

Czech University of Life Sciences Prague
Faculty of Environmental Sciences



Diploma Thesis

**Greywater infiltration treatment efficiency for the
chemical load removal in different hydraulic loads using
different biofilters**

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DIPLOMA THESIS ASSIGNMENT

Bayan Hamdan

Land scape Engineering
Environmental Modelling

Thesis title

The efficiency of the infiltration treatment in removing the chemical load in greywater in different hydraulic load and biofilters.

Objectives of thesis

The main objective of the thesis is to evaluate the efficiency of the infiltration on treatment in removing the chemical load from the greywater and to compare the treatment efficiency of different hydraulic loads and different filtra on materials used in the greywater infiltration on the system.

Methodology

- Utilization of physical model of infiltration on the unit to treat greywater.
- Running experiment with different hydraulic loads (amount of water) and differfilter materials.
- Laboratory analyses of greywater before and after treatment, by measuring the total organic carbon (TOC), Chemical Oxygen Demand (COD), Electrical Conductivity (EC), Total Phosphorous (TP), Anionic surfactants, Ammonia, pH, metals, and Organic compounds (pharmaceuticals).
- Statistical analyses of the experimental data.
- Evaluation of different filtra on materials treatment efficiency

The proposed extension of the thesis

70 pages

Keywords

greywater, infiltration, chemical load, hydraulic load, water treatment

Recommended information sources

- Al-Gheethi, A.A.S., Noman, E.A., Mohamed, R.M.S.R., Bala, J.D. and Kassim, A.H.M., 2019: Qualitative Characterization on of Household Greywater in Developing Countries. Book of Management of Greywater in Developing Countries. pp. 1-31.
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 - Sun, Y. et al. 2018. Organics removal, nitrogen removal, and N₂O emission in subsurface wastewater infiltration on systems amended with/without biochar and sludge. *Journal of Bioresource Technology* 249, pp. 57–61.
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I hereby declare that I have independently elaborated the diploma/final thesis on the efficiency of the infiltration treatment in removing the chemical load from the greywater in different hydraulic and filter materials. And that I have cited all the information sources that I used in the thesis and that are also listed at the end of the thesis in the list of used information sources. I am aware that my diploma/final thesis is subject to Act No. 121/2000 Coll., on copyright, on rights related to copyright, and on amendment of some acts, as amended by later regulations, particularly the provisions of Section 35(3) of the act on the use of the thesis. By submitting the diploma/final thesis, I am aware that I agree with its publication under Act No. 111/1998 Coll., on universities and the change and amendments of some acts, as amended, regardless of the result of its defense. With my signature, I also declare that the electronic version is identical to the printed version and the data stated in the thesis has been processed about the GDPR.

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Abstrakt

Tento výzkum zkoumal účinnost infiltrační léčby šedé vody pod dvěma úrovněmi saturace 70 a 30% a pěti různými typy biofiltrů (default, štěpek, mykorhiza, biochar 5% a biochar 10%). Pro zkoumání jejich účinku účinnosti léčby byla provedena laboratorní analýza, která obsahuje analýzu pH, EC, TN, TP, amonného dusíku, těžkých kovů, TOC, povrchově aktivních látek, léčiv a různých iontů, jako je síran, chlorid, fluorid, dusitan a dusičnan, který poskytuje obecný údaj o kvalitě vody. Bylo zjištěno, že odtok šedé vody má neutrální hodnoty pH a EC, které leží v normách kvality vody pro zavlažování (EPA, 2012). Maximální účinnost odstraňování byla pozorována při 30% úrovni nasycení a nejvyšší výkon byl biochar pro účinnost odstraňování TOC, TN, povrchově aktivních látek, DEET a diklofenaku a kovů. Nejvyšší účinnosti odstranění DEET a diklofenaku bylo dosaženo při 30% saturaci pro biochar 5% s 91,6% a 98,1% příslušně. Kovy byly úspěšně odstraněny biofiltry s účinností odstraňování mezi 90,6 - 95,93%. Bór však byl nalezen vyluhovaný. Pokud jde o amonný dusík a celkový fosfor, spolu s biocharem dosáhla štěpka slibné účinnosti odstraňování. Uvedené výsledky naznačují biochar jako účinný biofiltr, levný a udržitelný kromě nízké úrovně nasycení pro optimální kvalitu odpadních vod.

Klíčová slova: šedá voda; účinnost odstraňování; hydraulické zatížení; chemické zatížení; povrchově aktivní látky; léčiva; syntetická šedá vody

Abstract

This research studied the efficiency of the infiltration treatment of greywater under two levels of saturation 70 and 30%, and five different types of biofilters (default, woodchip, mycorrhiza, biochar 5%, and biochar 10%). To examine their effect of the efficiency of the treatment a laboratory analysis was conducted that contains an analysis of pH, EC, TN, TP, ammonium nitrogen, heavy metals, TOC, surfactants, pharmaceuticals, and different ions such as sulphate chloride, fluoride, nitrite, and nitrate that gives a general indication of the water quality. It was found that greywater effluent has neutral pH and EC values that lie within water quality standards for irrigation (EPA, 2012). The maximum removal efficiency was observed at a 30% saturation level and the highest performance was biochar for the removal efficiency of TOC, TN, surfactants, DEET and diclofenac, and metals. The highest removal efficiencies of DEET and diclofenac were achieved in 30% saturation for biochar 5% with 91.6% and 98.1% respectively. The metals were successfully removed by the biofilters with removal efficiencies ranging between 90.6 – 95.93%. However, the boron was found leached. Biochar 10% and woodchip have the highest removal efficiency of ammonium nitrogen and phosphorus and the optimum saturation is 30%. Given results suggest the biochar is an effective biofilter, inexpensive and sustainable in addition to a low saturation level for optimum effluent quality.

Keywords: greywater; removal efficiency; hydraulic load; chemical load; surfactants; pharmaceuticals; synthetic greywater

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List of Abbreviations

BOD	Biological Oxygen Demand
COD	Chemical Oxygen Demand
CW	Constructed Wetlands
DEET	Diethyltoluamide
DO	Dissolved Oxygen
EC	Electrical Conductivity
FWS	Free Water Surface
GROW	Green Roof Water Recycle
GW	Greywater
HGW	Heavy Greywater
HLR	Hydraulic Load Rate
HPLC	High-Performance Liquid Chromatography
HSSF	Horizontal Subsurface Flow
IC	Inorganic Carbon
LAS	Linear Alkylbenzene Sulfonate
LGW	Light Greywater
NBS	Nature-Based Solutions
NSAID	Non-Steroidal Anti-Inflammatory Drug
OMP	Organic Micropollutants
PCP	Personal Care Products
PPCP	Pharmaceuticals and Personal Care Products
RVF	Recycling Vertical Flow
SGW	Synthetic Greywater
SZ	Saturated Zone
TC	Total Carbon
TN	Total Nitrogen
TOC	Total Organic Carbon
TOrCs	Trace Organics Compounds
TP	Total Phosphorus
TSS	Total Suspended Solids
VF	Vertical Flow
XOC	Xenobiotic Organic Compounds

1 Introduction

Shortage in freshwater sources on the planet has been associated with the increasing human population, economic development, and living standards. This has resulted in more water reclamation and reuse, a significant issue at hand. Due to the pressing need for new water resources in recent years, greywater treatment and reuse are increasing attention (Revitt et al., 2011). Greywater represents up to 75% of total domestic wastewater and ranges between 100-150 l/p/d (liter/person/day) in high-income countries and smaller volume in low-income countries (Boano et al., 2020), which can be reduced by 40-60 l/p/d by reusing the Greywater (Friedler et al., 2005). Greywater treatment and reuse for non-drinking purposes is one practical approach in reducing the consumption of purified water and solving the water resources scarcity in urban areas so that it can save 29%-47% of drinking water consumption (Mahmoudi et al., 2021). Moreover, treated greywater is considered suitable for non-potable purposes such as toilet flushing and irrigation (Eriksson et al., 2009).

2 Objectives

This study aims to investigate the efficiency of the greywater infiltration treatment to remove the chemical load in two different hydraulic loads; 30 and 70%. In addition to examining the efficiency of the different biofilters and their contribution to the removal efficiency. The biofilters used in this study are biochar 5%, biochar 10%, woodchip, mycorrhiza, and default (without any additive).

Hence, the objectives of the study are:

To evaluate the greywater infiltration treatment in different hydraulic loads 30 and 70%.

- To evaluate the efficiency of the different biofilters.
- To evaluate the removal of the surfactants and the pharmaceuticals, DEET, and diclofenac.
- To evaluate the removal of heavy metals, boron, zinc, nickel, and copper.
- To evaluate the removal of total nitrogen, ammonium nitrogen, nitrate, and nitrite.
- To evaluate the removal of phosphorus and the different ions, sulphate, chloride, and fluoride.

3 Literature Review

3.1 Greywater origin

Domestic wastewater is divided into two main categories; black water, which originated from the toilet, and greywater (GW), which comprises wastewater from bathtubs, showers, laundry machines, washbasins, and kitchen sinks (Boyjoo et al., 2013). Figure 1 below demonstrates the division of household water consumption. Generally, the most significant water consumption is from the laundry (33%), shower (23%), and toilets (20%) (Al-Gheethi et al., 2019). The composition and the quantity of the GW produced in households vary depending on the amount and the type of chemicals used (detergents, soaps, shaving cream, fats, grease, toothpaste, etc.), household occupancy, water availability, gender, age, country, etc. Moreover, GW production varies during the day, with the highest amounts before and after the working hours (Boyjoo et al., 2013).

Greywater's quality is higher than that of black water. Even though its quality does not allow direct use, its better quality makes the water amenable for on-site treatment and reuse, such as irrigation and toilet flushing (Gross et al., 2007). For example, in Israel, more than 70% of its treated GW is reused for agriculture irrigation (Travis et al., 2010). The usage of greywater for irrigation in gardens and small-scale agriculture has the potential to maximize the use of limited water supplies and improve the food security of the rural poor since the greywater contains macronutrients, particularly nitrogen and phosphorus, the availability of these nutrients in proper amount may benefit the plants' growth (Radingoana et al., 2020). Indoor application of treated Greywater for toilet flushing in urban areas may reduce ca, 30% of building's water consumption (Maimon and Gross, 2018).

In addition to GW reuse for toilet flushing and irrigation, treated GW can be used for vehicle and window washing, fire extinguishing, concrete production, golf course irrigation, fertilization of crops and groundwater recharges (Al-Ismaili et al., 2017).

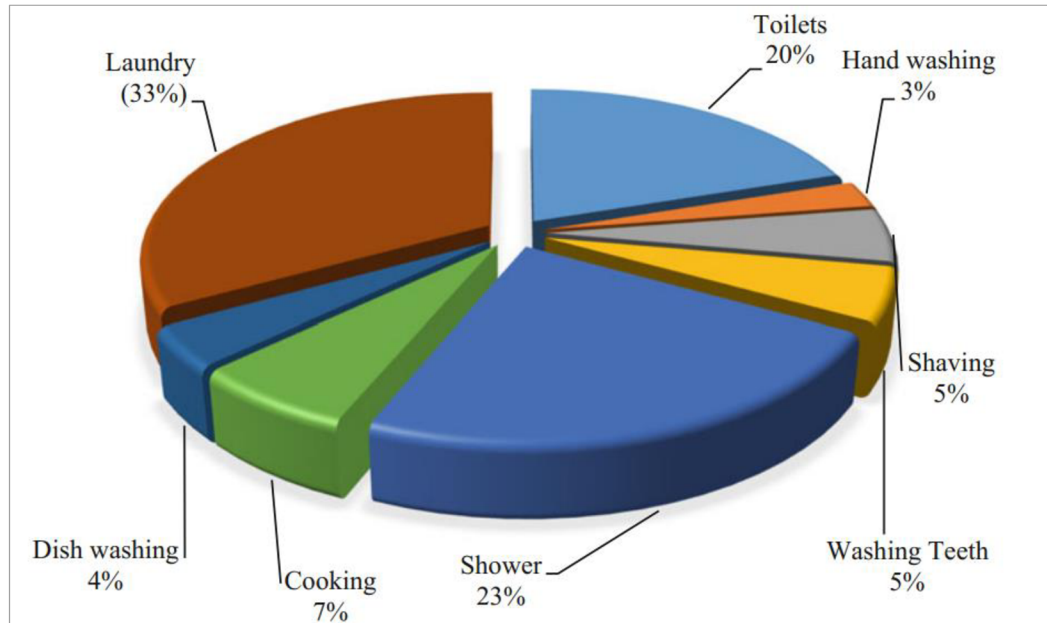


Figure 1: Domestic water consumption per capita per day. (Adopted from Al-Gheethi et al 2019)

3.2 Greywater classification

Greywater composition varies significantly due to the lifestyle of household members and the chemical compounds used in laundry, bathing, and cleaning. The mixture of the materials used in the household provides different kinds of pollutants into the Greywater. Another essential factor that influences the composition of Greywater is the type of the distribution network, chemical and biological degradation of certain compounds in the network storage (Al-Gheethi et al., 2019), (Oteng-Peprah et al., 2018). Household wastewater can be classified into six categories including brown water (wastewater with feces), yellow water (urine), black water (wastewater containing both urine and feces), greywater (mainly from detergents derived from laundry and sinks, bathing, and self-care products), green water (food particles, wastewater derived from dishwashing machines and kitchen sinks), and stormwater (rainwater) (Al-Gheethi et al., 2019).

