

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

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13 Household slow sand filter (HSSF) has been used as an alternative to drinking
14 water treatment in rural communities worldwide; however, its performance to
15 treat influent water with quality similar to rainwater still needs further studies.
16 Rainwater presents low pH and slight mineral ion concentrations, an aspect that
17 can modify the filter media and consequently the HSSF efficiency. Furthermore,
18 house roofs used in rainwater harvesting can be made of plastic. Therefore, it
19 can introduce chemicals such as Bisphenol A (BPA) in the water. In this context,
20 two pilot-scale HSSFs operated in continuous and intermittent flows were
21 evaluated to treat water containing BPA and low mineral ion concentrations in

22 order to assess the filter performance. Filter media leaching was noticed in the
23 trials; thus, filter media and construction material selection must be carefully
24 evaluated to eliminate risks of pollutant occurrence in drinking water.

25 Operational differences between continuous and intermittent flows influenced
26 the HSSF efficiency for BPA and DOC removals; even so, the filters'
27 performance was low probably due to the slow *schmutzdecke* development.

28 According to tracer test results, HSSF can be classified as a plug flow reactor
29 and strategies to improve its hydraulic performance are not required.

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

32 **1. Introduction**

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

46 **1.1. HSSF basic concepts**

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

61

62 **Table 1.** Influent water quality and HSSF efficiency without pre or post treatment.

Author	Influent Water	Turbidity		<i>Escherichia coli</i>	
		Initial value (NTU)	Removal (%)	Initial value (CFU/100 mL)	Reduction (log)
Elliott et al. (2008)	Raw water	1.86-8.96	74.8(a)*	255 ± 33	0.5-1.9*
Faria Maciel and Sabogal-Paz (2018)	Mixture of well water with kaolinite	10.92- 11.75 (a)	85(a)	3,969 - 5,021(a)	1.26- 2.29(a)
Frank et al. (2014)	Mixture of tap water with sewage	19.9 (a)	75.4(a)	39,400(a)	1.88(a)*

Lynn et al. (2013)	Mixture of raw water with sewage	7.5(a)	86(a)	1.11×10^4 (a)	1.7(a)
Murphy et al. (2010)	Deep well	10.4(a)	98(a)*	109(a)	0.92(a)*
	Surface water	9.4(a)	90(a)*	5,842(a)	1.74(a)*
Young-Rojanschi and Madramootoo (2014)	Raw water	12.6 ± 7.3	87-96(a)	410 ± 60	1.67-3.71

Notes: the greatest efficiency happens when the biological layer is formed; the symbol (a) means average; *values calculated by the authors with the data available in the manuscript.

63

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

67

68 **1.2. HSSF in intermittent and continuous flows**

69 HSSF is a modified SSF which works with a higher filtration rate (up to 29 times) and a
 70 smaller sand layer (up to 50% less) than the conventional filter. HSSF cleaning
 71 processes do not require removing the top of the filter media (Cawst, 2012) and it has
 72 reduced the scale, compatible with a household water treatment (WHO, 2016). A single
 73 user can build an HSSF with easily accessible materials (Faria Maciel and Sabogal-Paz,
 74 2018) and it can operate with intermittent flow, an operational aspect not possible in
 75 conventional SSF. Furthermore, HSSF can improve its performance by installing a non-
 76 woven synthetic fabric on the top of the filter media (Faria Maciel and Sabogal-Paz,
 77 2018), which can be easily positioned and fixed because the filter has a small superficial
 78 area, usually, up to 0.1 m^2 .

79 HSSF in intermittent flow (I-HSSF) can operate with filtration rates up to 29
 80 $\text{m}^3 \cdot \text{m}^{-2} \cdot \text{day}^{-1}$ (1.2 m/h), depending on the hydraulic head (Elliott et al., 2006). Water to

81 be treated has to rest in the pores of the filter media for a period of 1 to 48 h (i.e. pause
82 period) between each batch operation (Cawst, 2012). This pause period is important to
83 allow physico-chemical and biological processes to act on the *schmutzdecke* to treat
84 water. The pause period is a design parameter directly related to the HSSF efficiency
85 and its establishment (1 to 48 h) is not yet fully understood. The user feeds the I-HSSF
86 manually with 15-20 L directly into the unit after the pause period. The treated volume
87 corresponds to the water that is retained in the filter media; consequently, a unit can
88 usually produce up to 80 L day⁻¹ according to the pause period adopted (Schmidt and
89 Cairncross, 2009). The I-HSSF area occupied inside the residence is around 0.1 m².

90 HSSF in continuous flow (C-HSSF) usually works with lower filtration rates, up
91 to 9.6 m³.m⁻²day⁻¹ (Faria Maciel and Sabogal-Paz, 2018). The filter can produce up to
92 200 L day⁻¹ of filtered water, depending on filter configuration. C-HSSF can be fed by
93 gravity (using an elevated tank) or by direct pumping. This filter needs a filtration rate
94 control and may require more area inside the home (± 1.0 m²) as it demands an external
95 supply unit (i.e. elevated tank or pump).

96

97 **1.3. HSSF hydraulic behaviour**

98 HSSF flow characterisation is an important operational parameter (e.g. it can
99 define the water sampling time) and few studies have considered this aspect. Bradley et
100 al. (2011), Elliott et al. (2008) and Lynn et al. (2013) have evaluated I-HSSF hydraulic
101 behaviour and classified it as a plug flow reactor. The C-HSSF has been also classified
102 as a plug flow reactor by Faria Maciel and Sabogal-Paz (2018), Terin and Sabogal-Paz
103 (2019), and Young-Rojanschi and Madramootoo (2015). However, relatively little
104 attention has been given to the hydrodynamics of these filters.

