider the health risk based on the scientific literature relating adverse effects on animals in dosages considered safe.

Regarding biological treatment, bacteria and fungi can degrade BPA (Kang et al., 2006) and this opens up space to treat water affected by endocrine disruptors by HSSF. However, BPA metabolites generated after treatment may have estrogenic effects (Huang et al., 2012; Kang et al., 2006).

BPA detection in environment matrixes has generated the development of chromatographic techniques. Methods based on high performance liquid chromatography (HPLC) have usually been used for BPA analyses (Rodriguez-Mozaz et al., 2004). HPLC may be impracticable in developing countries due to high cost and technical complexity. Therefore, simpler methods that can detect BPA are needed to assess the drinking water risk. From this perspective, UV absorbance of BPA can be measured with a spectrophotometer, allowing its quantification in ppm (Cao et al., 2014).

1.6. Rainwater harvesting and treatment

Rainwater is slightly acidic and has very low dissolved mineral concentration. Thus, it is relatively aggressive and it can dissolve metals and impurities from catchment and storage tanks, resulting in unacceptably high pollutant concentrations in the water (WHO, 2017). BPA may be present in plastic materials, pipes, fittings and tanks (Huang et al., 2012) that can be used for rainwater harvesting, an aspect that needs more studies. Slow sand filtration is a technology that may be used to treat rainwater in developing countries (Helmreich and Horn, 2009) and its performance should be better understood.

Bearing in mind the lack of research about the endocrine disruptor's removal from rainwater, this paper aims to investigate the potential of two pilot-scale HSSFs (operating in intermittent and continuous flow regimes) in the BPA removal from water containing low mineral ion concentrations.

2. Materials and Methods

2.1. HSSF characteristics

Two pilot-scale HSSFs were constructed in acrylic with a 98 mm inside diameter (cross sectional area = 0.0075 m^2). One HSSF was designed to operate intermittently (I-HSSF) and the other to operate continuously (C-HSSF). The filters were covered to protect them from light. HSSFs schemes can be found in Fig. 1.

[Figure 1 near here]

The HSSF filter media was a 55 cm fine sand layer (0.09 mm to 0.5 mm) with an effective size (D_{10}) of 0.18 mm and uniformity coefficient (UC) of 1.64. Fine sand used (CH52, Minerals Marketing, UK) presented the following chemical composition: SiO_2 = 97.3%, Fe_2O_3 = 0.1%, Al_2O_3 = 1.37%, K_2O = 0.83% and loss-on-ignition = 0.25%. Support media consisted of a 5 cm layer of coarse sand (1 to 3 mm), 5 cm layer of fine gravel (3 to 6 mm) and 7.5 cm layer of coarse gravel (10 to 12 mm). The average porosity of the filter materials was 32%. Fine sand and support media were washed in tap water prior to their introduction inside each unit. Acrylic columns were filled with tap water before inserting the filter media to avoid air pocket formation and to allow fine sand stratification as well.

Finally, a non-woven synthetic fabric (specific gravity: ± 0.2 g cm^{-3}, composition: 100% polyester, and thickness = 2.8 mm with 25 µm fibres) was positioned at the filter media top. After the HSSF assembling, deionised water continuously fed each filter by 24 h to remove the chlorine from the tap water.

Water from Regent's Park Lake (London, UK) was used as a ripening agent (i.e. agent to accelerate the filter maturation in a simple way) and was only added at the beginning of the HSSF operation. The filter volume (i.e. sum of standing water volume, outlet pipe volume and filter media and support layer pore volumes) was introduced twice to each HSFF (i.e. 2 L from Regent's Park Lake) and it was left for one day before starting off the operation with influent water. Regent's Park water quality comprised total coliforms of 1.8×10^4 CFU 100 mL^{-1}, *Escherichia coli* of 200 CFU 100 mL^{-1}, turbidity of 2.02 NTU, conductivity of 1158 µS m^{-1}, pH of 7.69, temperature of 23 ^oC, dissolved oxygen (DO) of 4.34 mg L^{-1} and dissolved organic carbon (DOC) of 19.7 mg L^{-1}. Filtered water samples were collected one day after the maturation process, when the filters started the operation with influent water, to assess the efficiency of the HSSFs.