In some places, the term Greywater does not include kitchen wastewater due to its high rate of organic load originating from oil, grease, and detergents (Organization, 2006). Therefore, (Bodnar et al., 2014) divided Greywater into two main categories: light and dark Greywater. Light Greywater originates from bathing as well as that from laundry and kitchen as dark Greywater.

3.2.1 The chemical and physical composition

The main parameters of GW include biochemical oxygen demand (BOD), chemical oxygen demand (COD), suspended solids, turbidity, total nitrogen (TN), total phosphorus (TP), pH, alkalinity, electrical conductivity, heavy metals, surfactants, and detergents (Al-Gheethi et al., 2019). Temperature, color, turbidity, and content of suspended solids are referring to physical parameters. High temperatures may be unfavorable since they favor microbial growth and could in supersaturated waters, induce precipitation (e.g., calcite) (Eriksson et al., 2002). Boyjoo et al. (2013) show in their study detailed tables illustrating the chemical composition of both light greywater (LGW) and dark (high) greywater (HGW) in various countries (Table 1).

In term of quality and chemical composition, the GW contain chemical compounds such as soaps, detergents, oils and grease, metal salts, chemical dye, and pathogens (Eslami et al., 2017). In the laundry, the main chemical compounds that can be found are cations such as Ca, Mg, K, nitrate, sulphate anions, carbonate, and chloride as well as organic micropollutants (OMPs) (Mohamed et al., 2018). In addition, Linear Alkylbenzene Sulfonate (LAS) is a conventional anionic detergent that is used in 80% of household detergent and can be considerably found in greywater, it is important to mention that LAS is very toxic to marine life and result in foam formation in water resources (Eslami et al., 2017).

Kitchen greywater contains alkaline chemical pollutants such as detergents and cleaning agents, fats and oils, food residues, traces of food preservatives, raw meat washing, and vegetables and fruits peels. Kitchen GW also contains the highest rate of turbidity, suspended solids, organic substances, and nitrogen. Automatic kitchen dishwasher originates high pH, salinity, oil and grease, bacteria, organic matter, turbidity, soaps, odor, and suspended solids (Shaikh and Ahammed, 2020). The quality and composition of Greywater are like a quantity of the Greywater, it is varied in time and location (Eriksson et al., 2002).

Table 1: LGW characteristics from several countries. (B – Bath GW, F – Floor cleaning GW, S – Shower GW, W – Washbasin GW, BOD5 – 5-day BOD, * – 7-day BOD (BOD7). (Boyjoo et al., 2013)

Light GW characteristics	Units	Australia (Christova-Boal et al. 1996) B	Taiwan (Lin et al. 2005) S	Korea (Kim et al. 2007) F	France (Chaillou et al. 2011) B, S	Germany (Nolde 2000) B, S	UK (Pidou et al. 2008; Winward et al. 2008b) B, S, W	Spain (March et al. 2004; Gual et al. 2008) B, S, W	Israel (Ramona et al. 2004; Friedler et al. 2008, 2005; Friedler & Gilboa 2010) B, S, W	Morocco (Merz et al. 2007; Scheumann et al. 2007) S	Oman (Prathapar et al. 2005) S, W
pH		6.4–8.1	7	7.27	7.58	–	6.6–7.3	6.8–7.6	7.5	7.6	7.1–7.4
EC	$\mu\text{S cm}^{-1}$	82–250	–	194	468	–	–	921	1,241	645–855	14–15
Turbidity	NTU	60–240	43.1	12.6	150	–	35–42	20–38.8	23–34	29	133–375
SS	mg L^{-1}	48–120	29	–	125	–	29	32.2–44	29.8–61.3	–	353–505
Nitrate (NO_3^-)	mg L^{-1}	<0.05–0.20	–	–	–	–	3.9–7.5	–	0.67	0	10.2–28.7
Ammonia ($\text{NH}_3/\text{NH}_4^+$)	mg L^{-1}	<0.1–15	0.146	–	–	–	0.7–1	–	2.7	6.6–11.8	–
Total Kjeldahl nitrogen (TKN)	mg L^{-1}	4.6–20	–	–	–	–	–	–	–	11.9–15.2	–
Total nitrogen (TN)	mg L^{-1}	–	–	–	9.5	5–10	7.6–16.4	4.1–11.4	–	–	–
Phosphate (PO_4^{3-})	mg L^{-1}	–	–	–	–	–	0.5–1.3	–	0.09	1	–
Total phosphorus (TP)	mg L^{-1}	0.11–1.8	–	–	0.42	0.2–0.6	–	–	–	0.98–1.6	–
BOD ₅	mg L^{-1}	76–200	23	–	240	50–300*	20–166	–	59–104	53–59	42.1–130
COD _{total}	mg L^{-1}	–	–	–	399	–	–	–	148–170	–	–
COD _{dissolved}	mg L^{-1}	–	55	22.9	136	100– 633	86–575	72.7–171	86–110	109–122	58–294.3
TOC	mg L^{-1}	–	–	–	50.6	26–95	12–56	41–58	–	–	70.2–83.5
Surfactants	mg L^{-1}	–	–	–	6.8	–	–	–	–	–	14.9–41.9
Heterotrophic plate count	Counts/100 mL	–	–	–	1.87×10^9	1×10^5 to 1×10^6	–	–	1.3×10^7 to 2.0×10^9	–	–
Total coliforms	Counts/100 mL	500 to 2.4×10^7	–	0	–	10 to 1.0×10^5	4.0×10^5	–	–	–	> 200.5
Faecal coliforms	Counts/100 mL	170 to 3.3×10^3	–	–	3.42×10^5	0.1–10	–	–	5.6×10^5	1.4×10^5 to 2.48×10^5	–
Faecal streptococci	Counts/100 mL	79 to 2.4×10^3	–	–	–	–	–	–	–	–	–
<i>Escherichia coli</i>	Counts/100 mL	–	5.1×10^3	–	4.76×10^5	–	–	–	–	–	> 200.5

Table 14: HGW chemical characteristics in different countries. K-Kitchen GW, L-Laundry GW, M-Mixed GW, S-Shower GW, * - BOD7. (Boyjoo et al., 2013)

Moderate/ heavy GW characteristics	Units	Australia	Japan	Korea	India	Nepal	Costa	Brazil	Holland	Slovenia	Italy	Germany	Sweden	Israel	Turkey	Jordan	Oman
		(Christova- Boal et al. 1996) L	(Itayama et al. 2006) K	(Kim et al. 2009) K, S	(Mandal et al. 2011) M	(Morel & Diener 2006) M	(Dalias et al. 2004) M	(Paulo et al. 2009) S, K, L	(Hernandez Leal et al. 2010b, 2011a, 2007) M	(Sostar- Turk et al. 2005) L	(Ciabattia et al. 2009) L	(Li et al. 2003; Elmitwalli & Otterpohl 2007) M	(WHO (WHO 2006) M	(Gross et al. 2005) M	(Atasoy et al. 2007; Masi et al. 2010) M	(Halalshah et al. 2008) M	(Prathapar et al. 2005) L
pH		9.3-10	-	-	7.3-8.1	-	-	-	-	9.6	7-9	6.9-8.1	-	6.7	7.1-7.2	6.35	8.3
EC	$\mu\text{S cm}^{-1}$	190-1,400	-	-	489-550	-	-	-	-	-	1,300-3,000	-	-	-	401-495	1,830	-
Turbidity	NTU	50-210	-	19-84.8	20.6-38.7	-	96	254	-	-	40-150	-	-	-	-	-	444
SS	mg L^{-1}	88-250	105	30-130	12-17.6	98	-	120	-	35	90-200	-	-	138	48-54	168	315
NO_3^- - N	mg L^{-1}	0.1-0.31	-	-	0.5-0.63	-	-	0.05	0.12-0.77	-	-	-	-	-	0.13-1.3	-	25.8
Ammonia ($\text{NH}_3/\text{NH}_4^+$)	mg L^{-1}	<0.1-1.9	-	-	-	13.3	-	2.4	0.8-11.8	2.45	-	-	-	-	1.2-1.3	75	-
TKN	mg L^{-1}	1-40	-	-	-	-	-	-	-	-	-	27.2	-	-	7.6-9	128	-
Total N	mg L^{-1}	-	21	-	42.8-57.7	-	-	8.8	26.3-35.2	2.75	-	9.7-16.6	13.6	14	-	-	-
PO_4^{3-} - P	mg L^{-1}	-	-	-	1.52-3.36	3.1	16	5.6	2.3-2.36	-	-	9.8	-	-	-	-	-
Total P	mg L^{-1}	0.062-42	4	-	-	-	-	-	6.2-7.8	9.9	-	5.2-9.6	5.2	17.7	7.2-7.3	19.5	-
BOD_5	mg L^{-1}	48-290	477	-	56-100	200	167	435	-	195	-	-	260*	270	90-116	1,056	179.7
COD total	mg L^{-1}	-	-	-	-	-	-	-	724-1,004	-	-	640	-	-	245	-	-
COD dissolved	mg L^{-1}	-	271	50-400	244-284	411	-	646	210-376	280	400-1,000	125-354	520	686	177-277	2,568	231.3
TOC	mg L^{-1}	-	-	-	-	-	-	-	157-184.3	-	-	80.2-93.8	-	-	-	-	174.6
Surfactants	mg L^{-1}	-	-	-	-	-	-	-	43.5-54	10.1	0.01-25	-	-	40	-	-	118.3
Total coliforms	Counts/ 100 mL	2.3×10^3 to 3.3×10^5	-	1.2×10^3	3.74×10^4 to 3.8×10^4	-	-	5.4×10^8	-	-	-	-	-	-	1.36×10^4	1.0×10^7	>200.5
Faecal coliforms	Counts/ 100 mL	110 to 1.09×10^3	-	-	3.48×10^4 to 3.56×10^4	-	4.6×10^8	-	-	-	-	-	-	1.0×10^6	3.57×10^3 to 1.1×10^4	3.0×10^5	-
<i>E. coli</i>	Counts/ 100 mL	-	-	4.0×10^3	-	-	-	5.4×10^6	-	-	-	7.5×10^3 to 2.6×10^5	-	-	-	2.0×10^5	>200.5
<i>Staphylococcus aureus</i>	Counts/ 100 mL	-	-	1.8×10^3	-	-	-	-	-	-	-	-	-	-	-	-	-
<i>Salmonella typhimurium</i>	Counts/ 100 mL	-	-	5.4×10^3	-	-	-	-	-	-	-	-	-	-	-	-	-

3.3 Surfactants

Surfactants (surface active agents) are organic molecules consisting of hydrophilic and hydrophobic groups (Wiel-Shafran et al., 2006). Surfactants represent the major organic micropollutants in Greywater because they are used in detergents and hygiene products. The surfactants included the compounds generated from amphoteric, cationic, anionic, and nonionic detergents (Al-Gheethi et al., 2018). According to Wiel-Shafran et al. (2006) results, the surfactant concentration in the Greywater was higher than that in the raw domestic wastewater because it contains mainly wash water. The laundry effluent had the highest mean surfactant concentration, followed by kitchen and bath effluents.

Surfactants along with Personal Care Products (PCPs) enter the wastewater stream due to high solubility without undergoing any metabolic changes. As a result, these compounds persist in the recipient environment for a long time resulting in many adverse effects (Ramprasad and Philip, 2016). Maimon and Gross (2018) reported in their study that surfactants are one of the elements that pose an environmental risk that makes the GW reuse more challenging.

Eslami et al. (2017) conducted a study on the LAS surfactant removal by activated sludge, and it has been conducted that the LAS removal efficiency reached 94.24%. Likewise, Ramprasad and Philip (2016) indicated in their study on the surfactant's removal by a vertical constructed wetland that the surfactant concentrations were not very significant, and they were meeting the USEPA standards for GW reuse.