105

106 **1.4. HSSF versus emerging contaminants**

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

115

116 **1.5. Bisphenol A, risk and detection**

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

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

125 It has been detected in drinking water and food and has been banned from plastic
126 containers in Europe and Canada (Rogers et al., 2013). BPA in drinking water may arise
127 from its contact with polycarbonate plastics and epoxy resins (FAO and WHO, 2011) or
128 contaminated raw water. According to Vom Saal and Hughes (2005), 115 *in vivo*
129 studies were published regarding the effects of low BPA dosages and 94 indicated
130 significant effects. In addition, in 31 publications on vertebrates and invertebrates,

131 endocrine changes were found with apparently safe dosages (<50 $\mu\text{g kg}^{-1}\text{day}^{-1}$). An
132 estrogenic effect was confirmed by *in vitro* tests with disruption of cell function
133 (Beausoleil et al., 2018; Vom Saal and Hughes, 2005). Finally, the above authors
134 reported that there is a need to consider the health risk based on the scientific literature
135 relating adverse effects on animals in dosages considered safe.

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

140 BPA detection in environment matrixes has generated the development of
141 chromatographic techniques. Methods based on high performance liquid
142 chromatography (HPLC) have usually been used for BPA analyses (Rodriguez-Mozaz
143 et al., 2004). HPLC may be impracticable in developing countries due to high cost and
144 technical complexity. Therefore, simpler methods that can detect BPA are needed to
145 assess the drinking water risk. From this perspective, UV absorbance of BPA can be
146 measured with a spectrophotometer, allowing its quantification in ppm (Cao et al.,
147 2014).

148

149 **1.6. Rainwater harvesting and treatment**

150 Rainwater is slightly acidic and has very low dissolved mineral concentration. Thus, it
151 is relatively aggressive and it can dissolve metals and impurities from catchment and
152 storage tanks, resulting in unacceptably high pollutant concentrations in the water
153 (WHO, 2017). BPA may be present in plastic materials, pipes, fittings and tanks (Huang
154 et al., 2012) that can be used for rainwater harvesting, an aspect that needs more studies.

155 Slow sand filtration is a technology that may be used to treat rainwater in developing
156 countries (Helmreich and Horn, 2009) and its performance should be better understood.

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

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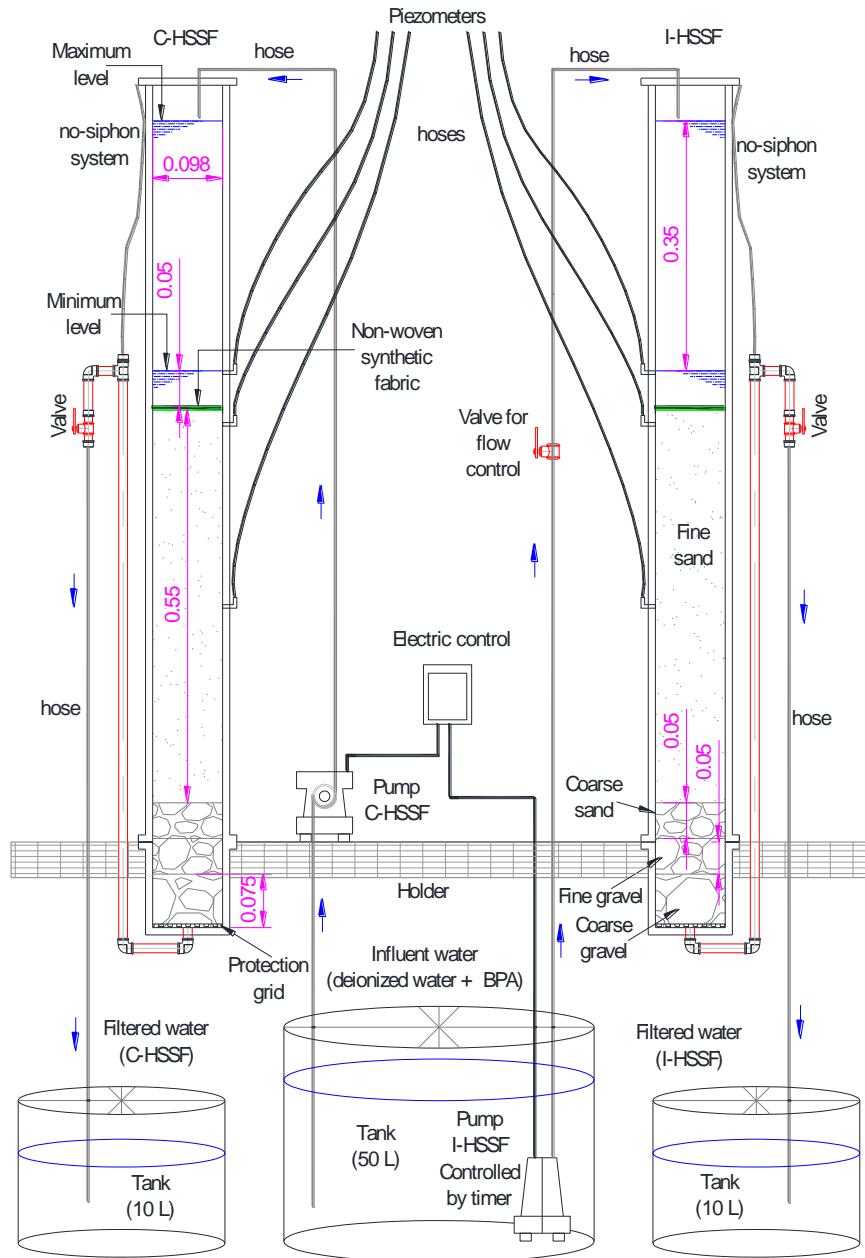
162 **2. Materials and Methods**

163

164 **2.1. HSSF characteristics**

165 Two pilot-scale HSSFs were constructed in acrylic with a 98 mm inside diameter (cross
166 sectional area = 0.0075 m²). One HSSF was designed to operate intermittently (I-HSSF)
167 and the other to operate continuously (C-HSSF). The filters were covered to protect
168 them from light. HSSFs schemes can be found in Fig. 1.