HSSFs were cleaned when they reached the maximum hydraulic head. Maintenance consisted of removing the synthetic fabric, scraping off the top and draining the supernatant without removing the sand from the top. The fabric was washed in deionised water and it was then placed back on the filter.

2.2. HSSF operation

HSSFs were operated for 90 continuous days. Influent water was prepared weekly by diluting BPA (Alfa Aesar ®, 97%) stock solution in deionised water to simulate rainwater contaminated by endocrine disruptor (Table 2).

[Table 2 near here]

HSSF filtration rates were calculated considering a daily production of 2.9 ± 0.9 L for the C-HSSF and 2.6 ± 0.8 L for the I-HSSF. The flow rate in the C-HSSF was controlled by a peristaltic pump (Watson-Warlow, MHRE 100) producing a filtration rate of 0.38 ± 0.13 $m^3 m^{-2} day^{-1}$. The I-HSSF hydraulic head was variable generating a filtration rate between 0 to 21 $m^3 m^{-2} day^{-1}$. The I-HSSF was filled with 1.0 L (filter volume) three times per day by a submersible pump (Jeneca ®, HM 5063) controlled with a valve and timer, causing an 8-hour pause period.

2.3. Tracer tests

HSSF flow characterisation was carried out using 200 mg L^{-1} sodium chloride (NaCl) solution as a tracer, prepared with tap water (the tests were performed in triplicate). Electric conductivity variation in the filtered water was detected using a conductivity probe (*Vernier*, USA) situated in the outlet hose. Data was collected by *Logger Lite* software (*Vernier*, EUA) and it was processed by Excel 2013 (*Microsoft*, EUA) and Origin 8.6 (*OriginLab*, EUA). In each tracer test, the HSSFs were cleaned with tap water until the salt solution from the previous test was completely removed.

NaCl solution was applied to the C-HSSF as a step input and the probe allowed a correlating conductivity variation with tracer concentration. The filtration rate was kept on 0.5 $m^3\ m^{-2}\ day^{-1}$ and the hydraulic retention time (HRT) was determined. The flow pattern was adjusted into three hydrodynamic mathematical models: dispersion models (low and high dispersion) and N-continuous stirred tank reactors (N-CSTRs), as reported by Levenspiel (1999).

The first filling to the I-HSSF was carried out with NaCl solution and the subsequent feedings were with tap water. The filtration rate declined to zero when the hydraulic head reached the lowest level, at which time a new water charging was performed (V = 1.0 L). Salt concentration versus filter volume curves produced a positive step followed by a negative step (increased and decreased concentrations). Afterwards, the Morrill Dispersion Index (MDI) and the modified MDI (mMDI) were calculated as described by Tchobanoglous et al. (2003) and Lynn et al. (2013), respectively.

2.4. BPA detection

BPA was measured by UV-Vis spectrophotometer (Shimadzu UV 2600, Japan). UV absorbance for six BPA concentrations (0 to 12 mg L^{-1}) was measured from 200 to 1000 nm wavelengths, in triplicate, in order to identify the characteristic absorbance peak (it was detected at 224 nm). Afterwards, the BPA standard curve was made from data obtained at 224 nm. The relationship between UV absorbance and BPA concentration was established [UV absorbance = 0.0748 x BPA concentration (mg L^{-1})]. The calibrated curve showed r^2 of 0.94, detection limit of 0.03 mg L^{-1} and limit of quantification of 0.10 mg L^{-1}.

2.5. *Schmutzdecke* evaluation

Scanning electron microscope (SEM) and flow cytometry (FC) were used to evaluate the biological layer (*schmutzdecke*) at the end of the HSSF operating period.