3.4 Pharmaceutical and personal care products (PPCPs)

Pharmaceuticals and personal care products are part of the Xenobiotic Organic Compounds (XOCs) (Al-Gheethi et al., 2019). And they can be divided into two main categories, PPCPs including pharmaceuticals, for instance, antibiotics, anti-inflammatory drugs, hormones, cytostatic drugs, contrast media, beta-blockers, blood lipid regulators, antiepileptic drugs; and the second category is the personal care products (PCPs), such as antimicrobial, ultra-violet filters, preservatives, insect repellent, and synthetic masks are the collection of the chemical substances used in daily activities of the humans' life (Keerthanan et al., 2021). PPCPs have been widely used in many fields such as medicine, industry, livestock farming, aquaculture, and people's daily life. These products have attracted much attention due to their impact on the environment, public health, and living microorganisms even at extremely low concentrations (Najmi et al., 2020). Wang and Wang, (2016) mentioned in their study that PPCPs occurred in surface water at a concentration ranging from ng/l to $\mu\text{g/l}$, in groundwater at the concentration level of ng/l to mg/l, in sediment with concentration $\mu\text{g/kg}$ and soil achieving $\mu\text{g/kg}$.

Due to the physicochemical properties of the PPCPs, many are not easily removed by conventional water treatment processes, as demonstrated by their existence in the drinking water. Thus, PPCPs are recognized as pseudo persistent organic pollutants in the environment, which may display the same risk to the environment as the truly persistent organic pollutants due to their continuous introduction into the environment, which leads to a potential risk to aquatic organisms and public health (Wang and Wang, 2016); (Ebele et al., 2017). This study contains two different kinds of PPCPs, diclofenac, and DEET.

3.4.1 Diclofenac

Diclofenac (2-(2-(2,6-dichlorophenylamino) phenyl) acetic acid), is a non-steroidal anti-inflammatory drug (NSAID), and it is one of the most consumed drugs in the world. Part of the consumed diclofenac is excreted in its original form so entering municipal wastewater, where its concentration reflects its consumption by the resident in a specific sewer system (Escapa et al., 2016). Westhof et al, (2016) study on micropollutants in the wastewater streams has found that Diclofenac's concentration in the Greywater is much higher than its concentration in the blackwater, and that's since Diclofenac is broadly applied in salve form and is washed off easily.

Like other kinds of pharmaceuticals, Diclofenac has high potential toxicity towards bacteria and can pose a significant risk to human health, aquatic environment, and environmental species (Khasawneh and Palaniandy, 2021).

3.4.2 N, N-diethyl-m-toluamide (DEET)

Personal care product, commonly used as an insect repellent. Washed off the skin easily, it is found in Greywater (Geiling, 2015). Owing to the large consumption in PCP and agriculture, DEET has been detected in wastewater, surface water groundwater and even drinking water with exposure concentrations ranging from not detected to 24 µg/l (Gao et al., 2020). Although DEET is found to be slightly toxic to aquatic invertebrates and freshwater fish, it owns potential carcinogenic properties in human nasal mucosal cells and even the intake of low doses of DEET in children often leads to coma and seizures (Cai et al., 2022). Hence, it is crucial to remove PPCPs such as DEET from the treated GW if we aim to reuse the GW, especially if the treated GW contains a body contact in purpose to reduce the water demand.

According to Cai et al, (2022) study, results showed that UV light-emitting diodes combined with an iron-containing coagulant (UV-LED₂₇₅/FeCl₃) showed high efficiency in DEET removal with an effect of pH solution, DEET degradation exhibited a promoting effect under acidic conditions, and it showed an obstructive effect with the increase of the pH values. Sui et al, (2010) investigated DEET removal by Ultrafiltration, Ozone

(UF/Ozone), sand filter (SF), and Microfiltration/Reverse Osmosis (MF/RO). The study results showed that DEET removal efficiency was 0 – 50% and 50 – 80% for UF/ozone, 0 – 50% for SF, and > 90% for MF/RO.

3.5 Heavy metals

The greywater reuse can pose an environmental and health risk since the GW contain a contaminant such as nutrients (Wiel-Shafran et al., 2006), (Maimon and Gross, 2018), and metals (Eriksson et al., 2002). Metals in Greywater can originate from household products, household appliances, plumbing, and personal care products (Turner et al., 2016). Heavy metals result in Greywater from many sources. Metals including Pb, Hg, Cd, and Ni enter the Greywater through individual sources such as cosmetics, beauty creams, lipsticks, body lotions, jewelry, dental filling, cutlery, and plumbing materials. And their concentration in the untreated greywater can reach 0.12, 0.21, 0.0015, 0.17 g person⁻¹y⁻¹ respectively (Mahmoudi et al., 2021) (Eriksson and Donner, 2009). In addition, (Boyjoo et al., 2013) indicated that laundry detergents are a source of heavy metals such as Cd, Zn, Pb, Cu, and Cr.

Treated greywater, indeed, is a beneficial alternative for freshwater uses. However, (Turner et al., 2016), indicated that treated greywater used for irrigation is a potential source for metals such as Cu, Ni, Cr, Zn, and B in the soil, groundwater, and surface water where their concentration exceeding the environmental quality guidelines and it could potentially be harmful to the environment and the public health. For instance, boron is considered a plant micronutrient and is required in small concentrations, levels slightly higher than those considered beneficial can cause damage and even death to the plants. As a result, the existence of boron can limit the type of plants that can be irrigated with treated greywater (WHO, 2006), (Turner et al., 2016). Zhang et al, (2020) reported that recirculating standing hybrid constructed wetlands (RSHCWs) showed satisfactory and steady removal efficiencies for Pb, Cu, Cd, and Zn, with > 85% removal at a treating time of 24 h or even shorter.

3.6 Effect of the hydraulic load on the water treatment efficiency

Hydraulic load is defined as water flow divided by the cross-section area per period (Godoy-Olmos et al., 2019). Pang et al. (2020) mentioned in their study that the influent of the hydraulic load is an important factor for the wastewater infiltration system, which would affect the purification effect and long-time running. According to Dalahmeh et al. (2014) different hydraulic load affects differently the infiltration system. On one hand, the high hydraulic load increases the infiltration rate and thereby reduces the exchange between mobile water in macropores and water retained in macropores. On the other hand, low hydraulic load result in the greater relative exchange between the mobile and retained water and thus prolong the residence time.

Different studies conducted on the Greywater or wastewater treatment and hydraulic load were one of the factors that have been considered. (Zhang et al., 2005) reported that when the hydraulic load increased, soil clogging occurred due to overfeeding, and soil permeability decreased to a very small value, thus it affects the COD, TN, and $\text{NH}_4\text{-N}$. Fig. 2 and 3 show the removal rate of $\text{NH}_4\text{-N}$ and TN respectively in different hydraulic loadings. The $\text{NH}_4\text{-N}$ removal was increased from 70% to 90% at low hydraulic loading, even when the hydraulic load increased to 8cm d^{-1} (Fig.2). Similarly, to $\text{NH}_4\text{-N}$ removal rate, Fig.3 shows that the TN removal rate reached 90% at a low hydraulic load of 2cm d^{-1} . The removal rate of both, TN, and $\text{NH}_4\text{-N}$, was decreased at a high average hydraulic loading of 10cm d^{-1} , it was explained by soil clogging due to the overfeeding, and the soil permeability also decreased to a very small value.

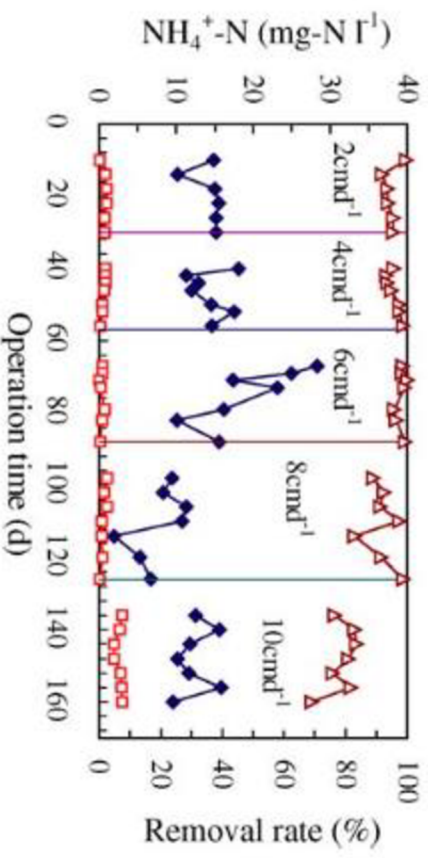


Figure 2: $\text{NH}_4^+\text{-N}$ removal performance in intermittent feeding mode (Zhang et al., 2005)

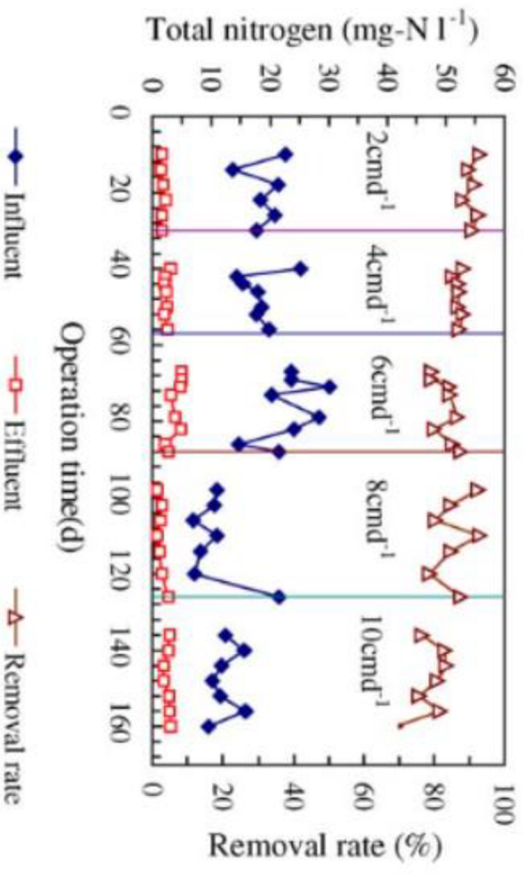


Figure 3: TN removal performance in the intermittent feeding mode (Zhang et al., 2005)

Pang et al. (2020) reported in their study that COD, NH₄-N, TP removal efficiencies decreased in the intermittent aeration WESIS (wastewater ecological soil infiltration system) when the hydraulic load increased. However, a hydraulic load of 0.2m/d achieved a high removal efficiency of 89.4%, 90.8%, 87.2%, 92.9% for COD, NH₄-N, TN, and TP respectively. Another study by Dalahmeh et al. (2014) about charcoal, sand, and bark filters for greywater treatment, indicated that different materials responded differently to the Hydraulic Load Rate (HLR). For instance, in bark filters, increasing the HLR leads to a decrease in the capacity of the filters to remove BOD₅, COD, TP, NH₄⁺-N, and NO₃-N. Therefore, when the HLR decreased, the removal of the pollutants increased again. While increasing the HLR for sand and charcoal filters, increased their capacity to remove the organic matters. Removing personal care products from greywater also can be affected by different hydraulic loads, Ren et al. (2021) indicating that the removal of four targets PCPs was higher at a hydraulic load of 5 cm/d than a hydraulic load of 8 cm/d.

(Trang et al., 2010) indicated in their study, pollutants removal from domestic wastewater by constructed wetlands, that there is an inverse relationship between hydraulic load and removal efficiencies of the pollutants. For instance, COD removal decreased from 84% at HLR of 31 mm day⁻¹ to 63% at HLR of 146 mm day⁻¹. Moreover, the most significant change was for ammonia NH₄⁺-N, from 91% at the lowest HLR of 31 mm day⁻¹ to 0% at the highest HLR of 146 mm day⁻¹. Also, the increase in the HLR affects negatively the removal of the TP and PO₄⁻-P.

3.7 Greywater treatment

Greywater has the potential to be a source of clean water. Focus on greywater treatment technology hovers around the easiness of its implementations, is low-cost, and does not produce any residue compounds. However, it is well known that Greywater contains a high level of organic matter, sulfates, suspended solids, and fecal contamination that can be found in small amounts which, as mentioned previously, can generate unpleasant

odor, and pose a microbial risk and environmental harm (Kurniawan et al., 2021). Before conducting greywater, treatment there are conventional assessed parameters (chemical and physical ones) needed to be considered to evaluate the GW quality, these parameters are shown in the table.3. For bacterial assessment, the most used is *Escherichia coli*, and the Xenobiotic Organic Compounds usually include surfactants, fragrance flavors, preservatives, and solvents (Boano et al., 2020).

The greywater characteristics depend firstly on the quality of the water supply, secondly on the type of distribution network for both drinking water and Greywater, and thirdly on the activities in the household (Eriksson et al., 2002). Furthermore, the compounds in the Greywater vary from source to source, where the lifestyle, costumes, installations, and use of chemical household products will be of importance.