169



170

171 **Fig. 1.** Cross-section of pilot-scale HSSFs (units in meters)

172

173 The HSSF filter media was a 55 cm fine sand layer (0.09 mm to 0.5 mm) with an
 174 effective size (D_{10}) of 0.18 mm and uniformity coefficient (UC) of 1.64. Fine sand used
 175 (CH52, Minerals Marketing, UK) presented the following chemical composition: SiO_2
 176 = 97.3%, Fe_2O_3 = 0.1%, Al_2O_3 = 1.37%, K_2O = 0.83% and loss-on-ignition = 0.25%.

177 Support media consisted of a 5 cm layer of coarse sand (1 to 3 mm), 5 cm layer of fine

178 gravel (3 to 6 mm) and 7.5 cm layer of coarse gravel (10 to 12 mm). The average
179 porosity of the filter materials was 32%. Fine sand and support media were washed in
180 tap water prior to their introduction inside each unit. Acrylic columns were filled with
181 tap water before inserting the filter media to avoid air pocket formation and to allow
182 fine sand stratification as well.

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

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

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

203 **2.2. HSSF operation**

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

207

208 **Table 2.** Influent water quality

Parameter	Mean value ± Standard deviation	Parameter	Mean value ± Standard deviation
Turbidity (NTU)	0.37 ± 0.11	Sulphate (mg L ⁻¹)	1.06 ± 1.63
Conductivity (μS m ⁻¹)	3.24 ± 0.78	Nitrate (mg L ⁻¹)	0.22 ± 0.71
pH	6.10 ± 0.14	Silicon (mg L ⁻¹)	0.002 ± 0.002
Temperature (°C)	22.2 ± 1.6	Aluminium (mg L ⁻¹)	0.006 ± 0.004
DO (mg L ⁻¹)	3.25 ± 0.53	Calcium (mg L ⁻¹)	0.196 ± 0.07
BPA (mg L ⁻¹)	2.35 ± 0.41	Iron (mg L ⁻¹)	0.002 ± 0.002
DOC (mg L ⁻¹)	132.9 ± 15.5	Calcium (mg L ⁻¹)	0.465 ± 0.66
Total coliforms (CFU/100 mL)	11.8 ± 4.3	Sodium (mg L ⁻¹)	0.04 ± 0.03
Phosphate (mg L ⁻¹)	0.64 ± 2.01	Potassium (mg L ⁻¹)	0.035 ± 0.06
Fluoride (mg L ⁻¹)	0.57 ± 0.71	Magnesium (mg L ⁻¹)	0.008 ± 0.004

Deionised water quality used to prepare influent water: turbidity (NTU) = 0.07 ± 0.01; conductivity (μS m⁻¹) = 0.85 ± 0.18; pH = 6.0 ± 0.1; temperature (°C) = 21.4 ± 1.3; DO (mg L⁻¹) = 3.6 ± 0.3; DOC (mg L⁻¹) = 0.1 ± 0.03; chloride (mg L⁻¹) = 0.03 ± 0.01; measured and undetected: coliforms, lithium, ammonium, phosphate, potassium, bromide, fluoride, sulphate, nitrite, nitrate, silicon, aluminium, calcium, iron, magnesium and sodium.

209

210 HSSF filtration rates were calculated considering a daily production of 2.9 ± 0.9
211 L for the C-HSSF and 2.6 ± 0.8 L for the I-HSSF. The flow rate in the C-HSSF was
212 controlled by a peristaltic pump (Watson-Warlow, MHRE 100) producing a filtration

213 rate of $0.38 \pm 0.13 \text{ m}^3 \text{ m}^{-2} \text{ day}^{-1}$. The I-HSSF hydraulic head was variable generating a
214 filtration rate between 0 to $21 \text{ m}^3 \text{ m}^{-2} \text{ day}^{-1}$. The I-HSSF was filled with 1.0 L (filter
215 volume) three times per day by a submersible pump (Jeneca ®, HM 5063) controlled
216 with a valve and timer, causing an 8-hour pause period.

217 **2.3.Tracer tests**

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

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

231 The first filling to the I-HSSF was carried out with NaCl solution and the
232 subsequent feedings were with tap water. The filtration rate declined to zero when the
233 hydraulic head reached the lowest level, at which time a new water charging was
234 performed ($V = 1.0 \text{ L}$). Salt concentration versus filter volume curves produced a
235 positive step followed by a negative step (increased and decreased concentrations).
236 Afterwards, the Morrill Dispersion Index (MDI) and the modified MDI (mMDI) were

237 calculated as described by Tchobanoglous et al. (2003) and Lynn et al. (2013),
238 respectively.