SEM with energy dispersive x-ray spectroscopy (EDS) (JEOL JSM-6480LV, Japan) was used to capture photomicrographs and chemical compositions from synthetic fabric and fine sand of dried samples at room temperature. Samples were analysed at different magnifications, variable pressure analytical scanning electron microscope with secondary electron imaging (SEI) and backscattered electron imaging (BEI) detectors and with an accelerating voltage of 15 kV. Individual particles and compacted samples were rigidly mounted on a specimen stub and they were coated with an ultrathin gold layer. EDS did more than a hundred spot analyses.

Bacteria cells (alive and dead) were determined by flow cytometry using Guava® easyCyte 5HT Benchtop Flow Cytometer (Millipore, UK). Samples from the biological layer for I-HSSF and C-HSSF at the end of the filter operation were collected and stored at 4 ºC before processing. LIVE/DEAD BacLight Bacterial Viability kit (Thermo Fisher Scientific, UK), with propidium iodide dye and SYTO® 9 dye, was prepared and applied according to the manufacturer´s instructions. 20 µL of sample (*schmutzdecke*) and controls (*E. coli* strain K-12 and deionised water) were added to 180 µL of the prepared stock staining into 1.5 mL microcentrifuge tubes.

E. coli was diluted before measuring in the flow cytometer in filtered deionised water (0.22 mm; PTFE Syringe, Gilson scientific). It was used as a biological positive control, and filtered deionised water was utilised as a control for background fluorescence. All prepared samples were incubated at room temperature in the dark for 15 min. The bacteria acquisition gate was determined according to forward scatter (FSC) and side scatter (SSC) channels to eliminate background noise and debris.

2.6. Sample collection and analysis

Influent water and filtered water samples were collected and analysed daily, according to the water sampling time defined by the tracer tests. The water quality parameters analysed were turbidity (Hach 2100N, USA), DO (Jenway 9200, USA), conductivity, temperature and pH (Mettler Toledo, S47K, USA), DOC (TOC-L, Shimadzu, Japan), cations and anions (IC1100, Dionex, USA and Varian ICP-AES 720-ES, USA), and coliforms (m-ColiBlue24®, Hach, USA). Standard methods defined by APHA, AWWA and WEF (2012) were followed to evaluate the above parameters. Head loss was measured daily in both filters.

2.7. Statistical analyses

Statistical analyses were performed using PAST 3 software (PAlaeontological STatistics) created by Hammer et al. (2018). The Kruskal-Wallis test was used to compare data from the filtered water samples among each other and with influent water (95% confidence interval). When statistical analyses showed that the mean values were significantly different, the Mann-Whitney test was selected to define which sample was different from another (95% confidence interval).

3. Results and Discussion

3.1. Tracer tests

Tracer test results for the I-HSSF are shown in Fig. 2a. Tracer concentration increased from 0 mg L^{-1} up to 182 mg L^{-1} and this 9% difference relative to the initial concentration (200 mg L^{-1}) can be attributed to the filter's hydraulic head, which may have diluted the tracer solution (Terin and Sabogal-Paz, 2019).

[Figure 2 near here]

According to the results, two feedings were required before collecting samples for the I-HSSF performance evaluation. Salt concentration decreased from the third filter volume and after the fifth feeding, the tracer left the filter (Fig. 2a). Similar behaviour was described by Bradley et al. (2011), Faria Maciel and Sabogal-Paz (2018) and Terin and Sabogal-Paz (2019), characterising a plug flow reactor for HSSF.

I-HSSF MDI was 1.54 ± 0.01, lower than the one observed by Young-Rojanschi and Madramootoo (2015), who found an MDI value of 1.8 and slightly higher than the ones reported by Elliot et al. (2008) and Bradley et al. (2011) of 1.3 and 1.4, respectively. As stated by USEPA (1986) and Tchobanoglous et al. (2003), this MDI characterises the I-HSSF as a plug flow reactor (MDI up to 2).

I-HSSF mMDI was 0.95 ± 0.1, lower than the one found by Lynn et al. (2013), who reported values of 2.86 and 3.01. According to Lynn et al. (2013), the calculated mMDI did not change significantly over time, which was a phenomenon noticed in our study. Consequently, additional strategies to improve the I-HSSF hydraulic performance in comparison to the ideal plug-flow reactor are not required.