Another parameter affecting greywater quality that needed to pay attention to is the storage of the Greywater. The storage system is one of the selected options which are used for Greywater before and after the treatment process. Storage systems are used mainly for the accumulation of the wastewater before subjecting it to the treatment process and for regulating the reuse in irrigation. In addition, the storage systems might the microbiological quality of the greywater in terms of pathogenic bacterial reduction due to the deficiency in the favorable conditions necessary for their growth such as nutrients, and increase the competition between the microorganisms in the greywater (Al-Gheethi et al., 2019). However, Liu et al. (2010) mentioned that BOD in Greywater reduced by 50% in 4 h period. Consequently, longer residence time in the storage tank can encourage bacteria re-growth and lead to degradation of water quality.

Table 22: Conventional parameters to characterize the GW quality.

(Boano et al., 2020)

Physical		Chemical	
Parameter	Units	Parameter	Units
Temperature	°C	pH	
Turbidity	NTU	Biochemical Oxygen Demand	mg/L
Total solids	mg/L	Chemical Oxygen Demand	mg/L
Total suspended solids	mg/L	Total Organic Carbon	mg/L
Total dissolved solids	mg/L	Dissolved Organic Carbon	mg/L
Biological		Nitrate	mg/L
Total coliforms	MPN/100 mL	Ammonium	mg/L
Faecal coliforms	MPN/100 mL	Oxidized nitrogen	mg/L
<i>Escherichia coli</i>	MPN/100 mL	Total Nitrogen	mg/L
F-RNA bacteriophage	MPN/100 mL	Total Phosphorus	mg/L
<i>Clostridium perfringens</i>	MPN/100 mL	Phosphate	mg/L
<i>Bacteroidales</i>	MPN/100 mL	Heavy metals	mg/L
		Xenobiotic Organic Compounds	mg/L

Another concern about long period greywater storage is the low levels of DO which are caused due to the oxidation and the degradation processes which consume the oxygen available in the Greywater. This suggests the degradation of the organic matter to simple substances and thus their more availability for the plants (Al-Gheethi et al., 2019). Similarly, (Shaikh and Ahammed, 2020) reported that storage of 24-48 h was beneficial in terms of partial removal of solids and organics. Yet, beyond 48 h, the storage affects negatively and disturbs the ambiance by smelling because of initiation of degradation.

Therefore, and after all, mentioned above, knowledge of characteristics and their variations is important in the selection of the greywater treatment system. Quality requirements, guidelines, and reuse limits depend on the type of reuse, the origin of greywater, and the possibility of human contact with recycled water. Several countries around the world have/ or working on the guidelines for the reuse of treated Greywater for non-potable reuse (Eriksson et al., 2002). (Boano et al., 2020) provided a detailed table for different countries' guidelines for GW reuse.

3.8 Greywater treatment technologies

Several technologies have been used for greywater treatment from physical filtration systems such as membranes and sand filters to highly automated and energy-intensive systems that include biological, chemical, and physical mechanisms. Constructed wetlands are often suggested as an economically and energetically efficient way of treating various wastewater streams (Arden and Ma, 2018).

3.8.1 Nature-Based Solutions

Nature-Based Solution (NBS) has been recently integrated into the new European framework program for research and innovation, whose main purpose is to preserve and maintain biodiversity and ecosystems in general, could also be a promising alternative for low-cost efficient treatment of emerging pollutants such as PPCPs. NBS includes constructed wetlands, wastewater stabilization ponds, and filters which are relatively cost-effective options (Nas et al., 2021). Another benefit for NBSs (Jung et al., 2019) is that NBSs have the added benefits of low energy consumption, low initial integration cost, and easy ongoing maintenance, as well as aesthetic appeal.

3.8.2 Constructed Wetlands

Constructed wetlands (CWs) are often used for the decentralized treatment of greywater. The technology is widely used due to the ability to combine simple techniques that do not require operational skills and low energy and implementation costs. On the other hand, the interaction between treatment processes, effluent characteristics, and a long period of system operation could result in gradual substrate clogging, which will negatively affect hydraulic performance and pollutant removal in CWs and as a result, reduce the life of the system. Clogging is one of the major disadvantages of the horizontal flow constructed wetlands where is caused by retention of organic and inorganic particles, pollutant precipitation, biofilm formation, and plant root growth (Bernardes et al., 2019). Constructed wetlands systems assessed

included free water surface (FWS), horizontal subsurface flow (HSSF), vertical flow (VF), recycling vertical flow (RVF), combination systems (e.g. FWS + HSSF), and green roof water recycle (GROW) (Arden and Ma, 2018).

The performance of the CW depends on the type of constructed wetland, vegetation, applied hydraulic load, and media used in the bed (Pared et al., 2021). The results indicated in the same study that CW has removal efficiency of 80%-91% BOD, 60%-85% COD, and 80%-95% TSS. Also, low hydraulic load provided better performance in a CW. Gross et al. (2007) indicated that after 8 h, the RVFW efficiently removed all TSS, BOD, 80% of COD, TN and TP were significantly reduced, and over 90% of surfactants also were removed after 8 h. However, after 12 h, most of the parameters reached a steady-state, with 90-99% of the TSS, BOD, TN, and anionic surfactants, 70-75% of the TP, and 50% of the total Boron having been removed.

3.8.3 Biofilters

Biofilters (also known as bioretention systems or rain gardens) are nature-based solutions (Zhang et al., 2019). Biofilters consist of a planted soil filter media, a sand transition, and a drainage layer and are operated via gravity-fed filtration (Jung et al., 2019). Recently, biofilters designs have been modified and tested with a raised outlet to create a saturated zone (SZ) or submerged anoxic zone where SZ inclusion improved pollutant removal by enhancing denitrification, maintaining plant life and filter media consistency during the dry weather periods, and retaining microorganisms in anoxic condition enhancing biological treatment. (Jung et al., 2019).

The properties of the filter's media are playing a major role in treatments containing filtration. Systems such as biofilters have demonstrated that media plays a critical role as it provides physical support for plants and facilitates the primary removal processes for pollutants such as suspended solids, phosphorus, and heavy metals (Prodanovic et al., 2017). Dalahmeh et al. (2014) highlighted the importance of the filter media properties for the treatment functioning. Sand filters, which have a macropore range, could remove organic matter from greywater through the attachment of the active

biofilms to the sand particles' surface to mineralize the organic matter. Similarly, to pore size of the sand, bark, and charcoal are distinguished by larger specific surface area, higher porosity, and higher organic content than the sand. Dalahmeh et al. (2014) find out that due to the properties of the bark and charcoal, they were more efficient in the removal of the organic matter and phosphorus, while sands filter properties didn't support the organic matter removal. Likewise, Zhang et al. (2019) find out in their study that the sand showed low removal efficiency of micropollutants <50% and limited ability to remove phosphorus. (Dalahmeh et al., 2018) find out that Biochar seems to be a promising material for on-site treatment, especially for non-biodegradable pharmaceuticals removal. Also, biochar shows removal efficiency for COD ($95\pm 3\%$) and TN ($>95\%$). (Dalahmeh et al., 2018) indicated in their study about biochar filters' potential for pharmaceuticals adsorption that biochar is an efficient medium for removal of nonbiodegradable pharmaceuticals, prominently by adsorption. Also, biochar filters in combination with active biofilm are more efficient than sand filters in the removal of organic matter and nitrogen while both media are poor in phosphorus removal. Another study that investigated the performance of different biofilters detected that biochar biofilters (10 and 5% volume) showed high removal efficiency for xenobiotic compounds and were less efficient in TN removal, while woodchip filters showed more promising efficiency in TN removal (Çömez. 2021).

3.9 The impact of greywater on the environment.

Population growth, urbanization, droughts, and climate change have contributed to the decline in both the quantity and quality of traditional water sources (Glover et al., 2021). Global climate change models are predicting that the Mediterranean climatic regions of South Africa, Australia, and Southwestern USA will become hotter and drier putting severe strain on freshwater recourses. This resulted in an increased global focus on wastewater reuse, in particular, greywater since greywater is less polluted and contaminated than other types of wastewaters (Hardie et al., 2021).

It is recommended to use treated greywater for irrigation in gardens. However, where water is scarce and/or water restrictions tight individual households may likely use untreated greywater on their gardens irrespective of regulations, guidelines, and safety.

Environmental concerns have been raised about the greywater reuse related to soil deterioration, surface, and groundwater since greywater contains heavy, micropollutants, etc. (Turner et al., 2019; Eriksson and Donner, 2009). Soil is the immediate recipient of greywater used for irrigation. Some studies have investigated the greywater ingredients that might change the soil properties such as elevated salinity, soil clogging, high pH, and change in the biochemical processes (Maimon & Gross, 2018). Moreover, Turner et al, (2016) stated in their study that it is important to understand the transport of the metals from the greywater as metals adsorption to soils and desorption can lead to off-site transport of metals via leaching resulting in contamination of groundwater and surface water. Erosion of contaminated soil can also cause surface water contamination. As laundry greywater is a source of metals and surfactants, Shafran et al, (2006) indicated in their study on the laundry greywater effect on the soil that greywater irrigated plants developed brown patches (chlorosis) in the tip of the leaves due to the elevated salinity and Boron levels in the leaves. Furthermore, applying 200 mm of powdered detergent to soils results in significant deterioration of soil quality in terms of sipping the soil of humus, decreasing the hydraulic conductivity (47 – 82% decrease), increasing the soil salinity, and alkalinity (Hardie et al., 2021).

Despite the importance of the pharmaceutically active compounds to ensure the health and the life quality of the population, the overuse of pharmaceuticals has been of concern in several countries since this contributes to the increase of these compounds in the environment. However, many treatments used in WWTPs are not effective for the complete removal of micropollutants including pharmaceuticals which is results in their presence of several pharmaceutical compounds intreated wastewater, surface water and even drinking water (Dos Santos et al. 2021). Additionally, Kadewa et al, (2020) explained the complexity of the greywater treatment due to the high variability of its physio-chemical and microbiological properties resulting from

the use of pharmaceuticals and personal care products. According to Glover et al (2021), in their study about the risk assessment of trace organic compounds (TOrcs) in greywater, they detected 12 compounds of TOrcs in untreated greywater and stated that due to incomplete removal of these compounds TOrcs have been detected in the aquatic environment and import toxicity through various mods of actions.

4 Methods and materials

This chapter reviews the methods and the materials used in this study by describing the exact procedure for the sampling and analysis of the samples. The first part describes the field experiment of the greywater system and sampling. Presented after are the laboratory analysis and the parameters used to assess the treated greywater quality.

It starts with a field experiment that contains a set of barrels divided into groups of five that contain different compositions of filtration materials. The barrels were loaded with synthetic greywater (SGW) in different hydraulic loads on daily basis except weekends. Samples of SGW before the treatment and samples of effluent were collected twice a week; SGW on Monday after 70 hr operation and on Friday, 22 hr operation.

In purpose to examine the quality and to analyze the samples collected from the field, laboratory testing was conducted and the parameters that used to assess the treated greywater are pH, electrical conductivity, anionic surfactants, ammonia, phosphorus, TN, TOC, TC, pharmaceuticals, Ions (nitrate, nitrite, chloride, sulphate, and fluoride) and metals.

To analyze the efficiency of greywater infiltration, samplings were carried out seasonally, as described in table 4. Starting from July 2021 till the 31st of October 2021.



Figure 4: Physical model of infiltration.

4.1 Barrels design and filter materials.

The physical model of infiltration contains 50 barrels. All the barrels have an identical volume of 75 L and dimensions of 63 cm height and 44 cm width, and the inner dimensions are 60 cm and 40.5 cm respectively. Each barrel consists of the same soil mixing which is based on the natural soil, sand, compost, and additive of the target filter material. In the bottom of the barrel is a 40 mm layer of gravel with a particle range of 4 – 8 mm. Followed by a 160mm layer of sand with a particles range of 0 – 4 mm. The upper layer is 350 mm of technogenic soil that in its base are soil, sand, and compost. The three layers were separated with a geotextile layer (300 g/m^3) to avoid clogging and mixing of the particles.

Table 23: The experiment time series.