239

240 **2.4. BPA detection**

241 BPA was measured by UV-Vis spectrophotometer (Shimadzu UV 2600, Japan). UV
242 absorbance for six BPA concentrations (0 to 12 mg L⁻¹) was measured from 200 to 1000
243 nm wavelengths, in triplicate, in order to identify the characteristic absorbance peak (it
244 was detected at 224 nm). Afterwards, the BPA standard curve was made from data
245 obtained at 224 nm. The relationship between UV absorbance and BPA concentration
246 was established [UV absorbance = 0.0748 x BPA concentration (mg L⁻¹)]. The
247 calibrated curve showed r² of 0.94, detection limit of 0.03 mg L⁻¹ and limit of
248 quantification of 0.10 mg L⁻¹.

249

250 **2.5. *Schmutzdecke* evaluation**

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

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

261 Bacteria cells (alive and dead) were determined by flow cytometry using
262 Guava® easyCyte 5HT Benchtop Flow Cytometer (Millipore, UK). Samples from the
263 biological layer for I-HSSF and C-HSSF at the end of the filter operation were collected
264 and stored at 4 °C before processing. LIVE/DEAD BacLight Bacterial Viability kit
265 (Thermo Fisher Scientific, UK), with propidium iodide dye and SYTO® 9 dye, was
266 prepared and applied according to the manufacturer's instructions. 20 µL of sample
267 (*schmutzdecke*) and controls (*E. coli* strain K-12 and deionised water) were added to
268 180 µL of the prepared stock staining into 1.5 mL microcentrifuge tubes.

269 *E. coli* was diluted before measuring in the flow cytometer in filtered deionised
270 water (0.22 mm; PTFE Syringe, Gilson scientific). It was used as a biological positive
271 control, and filtered deionised water was utilised as a control for background
272 fluorescence. All prepared samples were incubated at room temperature in the dark for
273 15 min. The bacteria acquisition gate was determined according to forward scatter
274 (FSC) and side scatter (SSC) channels to eliminate background noise and debris.

275

276 **2.6. Sample collection and analysis**

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

285 **2.7. Statistical analyses**

286 Statistical analyses were performed using PAST 3 software (PAIaeontological
287 STatistics) created by Hammer et al. (2018). The Kruskal-Wallis test was used to
288 compare data from the filtered water samples among each other and with influent water
289 (95% confidence interval). When statistical analyses showed that the mean values were
290 significantly different, the Mann-Whitney test was selected to define which sample was
291 different from another (95% confidence interval).

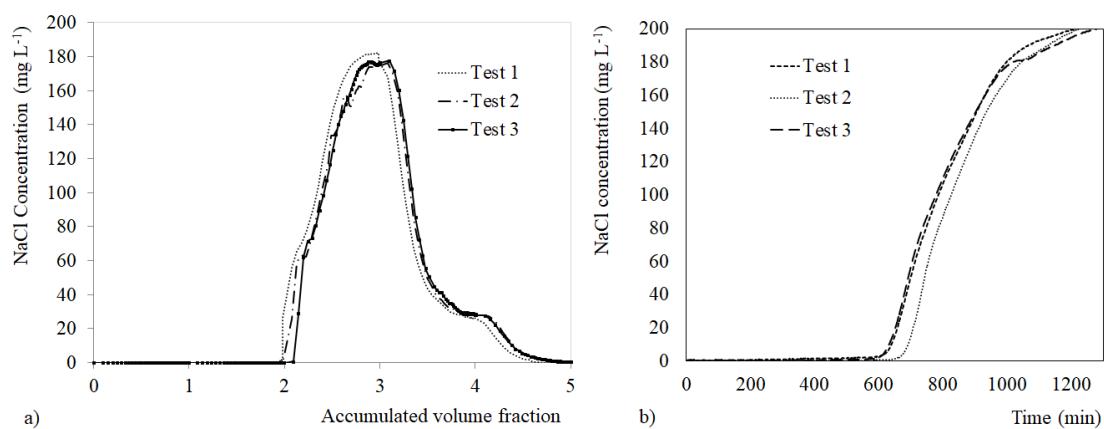
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293 **3. Results and Discussion**

294 **3.1. Tracer tests**

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

299



300 a) b)
301 **Fig. 2.** Tracer tests results for I-HSSF (a) and C-HSSF (b)

302

303 According to the results, two feedings were required before collecting samples
304 for the I-HSSF performance evaluation. Salt concentration decreased from the third

305 filter volume and after the fifth feeding, the tracer left the filter (Fig. 2a). Similar
306 behaviour was described by Bradley et al. (2011), Faria Maciel and Sabogal-Paz (2018)
307 and Terin and Sabogal-Paz (2019), characterising a plug flow reactor for HSSF.

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

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

318 Tracer test results for the C-HSSF are shown in Fig. 2b and Table 3. HRT was
319 857 ± 21 min and it was used to determine the sample collection time. The N-CSTR
320 model showed a better adjustment with an r^2 of 0.75 and N of 17. As indicated by
321 Levenspiel (1999), a high N value also designates a plug flow reactor.

322

323 **Table 3.** Tracer test results for the C-HSSF

Statistic	HRT	N-CSTR		Small dispersion model		High dispersion model	
		N	r^2	D/uL	r^2	D/uL	r^2
Mean	857	17	0.75	0.032	0.67	0.028	0.66
Standard deviation	21	5	0.02	0.008	0.03	0.007	0.03

Notes: N-CSTR: N-continuous stirred tank reactors model; N: number of stirred tank reactors; D/uL: dimensionless group characterising the spread in the whole reactor (close to zero denote negligible dispersion, hence, plug flow reactor); HRT: hydraulic retention time; and r^2 : coefficient of determination.