Tracer test results for the C-HSSF are shown in Fig. 2b and Table 3. HRT was 857 ± 21 min and it was used to determine the sample collection time. The N-CSTR model showed a better adjustment with an r^2 of 0.75 and N of 17. As indicated by Levenspiel (1999), a high N value also designates a plug flow reactor.

[Table 3 near here]

In the plug flow reactor, the fluid passes through the reactor (filter) with no mixing of earlier and later entering fluid (no overtaking). The necessary and sufficient condition for plug flow condition is that the residence time in the reactor must be the same for all elements of fluid (Levenspiel, 1999). In this context, a HSSF evaluated by Elliott et al. (2008) showed a minimal effect of dispersion by flow paths through the porous media, a result analogous to our study for both filters. Therefore, from the perspective of the biological layer development and microbial removal processes, the results suggest the same time is available for all portions of water that enter the HSSF, helping the water treatment.

3.2. HSSF operation

Filtered water quality and removal or variation rates are shown in Table 4. Turbidity removal showed a negative value for both filters (i.e. filtered water presented 74-76% higher turbidity) and there was no removal improvement over time (Fig. 3), contradicting the literature.

[Table 4 near here]

[Figure 3 near here]

Turbidity removal within the range of 70% to 96% in laboratory and field studies has been described worldwide with influent water turbidity up to 58 NTU (Cawst, 2012; Frank et al., 2014; Jenkins et al., 2011). However, according to Frank et al. (2014), HSSF generally has greater turbidity removal when influent levels are higher. This may explain the performance found in our study, since the influent water turbidity was only 0.37 ± 0.11 NTU (Table 2).

Another possible explanation for the increased filtered turbidity may be attributed to the filter media leaching. Thiry et al. (1988) reported this phenomenon, when the effect of groundwater in sands was analysed. This can be confirmed by the ion concentration increase in the filtered water for both filters (Table 4). It should be noted that the sands used in HSSF in real scale are washed only with water; therefore, it is not possible remove all the minerals prior to use. On the other hand, the HSSFs produced most of the time filtered water with turbidity below 1.0 NTU and this value is associated with 1-2 log and 2.5-3 log reduction of viruses and protozoa, respectively (WHO, 2017). There was no significant statistical difference between filter efficiencies when turbidity was evaluated ($p = 0.972$).

It is important to highlight that HSSF accepts a maximum turbidity of 50 NTU, according to Cawst (2012); however, high turbidity values often generate cleanliness of the unit, reducing the filter efficiency when the overall performance is evaluated. In this context, influent water with low turbidity is always desired.

Conductivity drastically increased in the filtered water with a statistically significant difference for I-HSSF ($p = 0.001$). However, the value was always below 50 $\mu S\ m^{-1}$ for both filters. Conductivity depends on ion concentration (i.e. phosphate, chloride, sulphate, nitrate, silicon, aluminium, calcium, iron, magnesium, sodium, etc.) and most of the time all these ions increased considerably after filtration (Table 4), and this may explain our findings. Likewise, Young-Rojanschi and Madramootoo (2015) noticed an increase in the conductivity and pH from filtered water and this anomaly was intensified when the influent water stayed longer in contact with the filter media (i.e. longer residence period) and they attributed this phenomenon to the filter media leaching. Therefore, this finding may explain why the conductivity was higher for the C-HSSF in our study (mean HRT = 14.3 h).

Increased pH (2-4%) in the filtered water was observed in both filters, a similar fact also reported by Young-Rojanschi and Madramootoo (2015). Murphy et al. (2010) attributed the increased pH to the calcium carbonate leaching from concrete-built HSSF walls. As the filters were acrylic fabricated in our study, the leaching from filter media may better explain this phenomenon. No significant statistical difference between filters was found for this parameter ($p = 0.061$).