<i>Season</i>	<i>Duration</i>	<i>Field experimental</i>	<i>Analysis</i>	<i>Saturation volume</i>
<i>Summer</i>	July - August, 2021	SGW irrigation, collecting samples and laboratory analysis	pH, TOC, TC, TN EC, metals, ammonium pharmaceuticals and anionic surfactants	70% and 30%
13/08/2021 - 01/09/2021 Break of two weeks				
<i>Fall</i>	September - October 2021	SGW irrigation, collecting samples and laboratory analysis	pH, TOC, TC, TN EC, metals, ammonium pharmaceuticals and anionic surfactants	Decrease of the saturation from 70% to 30%

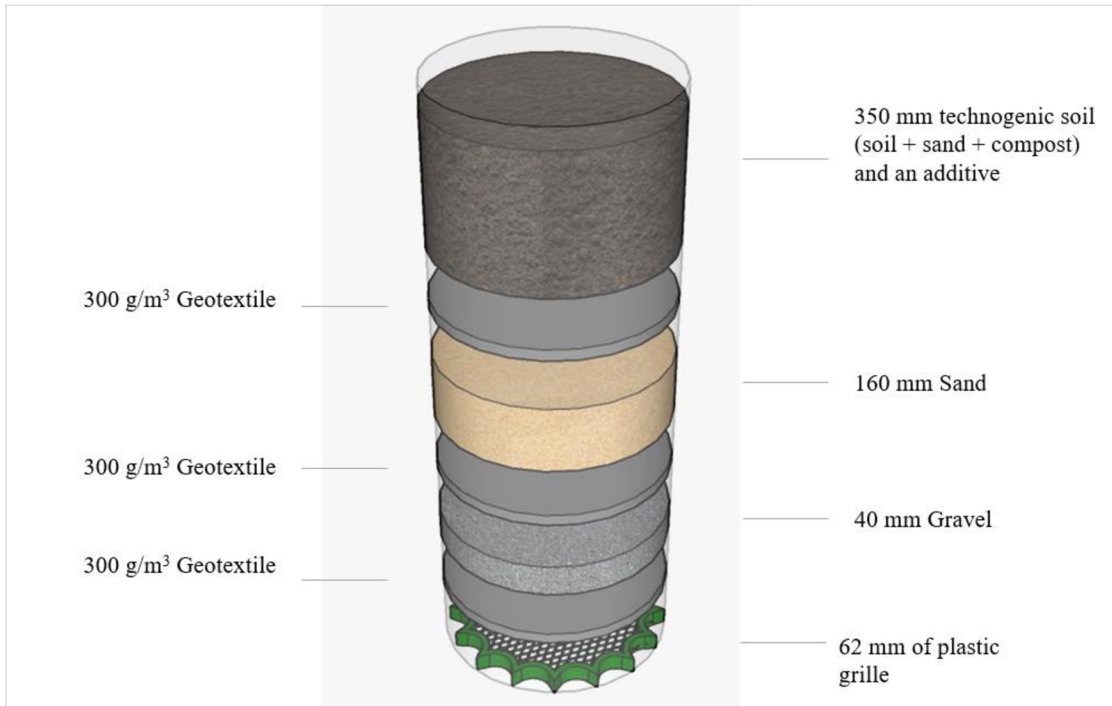


Figure 5: Design scheme of the barrel model.

Table.5 specifies the exact composition and their ratio of the technogenic soil and the additive of the filter media for each group of barrels. Besides the saturation ratios and the number of replicants. The filters media were chosen for the experiment are woodchip, mycorrhiza, biochar5%, biochar 10% (5% and 10% refers to the percentage of the biochar from the whole soil mixture), and the default that used as a control later in the results chapter.

Table 24: Barrels composition and proportions of the soil.

Type of filter media	Soil types (mixing ratio)	saturation volume	Number of replicates
Default	Sand - Compost - Natural Soil (5:3:2)	30%	5
Default	Sand - Compost - Natural Soil (5:3:3)	70%	5
biochar 5%	Sand-Compost-Natural Soil-Biochar (10:5:4:1)	30%	5
biochar 5%	Sand-Compost-Natural Soil-Biochar (10:5:4:1)	70%	5
biochar 10%	Sand-Compost-Natural Soil-Biochar (5:2:2:1)	30%	5
biochar 10%	Sand-Compost-Natural Soil-Biochar (5:2:2:1)	70%	5
Woodchip	Sand-Compost-Natural Soil-Woodchips (5:2:2:1)	30%	5
Woodchip	Sand-Compost-Natural Soil-Woodchips (5:2:2:1)	70%	5
Mycorrhiza	Sand-Compost-Natural Soil-Mycorrhiza (4:3:2:1)	30%	5
Mycorrhiza	Sand-Compost-Natural Soil-Mycorrhiza (4:3:2:1)	70%	5

4.2 Synthetic Greywater preparation

Mainly Synthetic Greywater is composed of technical quality chemical products to simulate organic and inorganic pollution of greywater from the bathroom (pollution from the human's body, body hygienic products, and makeup-related products) (Hourlier et al., 2010). The synthetic greywater is formulated and used for testing the greywater infiltration treatment efficiency. Household Greywater was created synthetically by using materials found usually in household greywater.

The SGW has been prepared according to synthetic greywater recipes from Abed & Scholz (2006) and Diaper et al (2008). The recipe comprised; Ariel Gel for Laundry, Nivea Fresh Natural Roll-on, Colgate Cavity Protection toothpaste, Head and Shoulders shampoo, Nivea Body Milk, and Dove Caring Hand Wash. These products are used in purpose to simulate the

pharmaceuticals and personal care products in cosmetic and household products and to be a source of such compounds such as anionic and nonionic surfactants and soap from the Ariel. The toothpaste is a source of Calcium Carbonate, Zinc Oxide, Bicarbonate, etc. In addition, a couple of solutions were added to the SGW which is contained; metals solution that contains $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ as Copper, H_3BO_3 as trace element boron, $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ as zinc, and $\text{NiSO}_2 \cdot 6\text{H}_2\text{O}$ as nickel, organic compounds; DEET as $\text{C}_{12}\text{H}_{17}\text{NO}$, and diclofenac as $\text{C}_{14}\text{H}_{10}\text{Cl}_2\text{NNaO}_2$. Table 6 shows the amount of the SGW composition in two different quantities for two different saturations, 70%, and 30%.

4.3 Irrigation and sampling process

The experiment has started on the 28th of June, and it has 3 different parts. First, it started by adding tap water to all the barrels to rinse the filtration materials and rinse the barrels from the excess materials and impurities that grow naturally inside them and collecting mixed samples from all the groups. Secondly, a kinetic experiment was aimed to distinguish the efficiency of the Greywater infiltration in different retention times; 2hr, 4hr, 24hr, and 4 days and the samples were collected from each barrel separately for laboratory analysis. The kinetic experiment was conducted as control only. Third, effluent filtration and collecting samples twice a week; Monday 70 hr retention time and Thursday 22 hr operation time, followed by SGW irrigation in two different saturations, 70%, and 30%. However, the hydraulic load volume has changed after the break. The hydraulic load had decreased after the break from 70 to 30% to examine the effect of the hydraulic load decreasing on the efficiency of the infiltration.

Table 25: Synthetic Greywater composition and compounds concentrations

<i>Parameter</i>	<i>The amount for 70% saturation (per 1000L)</i>	<i>The amount for 30% saturation (per 600L)</i>	<i>Unit</i>
<i>PCP</i>			
Shampoo	72	43,2	g
Soup	648	388,8	g
Body Milk	10	6	g
Toothpaste	32,5	19,5	g
Deodorant	10	6	g
Laundry Detergent	150	90	g
<i>Metals</i>			
ZnSO ₄ ·7H ₂ O	0.8	0.8	mg/L
H ₃ BO ₃	5.7	5.7	mg/L
NiSO ₂ ·6H ₂ O	0.8	0.8	mg/L
CuSO ₄ ·5H ₂ O	0.7	0.7	mg/L
<i>Pharmaceuticals</i>	10	6	ml
C ₁₂ H ₁₇ NO	5	5	mg/L
	5	5	mg/L

4.4 Laboratory samples processing and analysis

Sampling is done after draining the effluent first thing in the morning. 2L of effluent were taken from each barrel into a water container that represents the specific group, after collecting samples from all barrels, a 50ml of the mixed sample was taken into a clear centrifugal tube for the chemical analysis and another 10ml to separate tube for the metals analysis later. Table.7 presents the parameters that were tested on effluent and influent samples and the machines used for that purpose. Along with the parameters mentioned in the table, chemical compounds such as Fluoride, Chloride, Bromide, Nitrite,

Nitrate, Phosphate, and Sulphate are tested as a general indication for the water quality. The procedure of the analysis of the samples is **similar** for both affluent, and influent (SGW). The physio-chemical analysis is done right after collecting the samples from the field in the laboratory, starting with filtration of influent and effluent samples using Syrine filters ROTILABO[®] PES, 0,22 µm except for the 10 ml samples of the metals. Followed by pH and conductivity measurement. All the machines used to analyze the samples presents in table 7.

4.4.1 TOC, IC, TC, and TN analysis

The Total Organic Carbon (TOC), Inorganic Carbon (IC), total carbon (TC), and Total Nitrogen (TN) measurements were analyzed by feeding a 15ml test tube of filtered sample into a Skalar Formacs^{HT} TOC/TN Analyzer (fig.6). The Formacs analyze and measure the concentration of nitrogen and carbon fractions in the liquid sample using high-temperature catalytic combustion. The analyzer measure TOC by analyzing TC and IC. TC in the sample is obtained by catalytic oxidation, and the high temperature converts the TC present (organic and inorganic) into Carbon Dioxide (CO₂). Subsequently, the analysis is done by measuring the quantity of Carbon Dioxide formed during the acidification and the TOC is calculated by $TOC = TC - IC$.



Figure 6: Skalar Formacs^{HT} TOC/TN Analyzer

Table 26: Conventional parameter for effluent and influent analysis.

<i>Parameter</i>	<i>unit</i>	<i>Machine</i>
<i>pH</i>	mg/l	WTW Lab-pH Meter inoLab® pH 7110
<i>conductivity</i>	µg/l	Conductivity benchtop inoLab® cond7110
<i>N-N</i>	mg/l	Spectrometry - Agilent Technologies Cary
<i>T-P</i>	mg/l	60 UV-Vis
<i>TOC</i>	mg/l	SKALAR Formacs ^{HT} TOC/TN Analyzer
<i>TC</i>	mg/l	
<i>IC</i>	mg/l	
<i>TN</i>	mg/l	
<i>Tenzides</i>	mg/l	
<i>Heavy Metals</i>		
<i>Zn</i>		HACH DR3900 spectrophotometer
<i>B</i>	mg/l	
<i>Cu</i>		
<i>Ni</i>		
<i>Pharmaceuticals</i>		(HPLC) Coupled with a Diode Array
<i>Diclofenac</i>	mg/l	
<i>DEET</i>		

4.4.2 Anions

The Anions analysis contains Fluoride, Nitrate, Nitrite, Phosphate, sulphate, and Chloride. The analysis is done by loading a filtered sample to a 15 ml plastic tube and feeding it into the Ionic chromatography system MagIC NET™. These anions concentrations indicate the general water quality.

4.4.3 Anionic Surfactants (Tenzides)

Anionic surfactants analysis is done by LCK332 0.05-2.00 mg/L cuvette test (Hang Lange) (fig.7.A), which contains solution A and solution B and prepared cuvettes. By adding the sample, the anionic surfactants react with methylene blue to form complexes (fig.7.B), which are extracted in chloroform and evaluated photometrically using HACH LANG DR3900 spectrophotometer. Since the measurement range was between 0.05 – 2.00 mg/L, higher concentration samples were diluted, and the synthetic greywater sample was diluted 50 times due to the high concentration of surfactants in it.

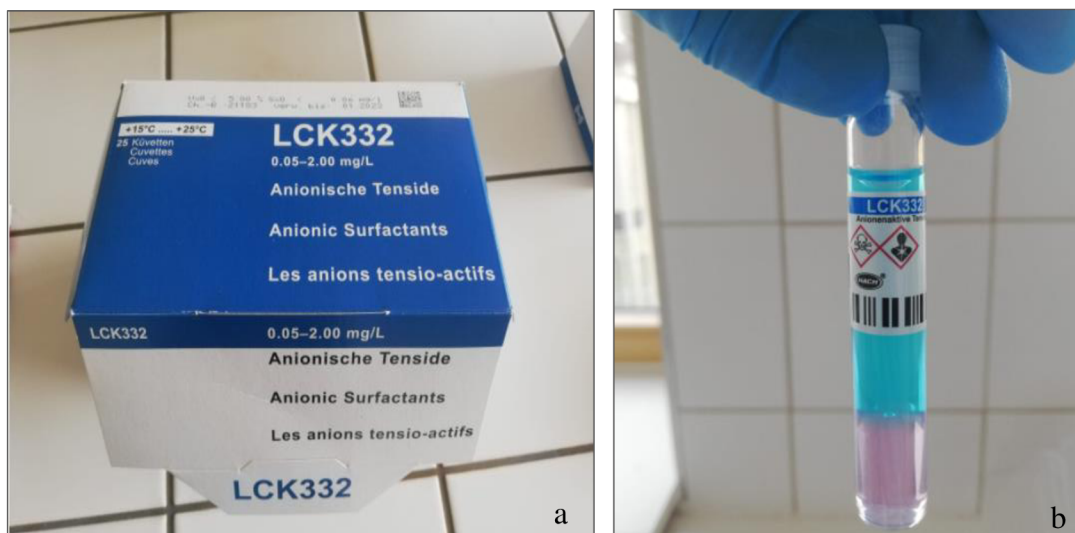


Figure 7: a) LCK332 cuvette test for surfactant analysis. b) prepared cuvette for measurement.