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

333

334 **3.2. HSSF operation**

335 Filtered water quality and removal or variation rates are shown in Table 4.
336 Turbidity removal showed a negative value for both filters (i.e. filtered water presented
337 74-76% higher turbidity) and there was no removal improvement over time (Fig. 3),
338 contradicting the literature.

339

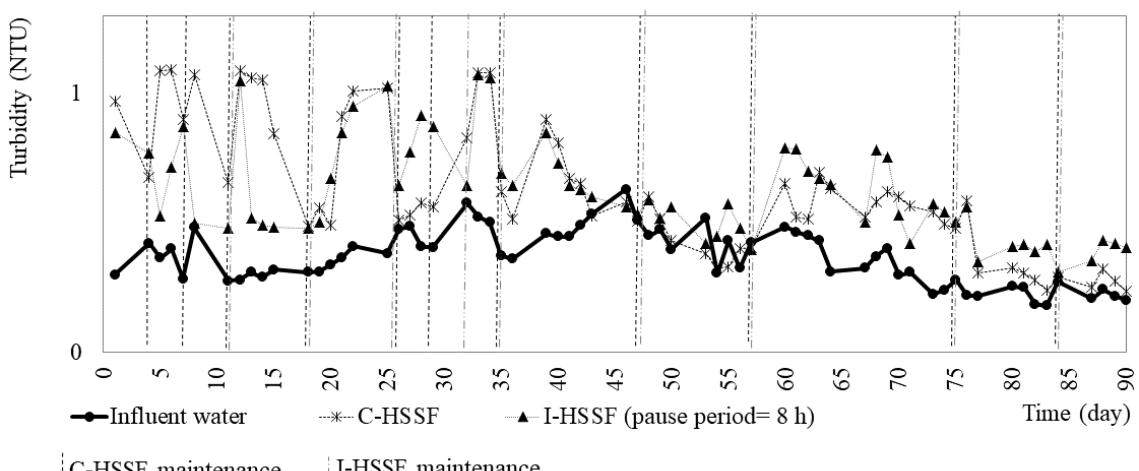
340 **Table 4.** Filtered water quality and removal or variation rates for I-HSSF and C-HSSF

Parameter	C-HSSF		I-HSSF		<i>p</i> -value
	Value (M \pm SD)	Removal (R) or variation (V) (%) (M \pm SD)	Value (M \pm SD)	Removal (R) or variation (V) (%) (M \pm SD)	
Turbidity (NTU)	0.62 \pm 0.3	-74 \pm 73 (R)	0.62 \pm 0.2	-76 \pm 53 (R)	0.972
Conductivity (μ S m $^{-1}$)	36.2 \pm 10.4	-1063 \pm 386 (V)	30.0 \pm 8.0	-868 \pm 313 (V)	0.001 SS
pH	6.2 \pm 0.3	-2 \pm 5 (V)	6.3 \pm 0.4	-4 \pm 6 (V)	0.061
Temperature (°C)	22.1 \pm 1.6	1 \pm 1 (V)	22.1 \pm 1.6	1 \pm 1 (V)	0.860
DO (mg L $^{-1}$)	1.2 \pm 0.8	60 \pm 28 (V)	1.1 \pm 0.6	66 \pm 22 (V)	0.181
BPA (mg L $^{-1}$)	2.65 \pm 0.37	-14 \pm 16 (R)	2.26 \pm 0.31	3 \pm 8 (R)	0.001 SS
DOC (mg L $^{-1}$)	115.9 \pm 14.8	12 \pm 9 (R)	123.2 \pm 12.3	7 \pm 6 (R)	0.003 SS

Total coliforms (CFU 100mL ⁻¹)	2.3 ± 1.7	0.78 log ± 0.3 log (R)	1.92 ± 1.38	0.84 log ± 0.25 log (R)	0.686
Phosphate (mg L ⁻¹)	1.86 ± 3.10	-12 ± 58 (V)	2.50 ± 3.67	-18 ± 60 (V)	0.501
Fluoride (mg L ⁻¹)	0.44 ± 0.84	55 ± 65 (R)	0.11 ± 0.18	88 ± 17 (R)	0.045 SS
Chloride (mg L ⁻¹)	0.17 ± 0.20	-86 ± 202 (V)	0.17 ± 0.17	-60 ± 141 (V)	0.785
Sulphate (mg L ⁻¹)	1.70 ± 2.17	-18 ± 26 (V)	1.58 ± 2.24	-10 ± 74 (V)	0.844
Nitrate (mg L ⁻¹)	0.28 ± 0.87	-43 ± 141 (V)	0.38 ± 1.09	-89 ± 174 (V)	0.712
Silicon (mg L ⁻¹)	0.46 ± 0.05	-38804 ± 19491 (V)	0.43 ± 0.07	-36820 ± 20359 (V)	0.412
Aluminium (mg L ⁻¹)	0.01 ± 0.003	-533 ± 873 (V)	0.01 ± 0.002	-846 ± 1678 (V)	0.675
Calcium (mg L ⁻¹)	4.18 ± 0.32	-2271 ± 894 (V)	3.39 ± 0.30	-1883 ± 940 (V)	0.004 SS
Iron (mg L ⁻¹)	0.01 ± 0.004	-345 ± 372 (V)	0.01 ± 0.003	-349 ± 190 (V)	0.930
Potassium (mg L ⁻¹)	0.19 ± 0.10	-1523 ± 1275 (V)	0.13 ± 0.01	-919 ± 558 (V)	0.174
Magnesium (mg L ⁻¹)	0.92 ± 0.08	-14577 ± 8155 (V)	0.80 ± 0.07	-12644 ± 7717 (V)	0.036 SS
Sodium (mg L ⁻¹)	0.11 ± 0.06	-316 ± 289 (V)	0.10 ± 0.04	-312 ± 361 (V)	0.764