There was a slight temperature variation (1.0%) throughout the tests with around 22 ^{o}C in the filtered water. However, no significant statistical difference between filters was found ($p = 0.860$). Arnold et al. (2016) stated that HSSF could be effective at any temperature above freezing; nevertheless, the biological layer needs time to adapt to changes in the temperature. They also indicated that HSSF should be kept at warmer temperatures since the coldest temperatures have less bacteria removal in the operational beginning. In this context, this parameter was not pointed out as a limiting factor for the HSSF efficiency in our study.

DO reductions were detected in filtered water (60-66%); however, anoxic conditions were not noticed. No significant statistical difference between HSSFs was identified ($p = 0.181$). DO consumption is expected in HSSF due to the biological layer development (Young-Rojanschi and Madramootoo, 2015). According to Kennedy et al. (2012), both pH and DO decreased during the operation of their tested HSSFs and this phenomenon was most likely due to carbon oxidation. Young-Rojanschi and Madramootoo (2014) found anoxic conditions in HSSF and this condition is not desired since nitrate reduction may occur to nitrite, as observed by Murphy et al. (2010). Based upon our experimental results, DO cannot be considered as a restrictive factor for HSSF efficiency.

I-HSSF showed statistically significant BPA removal efficiency than the C-HSSF (p = 0.001). However, mean PBA removal was low (3%) and on some occasions, the PBA concentration was higher in the filtered water than the influent water (Fig. 4). BPA removal in the I-HSSF may be explained by biosorption from bacteria, as described by Vecchio et al. (1998), who evaluated heavy metal biosorption by bacterial cells, and by Vijayaraghavan and Yun (2008), who published a review about the status of biosorption technology.

[Figure 4 near here]

There was an unexpected BPA increase in the C-HSSF filtered water. Nonetheless, this may be explained by PBA desorption from the sand surface, as reported by Tran et al. (2002) for cadmium. In addition, this could be caused by BPA accumulation inside the living cells and when they die, the accumulated BPA may enter the water again, as reported by Terin and Sabogal-Paz (2019) for cyanobacteria and consequent microcystin production. Katayama-Hirayama et al. (2010) evaluated a lab-scale SSF efficiency to treat river water with tetrabromobisphenol A. They found low removal (20%) at the initial concentration of 100 μg L^{-1} throughout the experimental period (18 days). According to these authors, bisphenol removal by SSF may be related to the type of chemical structure, since hydroxylation of a phenol ring is an early step in microbial aromatic degradation. An attached group next to a hydroxyl group may inhibit phenol hydroxylation and this may explain the results obtained in our study.

Both filters showed low DOC removal (7 to 12%), however the C-HSSF had statistically significant DOC reduction efficiency (p = 0.003). This result agrees well with D'Alessio et al. (2015) and Terin and Sabogal-Paz (2019) who found TOC removals up to 11% in the filtered water. Contrary to other research, DOC in the influent water was higher (132.92 ± 15.50 mg L^{-1}) once Elliott et al. (2015) reported TOC values up to 12.5 mg L^{-1} in influent water to HSSFs.

According to PBA and DOC removals, HSSF as a single treatment was not effective in terms of eliminating organic compounds; therefore, activated carbon adsorption as an HSSF's post-treatment is recommended for generating safe water in rural communities. Li et al. (2018) obtained promising results when using granular activated carbon sandwich slow sand filtration to remove pharmaceutical and personal care products.

Both HSSFs did not show a significant statistical difference in the reduction of total coliforms (p = 0.686), with the mean in the range of 0.78 to 0.84 log. This efficiency was lower than the ones reported by Lynn et al. (2013) and Pompei et al. (2017) with 1.2 log and 2.0 log, respectively. Coliform removal depends on *schmutzdecke* development and a slow ripening may be responsible for the low reduction rate. The filters in our study needed frequent cleaning (vertical lines indicate maintenance activity in Fig. 3 and Fig. 4), since both HSSFs reached their maximum hydraulic head quickly, a fact that may have influenced the complete development of the biological layer.

Filtered water presented an increase in phosphate, chloride, sulphate, nitrate, silicon, aluminium, calcium, iron, potassium, magnesium and sodium concentrations for both HSSFs (Table 4). This indicates that there was a mineralisation in the filtered water. There was a higher calcium and magnesium increase in the C-HSSF ($p = 0.004$ and $p = 0.036$, respectively) and, on the other hand, for the other ions there were no significant statistical differences between filters.