4.4.4 Ammonia Analysis

Regarding Ammonia analysis preparations, a 4ml filtered sample was added 0.4ml of preprepared coloring agent, 0.4ml of alkaline solution, and 0.2 of distilled water (fig.8), blank sample was also prepared using 4 ml of distilled water instead of filtered sample and used as a control. The accepted concentration range is between 0.01 – 1 mg/L. Therefore, samples with possibly higher concentrations were diluted 3 times. The prepared samples were placed for 1 hr in the dark before the evaluation. The results were analyzed using Angilent Cary 60 UV-Vis with wavelength 190 – 1100 nm.

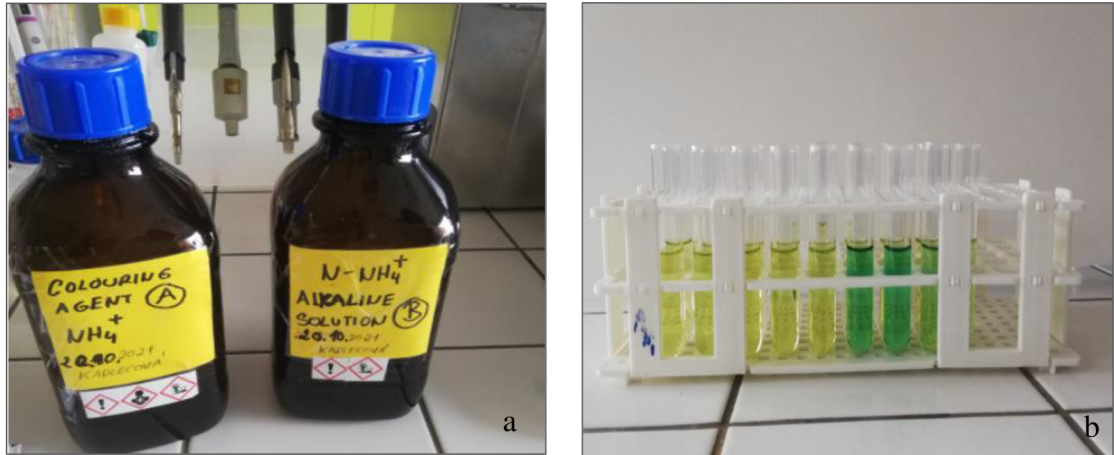


Figure 8: a) Solutions were added to the samples. b) prepared samples before measurement.

4.4.5 Metals Analysis

To analyze the metals in the synthetic greywater and the effluent, 10 ml of the well-mixed sample that presents each group of barrels was taken into a 15 ml tube and applied also for synthetic greywater. For both types of samples were added 250 $\mu\text{g/L}$ of Nitric acid (HNO_3^-) to avoid adsorption to the tube walls and the microbial degradation is minimized. Also, by adding Nitric acid to the samples, it converts metal ions into their highly soluble Nitrate salts. Afterward, the samples were sent for measurement. The samples of the metals were analyzed by ICP-OES, Agilent 5800 (Inductively Coupled Plasma Atomic Emission spectroscopy).

4.4.6 Pharmaceuticals Analysis

Pharmaceutical analysis was done using High-Performance Liquid Chromatography (HPLC) combined with Thermo Scientific™ Dionex™ Ultimate™ 3000 (fig.9). All the filtered samples were inserted into the HPLC on the same day or after overnight storage in the fridge away from the light in purpose to avoid a degradation and microorganisms' growth in the tube of any of the pharmaceutical materials.



Figure 9: HPLC combined with Thermo Scientific™ Dionex™ Ultimate™ 3000

4.5 Statistical analysis and calculations

The data obtained from the laboratory analysis were marched in an MS OFFICE Excel sheet. For the data analysis, RStudio Version 3.6.1 was used for testing and plotting the outputs. The statistical testing was done using ANOVA. In case the data did not meet the normality, statistical analysis was calculated by Kruskal-Wallis tests, which is a one-way analysis of ANOVA, and the test indicates that at least one sample stochastically dominates one other sample. The plots aim to show the efficiency of the additives to remove the specific chemical parameter

The removal ffoverboldas calculated using the formula:

$$E = \frac{C_{in} - C_{total}}{C_{in}} * 100$$

Where:

C_{in} – influent concentration (mg/l).

C_{total} – effluent concentration (mg/l).

E – removal efficiency.

5 Results

5.1 TC, IC, and TOC analysis

The TOC removal efficiency in summer for 70% saturation for default, woodchip, mycorrhiza, biochar 5%, and biochar 10% were 33.41, 36.68, 45.20, 56.65 and 67.18 % respectively (fig.9-a). While for 30% saturation the values are 36.03, 38.94, 41.60, 56.83, 73.53% respectively (fig.9-b). The biochar 10% achieved 67.18% and 73.53% removal efficiency, which was the highest treatment efficiency of TOC among both saturation levels, 70 and 30%. The mycorrhiza showed the lowest performance for 30% saturation. The ANOVA of TOC removal efficiency of the different filter media revealed a significant difference between the efficiencies mean of all the filters media (P.value<0.05), which means that there is a dependency between the biofilter type and the removal efficiency of TOC. The TC and IC results (see appendix 1) explain TOC removal by oxidizing the organic carbon to inorganic carbon and as a result, a higher concentration of IC and subsequently TC were found.

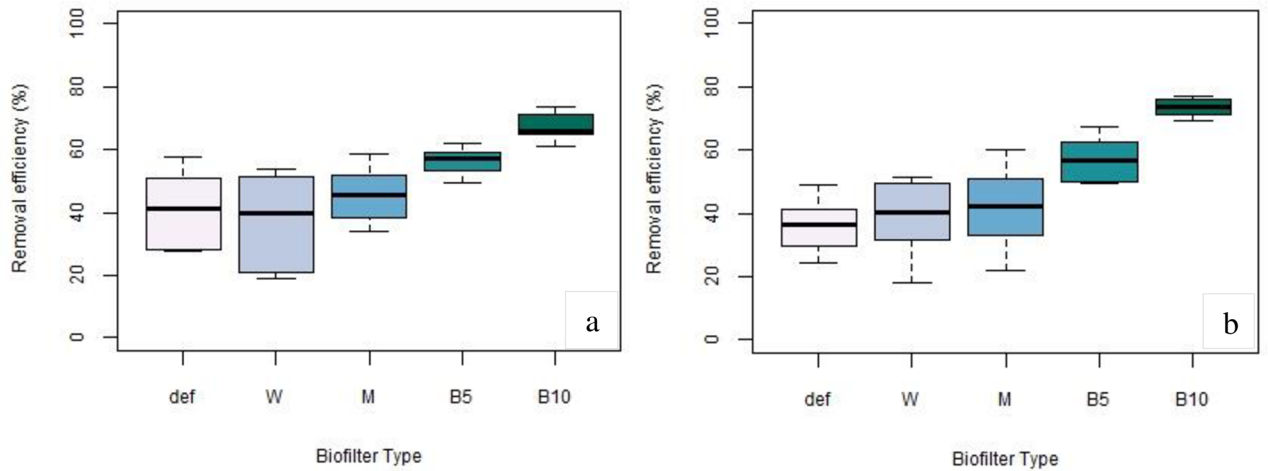


Figure 10: TOC removal efficiency in the summer season of all filters media for two different saturation levels; a) 70, and b) 30%.

Figure 10 represents the TOC removal efficiency after decreasing the 70% to 30% hydraulic load (fig.10 – a) and 30% without a change with the hydraulic load (fig10 – b). Decreasing the hydraulic load caused a higher performance among all biofilters, where the mean values of default, woodchip, mycorrhiza, biochar 5%, and biochar 10% were 68.39, 67.39, 69.75, 73.35, 78.92 % respectively. In addition, the biofilters showed a higher performance than the TOC removal efficiency for 30%

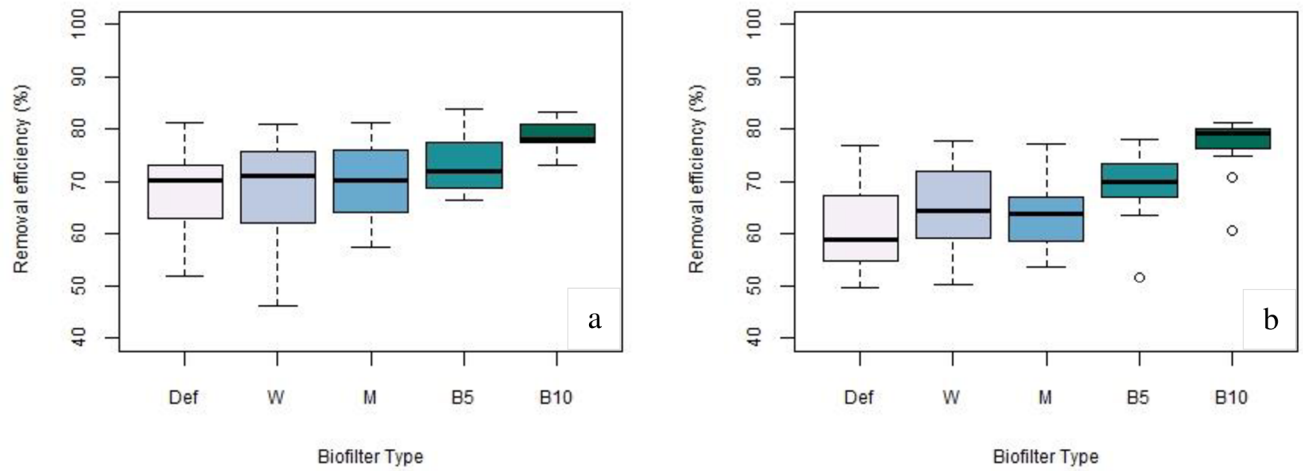


Figure 11: TOC removal efficiency in the fall season of all filters media for two different saturation levels; a) decrease from 70% to 30%, and b) 30%.

5.2 TN, Nitrate, and Ammonium removal.

The removal efficiency of TN is given in Figure 12. Biochar 10% showed the highest efficiency with an average removal of 80.81 and 89.80% for 70% and 30% hydraulic load. While the default showed the lowest efficiency of, 66.86% for 70% and 68.08 for 30%. Figure 12 – b revealed that lower hydraulic load leads to better removal efficiency among all the filters. The ANOVA test revealed absolute dependency between the biofilter type and the removal efficiency since the biofilter performance was significantly different from each other (T-test, P.value < 0.05).

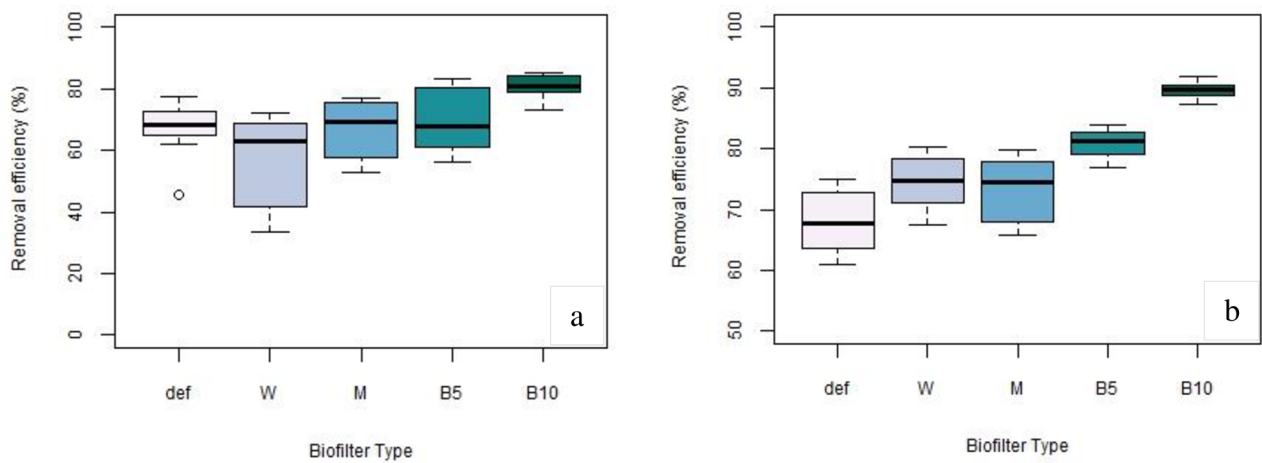


Figure 12: TN removal efficiency in the summer season of all filters media for two different hydraulic loads; a) 70% and b) 30%

Like TOC, the decrease in the hydraulic load from 70% to 30% affected the system positively (Fig.13-a). Compared to Figure.11-a, the removal efficiency increased to an average of 79.95% for default, 70.44% woodchip, 81.63% mycorrhiza, 81.24% biochar 5%, and 88.34% biochar 10%. Figure 13-b of 30% saturation during the fall shows higher performance compared to 30% in the summer and slightly higher values compared to the decrease from 70 to 30%. ANOVA test revealed dependency between the biofilter type and the removal efficiency of TN (T-test, P.value < 0.05).