Notes: M: mean; SD: standard deviation; statistically significant difference (SS) when p-value < 0.05

341



342

343 **Fig. 3.** Turbidity variation for the influent water and filtered water over time for I-HSSF
344 and C-HSSF

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

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

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

365 Conductivity drastically increased in the filtered water with a statistically
366 significant difference for I-HSSF ($p = 0.001$). However, the value was always below 50
367 $\mu\text{S m}^{-1}$ for both filters. Conductivity depends on ion concentration (i.e. phosphate,
368 chloride, sulphate, nitrate, silicon, aluminium, calcium, iron, magnesium, sodium, etc.)
369 and most of the time all these ions increased considerably after filtration (Table 4), and

370 this may explain our findings. Likewise, Young-Rojanschi and Madramootoo (2015)
371 noticed an increase in the conductivity and pH from filtered water and this anomaly was
372 intensified when the influent water stayed longer in contact with the filter media (i.e.
373 longer residence period) and they attributed this phenomenon to the filter media
374 leaching. Therefore, this finding may explain why the conductivity was higher for the
375 C-HSSF in our study (mean HRT = 14.3 h).

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

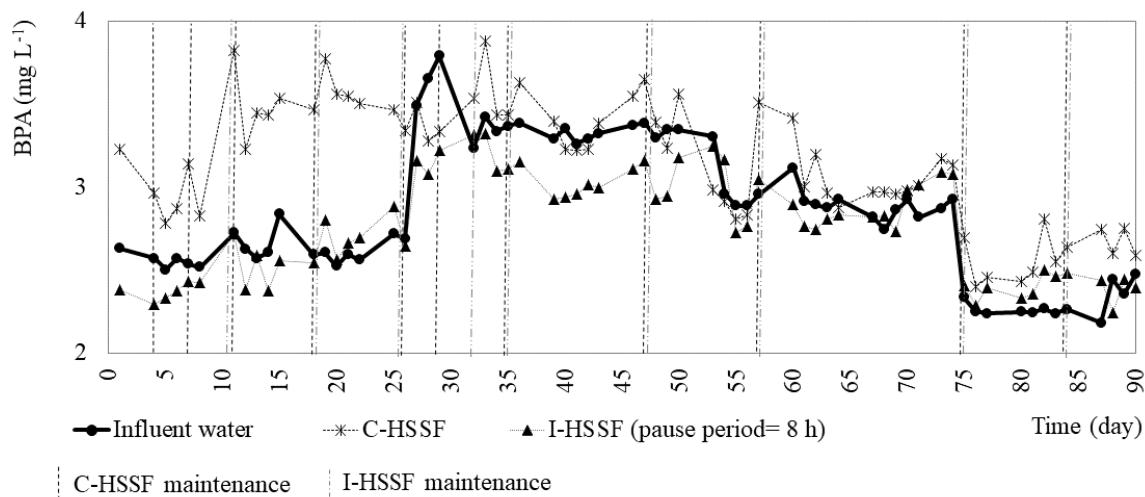
382 There was a slight temperature variation (1.0%) throughout the tests with around
383 22 °C in the filtered water. However, no significant statistical difference between filters
384 was found ($p = 0.860$). Arnold et al. (2016) stated that HSSF could be effective at any
385 temperature above freezing; nevertheless, the biological layer needs time to adapt to
386 changes in the temperature. They also indicated that HSSF should be kept at warmer
387 temperatures since the coldest temperatures have less bacteria removal in the
388 operational beginning. In this context, this parameter was not pointed out as a limiting
389 factor for the HSSF efficiency in our study.

390 DO reductions were detected in filtered water (60-66%); however, anoxic
391 conditions were not noticed. No significant statistical difference between HSSFs was
392 identified ($p = 0.181$). DO consumption is expected in HSSF due to the biological layer
393 development (Young-Rojanschi and Madramootoo, 2015). According to Kennedy et al.
394 (2012), both pH and DO decreased during the operation of their tested HSSFs and this

395 phenomenon was most likely due to carbon oxidation. Young-Rojanschi and
396 Madramootoo (2014) found anoxic conditions in HSSF and this condition is not desired
397 since nitrate reduction may occur to nitrite, as observed by Murphy et al. (2010). Based
398 upon our experimental results, DO cannot be considered as a restrictive factor for HSSF
399 efficiency.

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

407



408 **Fig. 4.** BPA in the influent water and filtered water over time for I-HSSF and C-HSSF

410

411 There was an unexpected BPA increase in the C-HSSF filtered water.

412 Nonetheless, this may be explained by PBA desorption from the sand surface, as

413 reported by Tran et al. (2002) for cadmium. In addition, this could be caused by BPA

414 accumulation inside the living cells and when they die, the accumulated BPA may enter
415 the water again, as reported by Terin and Sabogal-Paz (2019) for cyanobacteria and
416 consequent microcystin production. Katayama-Hirayama et al. (2010) evaluated a lab-
417 scale SSF efficiency to treat river water with tetrabromobisphenol A. They found low
418 removal (20%) at the initial concentration of $100 \mu\text{g L}^{-1}$ throughout the experimental
419 period (18 days). According to these authors, bisphenol removal by SSF may be related
420 to the type of chemical structure, since hydroxylation of a phenol ring is an early step in
421 microbial aromatic degradation. An attached group next to a hydroxyl group may inhibit
422 phenol hydroxylation and this may explain the results obtained in our study.