The presence of some of these ions may be a result of sand leaching, a fact that can be confirmed, since the fine sand presented SiO_2, Fe_2O_3, Al_2O_3 and K_2O in its composition, according to the supplier's information. The influent water (which simulated rainwater) was slightly acidic and had low mineral ion concentrations. Therefore, it was relatively aggressive and could dissolve some compounds from the filter media. WHO (2017) established guideline values for some of the above ions, and for those regulated, the drinking water recommendations were met.

Both filters removed fluoride (55 to 88%) as stated by Devi et al. (2008), who reached an 85.6% reduction by an HSSF. There was a significant statistical difference between filters in our study ($p = 0.045$) showing a better performance for the I-HSSF. According to the WHO (2017), the guideline value is 1.5 mg L^{-1} in drinking water; therefore, the filtered water in our study met this recommendation.

3.3. *Schmutzdecke* analysis

SEM photomicrographs and chemical compositions from synthetic fabric and fine sand are shown in Fig. 5. Potassium, silicon, aluminium, calcium, sodium, chloride and iron were detected in the original fine sand (Fig. 5a), an already expected composition, as discussed above. Potassium was not found in the original synthetic fabric (Fig. 5b). C-HSSF biofilm presented mainly silicon, potassium, magnesium and aluminium in its chemical composition (Fig. 5 c and Fig. 5 d); however, magnesium was not detected in the I-HSSF biofilm (Fig. 5 e and Fig. 5 f). Evidently, all the above ions helped the development of the biological layer in the filters (Fig. 6), providing essential nutrients. As established by Faria Maciel and Sabogal-Paz (2018), the increase of nutrients in HSSFs accelerates the filter maturation process.

[Figure 5 near here]

[Figure 6 near here]

Flow cytometry assay results are shown in Fig. 7. C-HSSF showed a high number of live and dead cells; however, I-HSSF presented slightly higher live cell percentages (99.7% vs 98.9%).

[Figure 7 near here]

According to Chan et al. (2018), flow cytometry with DNA staining can be used to study the microbial dynamics in both treatment and distribution of drinking water and, in the case of our study, the technique may evaluate the state of the biological layer in relation to the presence of live microorganisms, which can help the water treatment.

As reported by Hall-Stoodley et al. (2004), biofilms are structurally complex, dynamic systems with attributes of both primordial multicellular organisms and multifaceted ecosystems. Biofilm formation is a protected mode of growth that allows cells to colonise new niches or survive in adverse environments. Optimising nutrient and waste-product exchange provides the first link between form and function of the biofilm in both natural and fabricated environments. In addition, this theory can be applied to the *schmutzdecke* development in both filters of our study. Evidently, there is still a need to understand how the microorganisms grow in the HSSF biofilm, therefore, further research is recommended.

4. Conclusions

- Water with low mineral ion concentrations generated sand leaching, increasing the values of turbidity, conductivity, pH, phosphate, chloride, sulphate, nitrate, silicon, aluminium, calcium, iron, potassium, magnesium and sodium in the filtered water. In this context, when making the analogy with rainwater, care must be taken in relation to the selection of filter media and construction materials in order to reduce the risk of introducing pollutants in drinking water.
- Operational differences related to continuous and intermittent flow showed influence in the filter efficiency for BPA and DOC for the I-HSSF and C-HSSF, respectively, although the mean performance was low. Consequently, HSSF as a single treatment was not effective for the removal of organic compounds, possibly by the slow *schmutzdecke* development in both filters.
- Activated carbon adsorption as an HSSF's post-treatment must be researched to improve BPA and DOC removals in drinking water for rural communities.
- Strategies to improve the HSSF hydraulic performance compared to ideal plug flow reactor are not required. However, more research is needed to understand the role of the HSSF biological layer in water treatment.

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Statement

The authors hereby declare previous originality check, no conflict of interest and open access to the repository of data used in this paper for scientific purposes.

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