Ammonium concentration showed instability. Moreover, the removal efficiency of ammonium showed negative values which means inefficiency in the ammonium removal. According to the data obtained from the nitrate and nitrite concentration, it indicates a low concentration of nitrite (<LOQ), and a slightly higher concentration of nitrate. Which can explain the ammonium instability.

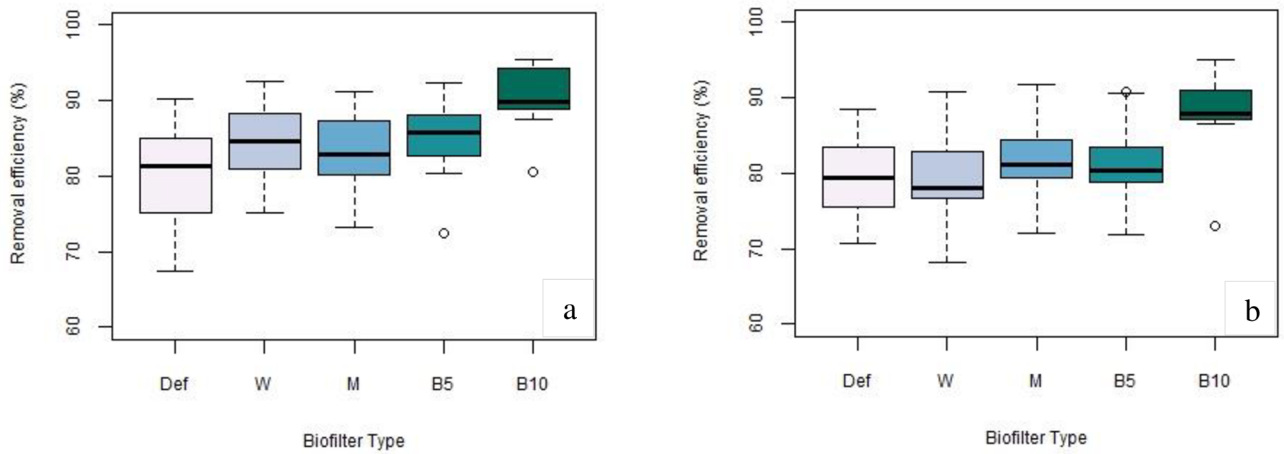


Figure 13: TN removal efficiency in the fall season of all filters media for two different hydraulic loads; a) decreasing from 70 to 30%, and b) 30%.

Figure 14 shows the nitrate concentration in saturations. Nitrate is an important supplement for plants. And it takes a major place in nitrification and denitrification processes. The average Nitrate concentration for the SGW was 26.24 mg/l. According to the results, for 70% and 30% in the summer, the nitrate concentration is unstable. After the 6th sample, the nitrate concentration in 70% saturation was higher than 30% saturation.

Nitrate concentration for the fall season for 30% and the decrease from 70 to 30% is shown in Figure 15. By changing the saturation from 70 to 30% in the fall, the filter materials showed a high concentration on the first day of the experiment and then the concentration drops between the 2nd until the 7th sample. The highest concentration is for biochar 5% and the lowest is for biochar 10% with 5.44 and 0.03 mg/l respectively. Overall, the lowest concentration is for biochar 10% with an average of 1.18 mg/l. The biochar 10% succeed to decrease the concentration from 3.34 mg/l on day 1 to 1.7 mg/l on the last day. No significant difference was observed between 30% in the summer and 30% in the fall.

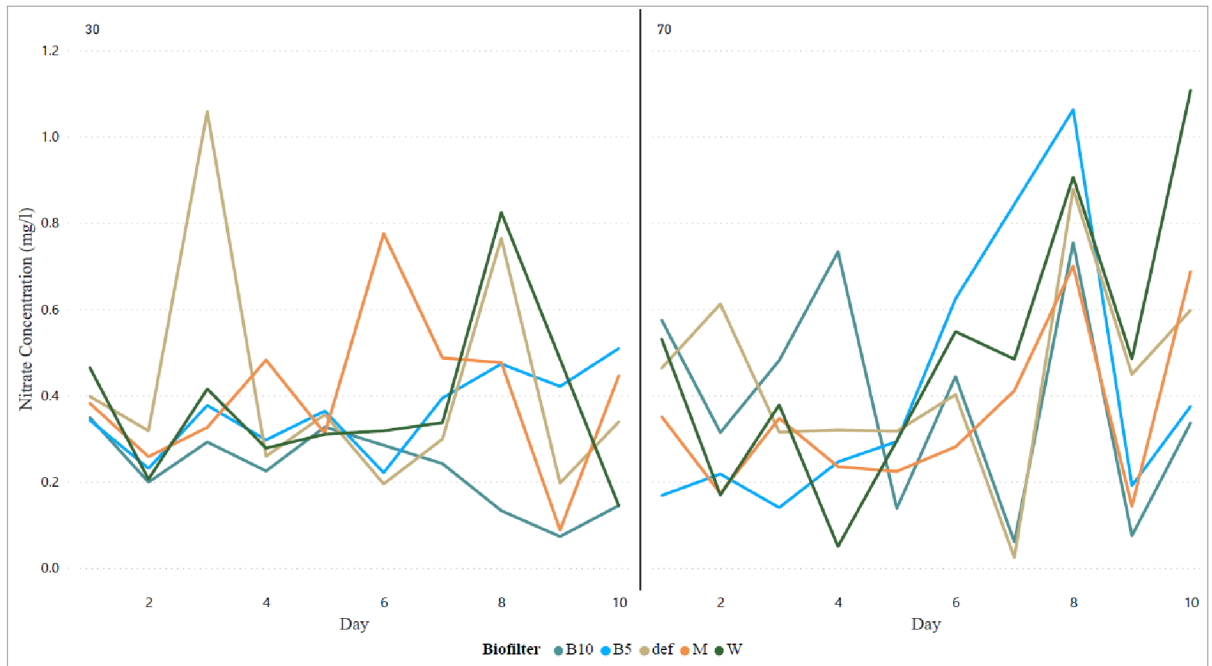


Figure 14: Nitrate effluent concentration for all the biofilters in the summer for 30% and 70% saturation.

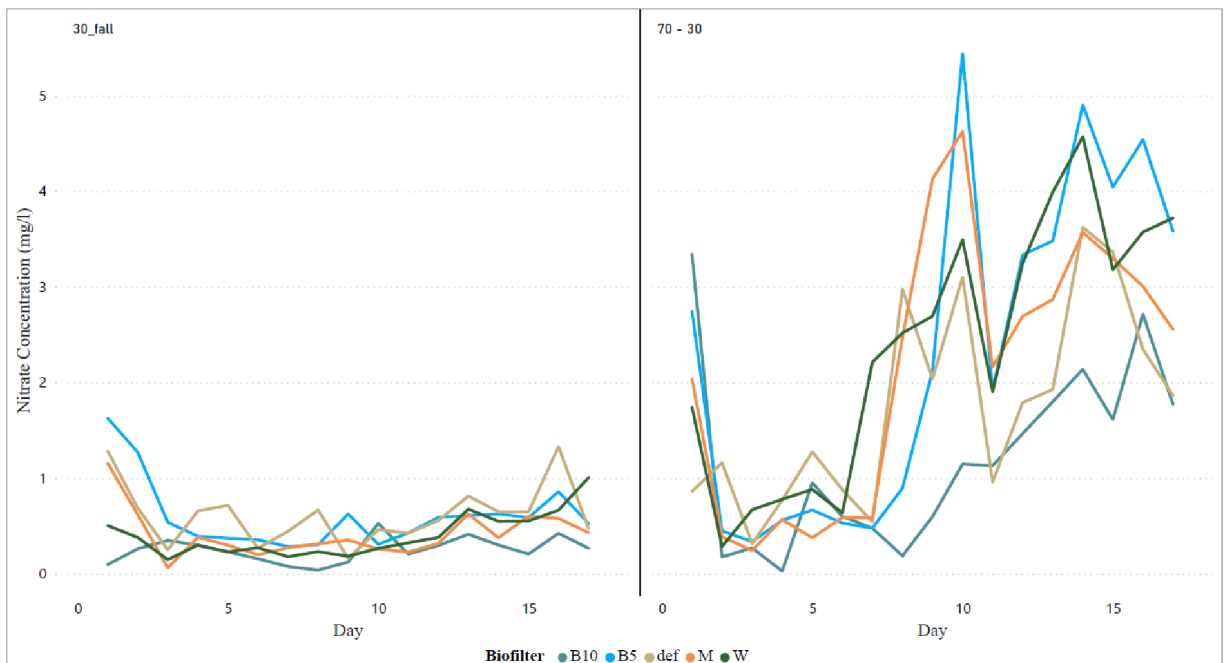


Figure 15: Nitrate effluent concentration for all the biofilters in the fall for 30% and the decrease from 70% - 30 % saturation.

The biofilters did not show efficiency in removing the ammonium from the influent where the average concentration in the SGW was 0.065 mg/l. The removal efficiency calculations showed a negative value among all the biofilters as well as the saturation level.

Figure 16 represents the ammonium concentration in the summer for 70% and 30% saturation, and the SGW concentration with a dashed line, 0.065 mg/l. For 70% saturation, none of the biofilters succeed to reduce the ammonium concentration so the concentration was higher than 0.065 mg/l. Furthermore, it shows an increase in the trend from the 2nd day until the last day of the experiment. However, it is noticeable that for 30 % saturation the ammonium concentration is lower than for 70% saturation. Additionally, as Figure 13 and Figure 12, the biochar 10% has a higher performance with an average concentration of 0.04 mg/l. While the highest concentration is for default with 0.15 mg/l.

Coupled with the removal efficiency calculation, it showed that the only biofilter that kept the removal efficiency in the positive range is the biochar 10% in 30 % saturation.

Figure 17 represents the Ammonium concentration for the fall season for 30% saturation and the decrease in the saturation level from 70 to 30% saturation, coupled with the ammonium concentration in the SGW with a dashed line on 0.065 mg/l. According to the results, the decrease in the saturation level did not improve the ammonium removal, and only after the 9th day only the biochar 10% succeed to reduce the concentration. On the other hand, it is clearly shown that there is a decrease in the ammonium concentration of all the biofilters during the fall in 30% saturation. Moreover, the lowest concentration of biochar 10% effluent with an average of 0.02 mg/l followed by mycorrhiza and woodchip of 0.048 mg/l and 0.047 mg/l respectively, and the last is biochar 5% and default of 0.05 mg/l and 0.062 mg/l respectively.

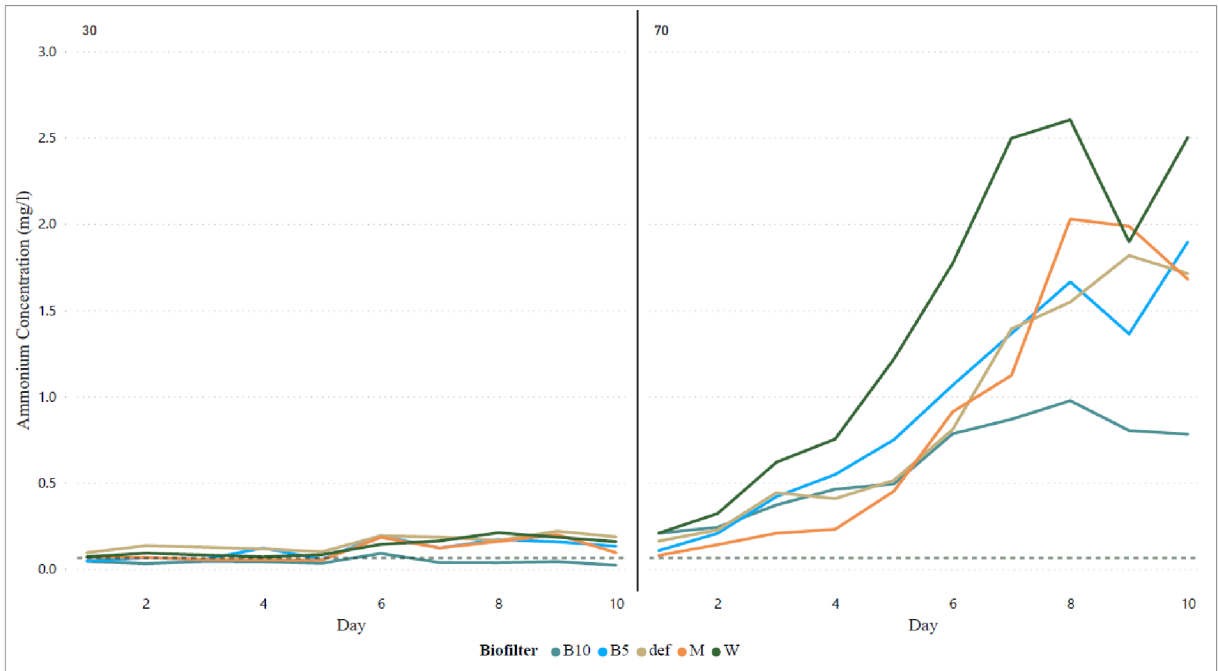


Figure 16: Ammonium effluent concentration for all the biofilters in the summer for 30% and 70% saturation.