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

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

435 Both HSSFs did not show a significant statistical difference in the reduction of
436 total coliforms ($p = 0.686$), with the mean in the range of 0.78 to 0.84 log. This
437 efficiency was lower than the ones reported by Lynn et al. (2013) and Pompei et al.
438 (2017) with 1.2 log and 2.0 log, respectively. Coliform removal depends on

439 *schmutzdecke* development and a slow ripening may be responsible for the low
440 reduction rate. The filters in our study needed frequent cleaning (vertical lines indicate
441 maintenance activity in Fig. 3 and Fig. 4), since both HSSFs reached their maximum
442 hydraulic head quickly, a fact that may have influenced the complete development of
443 the biological layer.

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

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

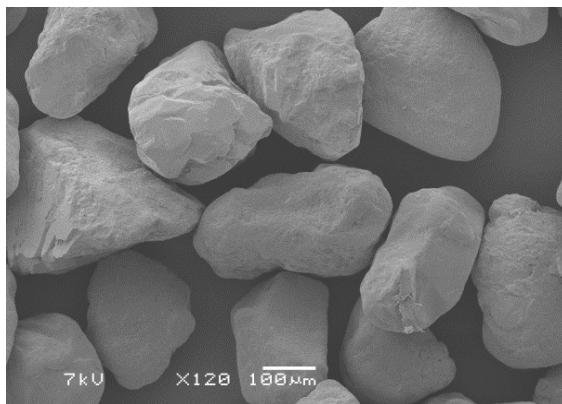
457 Both filters removed fluoride (55 to 88%) as stated by Devi et al. (2008), who
458 reached an 85.6% reduction by an HSSF. There was a significant statistical difference
459 between filters in our study ($p = 0.045$) showing a better performance for the I-HSSF.
460 According to the WHO (2017), the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

462

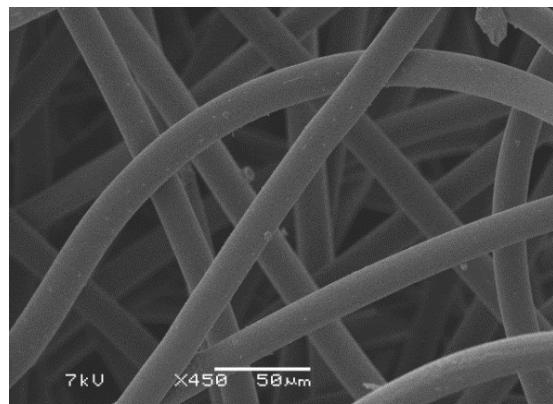
463 **3.3. Schmutzdecke analysis**

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

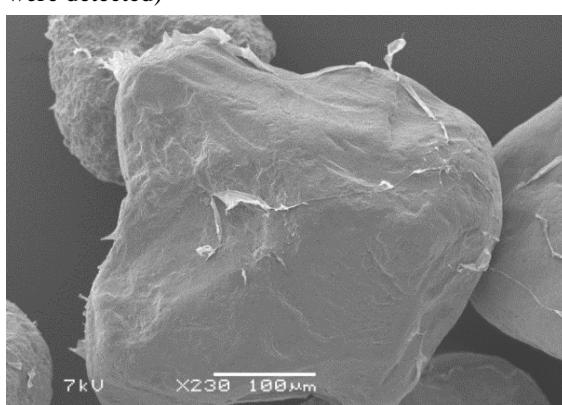
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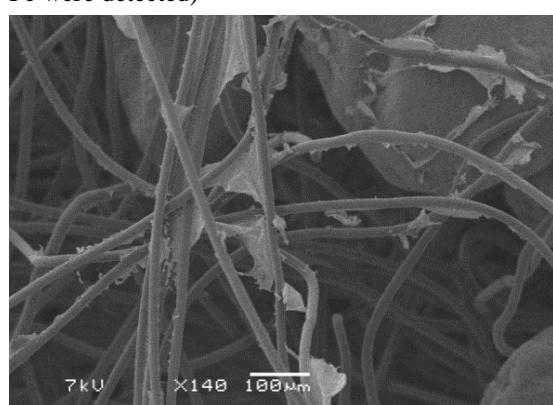
a) original fine sand (K, Si, Al, Ca, Na, Cl and Fe were detected)



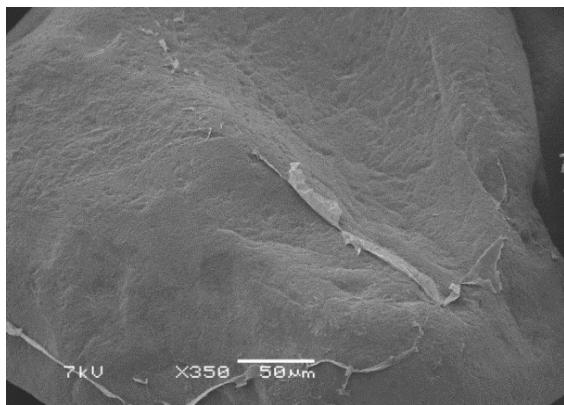
b) original synthetic fabric (Si, Al, Ca, Na, Cl and Fe were detected)



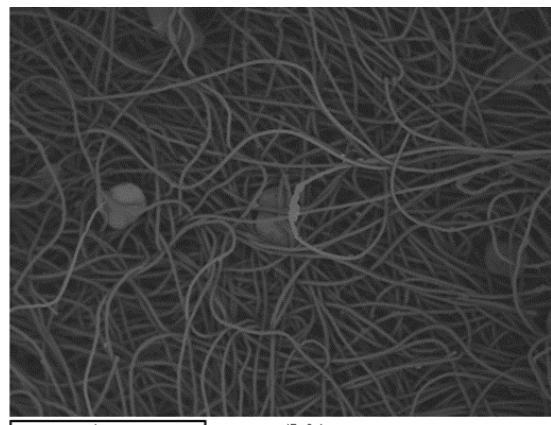
c) C-HSSF fine sand with formed biofilm (Si, K, Mg and Al were detected)



d) C-HSSF synthetic fabric with formed biofilm (Si, K and Al were detected)



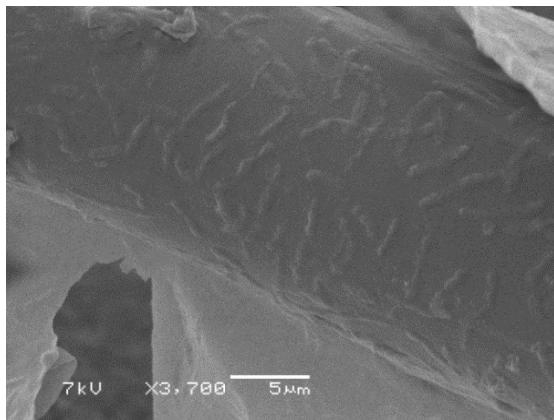
e) I-HSSF fine sand with formed biofilm (Si, Mg and Al were detected)



f) I-HSSF synthetic fabric with formed biofilm (Si, Al and Na were detected)

475 **Fig. 5.** SEM photomicrographs and chemical compositions from the synthetic fabric and
476 fine sand.

477



a) C-HSSF biofilm (synthetic fabric)



b) I-HSSF biofilm (fine sand)

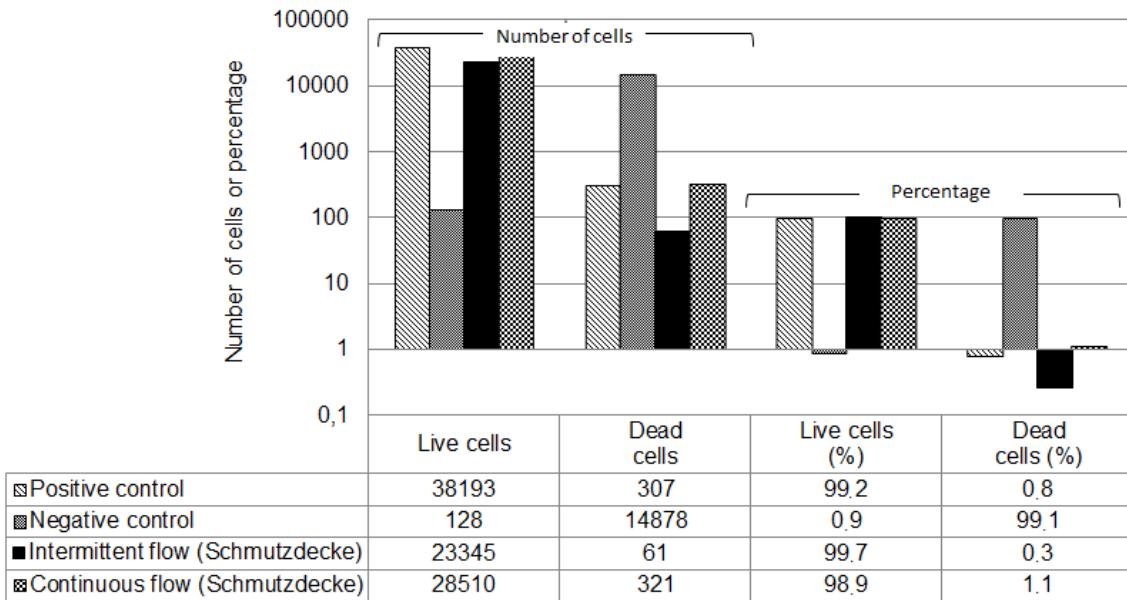
478 **Fig. 6.** Microorganisms visualised in the biofilms by SEM photomicrographs

479

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

483

484



485
486 **Fig. 7.** Flow cytometry results for samples from the biological layer collected at the end
487 of the HSSF operation.

488

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

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

502 **4. Conclusions**

503 • Water with low mineral ion concentrations generated sand leaching, increasing
504 the values of turbidity, conductivity, pH, phosphate, chloride, sulphate, nitrate,
505 silicon, aluminium, calcium, iron, potassium, magnesium and sodium in the
506 filtered water. In this context, when making the analogy with rainwater, care
507 must be taken in relation to the selection of filter media and construction
508 materials in order to reduce the risk of introducing pollutants in drinking water.

509 • Operational differences related to continuous and intermittent flow showed
510 influence in the filter efficiency for BPA and DOC for the I-HSSF and C-HSSF,
511 respectively, although the mean performance was low. Consequently, HSSF as a
512 single treatment was not effective for the removal of organic compounds,
513 possibly by the slow *schmutzdecke* development in both filters.

514 • Activated carbon adsorption as an HSSF's post-treatment must be researched to
515 improve BPA and DOC removals in drinking water for rural communities.

516 • Strategies to improve the HSSF hydraulic performance compared to ideal plug
517 flow reactor are not required. However, more research is needed to understand
518 the role of the HSSF biological layer in water treatment.

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522 **Statement**

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

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