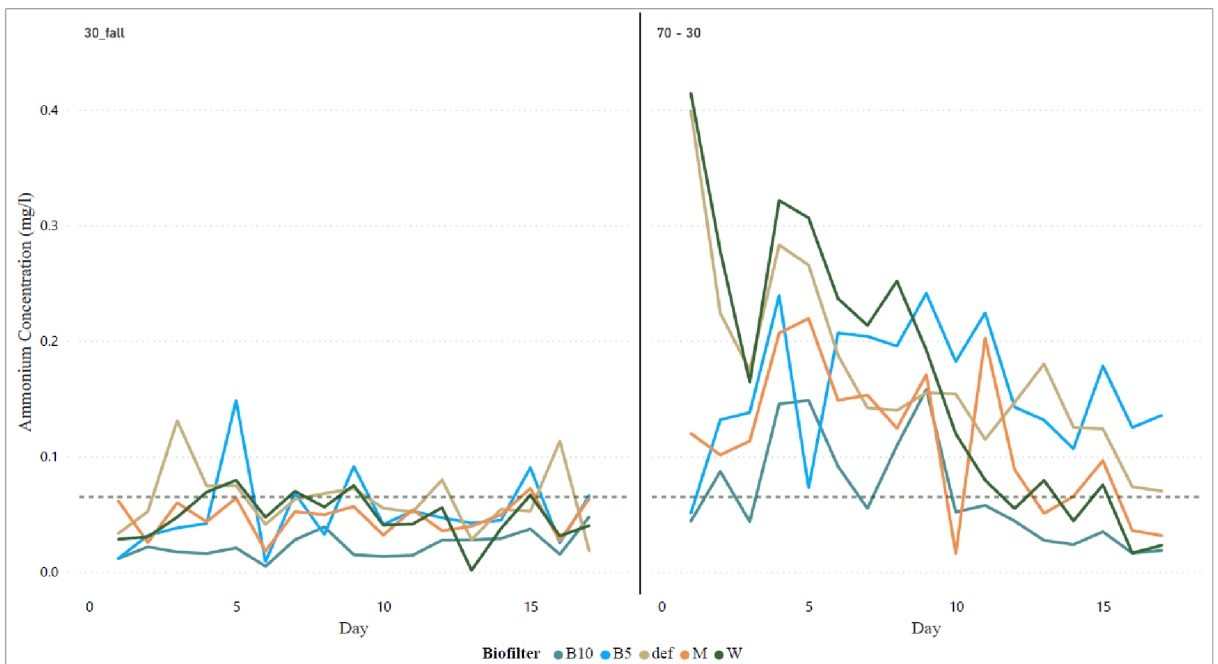


Figure 17: Ammonium effluent concentration for all the biofilters in the fall for 30% and the decrease from 70% - 30 % saturation.

5.3 Anionic Surfactants Removal Efficiency

Anionic surfactants removal efficiency is shown in Figure 18. At 70 and 30% saturation levels in the summer, all the filters showed a high efficiency in the anionic surfactants' removal which range between 97.58-99.15% for 70% saturation, and 98.92-99.15% for 30% saturation. The highest biofilter performance with a slight difference from the others goes to biochar 5% in both saturation levels.

Decreasing the saturation level from 70-30% (Fig.19-a) did not affect the removal efficiency of the anionic surfactants. The same applied for 30% saturation (Fig.19-b).

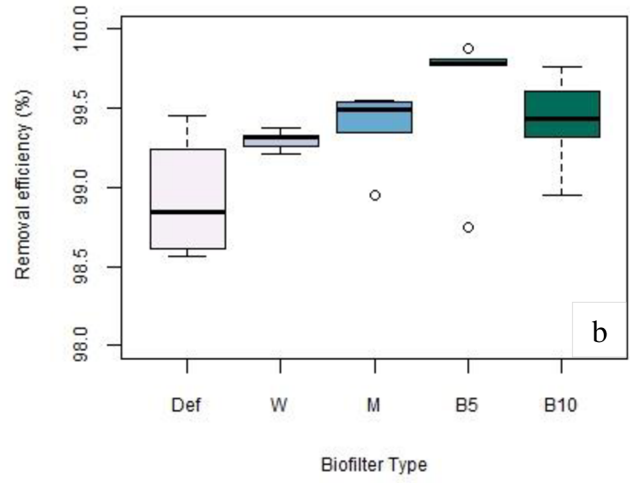
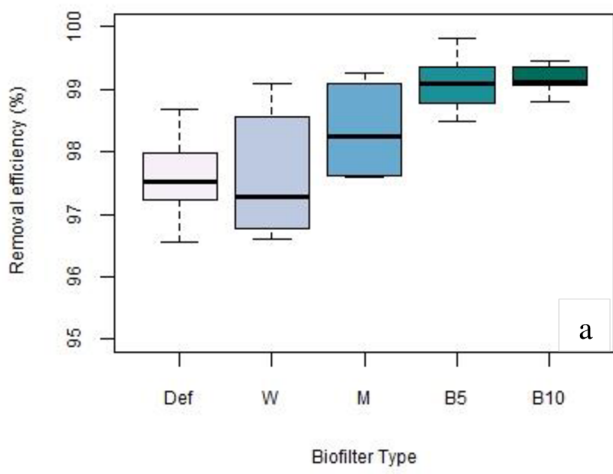


Figure 18: Surfactant's removal efficiency in the summer for all the filters media in two saturation levels; a) 70% and b) 30%.

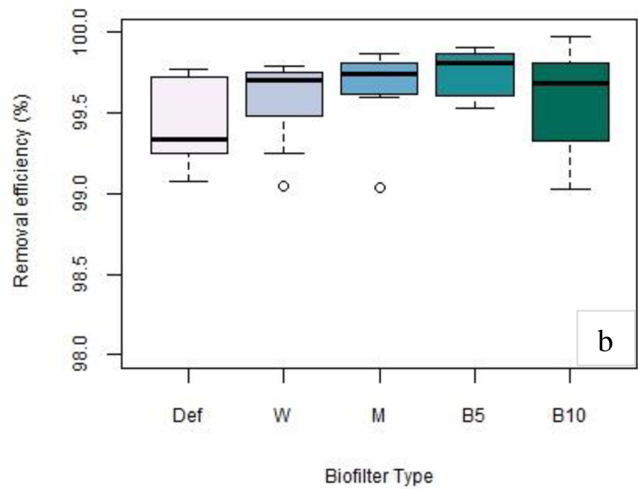
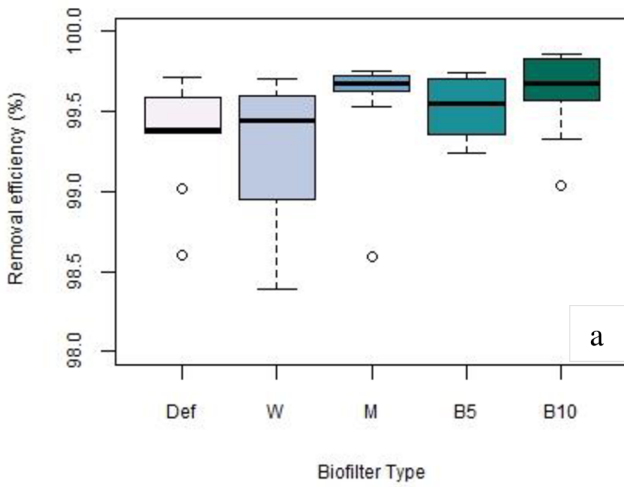


Figure 19: Surfactant's removal efficiency in the fall for all the filters media in two saturation levels; a) 70 - 30% and b) 30%.

5.4 Metal Removal Efficiency

Regarding the metals and the boron concentration in the effluent and the influent, it is presented in Table 27 which contains the mean value and standard deviation (SD), minimum concentration (min), maximum concentration (max), and under the detection limit (<LOQ). The trace element, boron, showed insufficient removal from the effluent. The mean value of boron concentration of the influent is 0.758 mg/l, all the filters' media showed higher concentration than the influent one. Consequently, the removal efficiency percentage was mostly negative during the experiment. The highest positive boron removal efficiency is 49.6% for biochar 10. The negative removal efficiency of boron can be caused by leakage from the filter materials to water and as a result, an increase in the boron concentration in the effluent compared with the influent. According to Zaman et al (2018), to avoid toxicity for plants, the boron concentration in the irrigation water should be lower than 0.3 mg/L.

Table 27: Metals concentration in the effluent and the influent.

<i>Filter media</i>		<i>Metals concentration (mg/l)</i>			
		<i>Boron</i>	<i>Zinc</i>	<i>Copper</i>	<i>Nickel</i>
SGW	<i>Mean ± SD</i>	0.758 ± 0.4	0.176 ± 0.09	0.156 ± 0.08	0.147 ± 0.08
	<i>Min</i>	<LOQ	<LOQ	<LOQ	<LOQ
	<i>Max</i>	1.107	0.28	0.23	0.21
Def	<i>Mean ± SD</i>	1 ± 0.12	0.01 ± 0.01	0.009 ± 0.006	0.009 ± 0.004
	<i>Min</i>	0.62	<LOQ	<LOQ	<LOQ
	<i>Max</i>	1.189	0.06	0.0205	0.0205
W	<i>Mean ± SD</i>	0.95 ± 0.2	0.013 ± 0.016	0.012 ± 0.008	0.011 ± 0.014
	<i>Min</i>	<LOQ	<LOQ	<LOQ	<LOQ
	<i>Max</i>	1.27	0.051	0.051	0.105
M	<i>Mean ± SD</i>	0.99 ± 0.17	0.01 ± 0.013	0.01 ± 0.001	0.009 ± 0.004
	<i>Min</i>	0.51	<LOQ	<LOQ	<LOQ
	<i>Max</i>	1.39	0.051	0.03	0.02
B5%	<i>Mean ± SD</i>	0.98 ± 0.11	0.01 ± 0.018	0.01 ± 0.008	0.007 ± 0.004
	<i>Min</i>	0.63	<LOQ	<LOQ	<LOQ
	<i>Max</i>	1.2	0.061	0.03	0.01
B10%	<i>Mean ± SD</i>	0.96 ± 0.15	0.02 ± 0.02	0.009 ± 0.006	0.009 ± 0.002
	<i>Min</i>	0.48	<LOQ	<LOQ	<LOQ
	<i>Max</i>	1.189	0.061	0.02	0.01

Additionally, the filters media successfully removed the metals zinc, copper, and nickel. According to the results, the metals concentration range between under the detection limit and 0.28 mg/l. The results did not record any significant difference in metals concentration during the experiment timeline. Table 28 presents the metals removal efficiency for the different filters' media. Furthermore, according to the results, the saturation level does not affect the removal efficiency. The lowest value is the zinc removal efficiency for biochar 10%, 90.67%.

Table 28: Metals removal efficiency for different filters media.

<i>Filter Media</i>	<i>Removal Efficiency (%)</i>		
	<i>Copper</i>	<i>Nickel</i>	<i>Zinc</i>
<i>Default</i>	95.05	94.75	94.14
<i>Woodchip</i>	93.51	93.75	93.65
<i>Mycorrhiza</i>	94.56	95.15	95.15
<i>Biochar 5%</i>	94.31	95.93	91.75
<i>Biochar 10%</i>	95.3	94.89	90.67

5.5 Phosphorus Removal Efficiency

The last important indicator is phosphorus. Phosphorus is playing a major role in plant and algae growth in surface water. Figures 20 and 21 represent the phosphorus concentration for all the biofilters in the different saturation levels and the concentration in the SGW with a dashed line and its 0.241 mg/l. According to the results, it shows a challenge in reducing the phosphorus from the greywater for the different biofilters. Results of the phosphorus effluent for 70% and 30% in the summer are shown in Figure 20.

In the summer, 70% saturation shows an increase of the phosphorus effluent after the first day of the experiment among all the biofilters. While 30% showed better performance, especially for biochar 10% and woodchips. For biochar the concentration was lower than 0.241 mg/l after the peak on the first day of the experiment, in which the concentration reached 2.16 mg/l, also woodchip showed a low concentration between the 3rd and 8th sample.

In the fall, the decrease from 70 to 30% saturation (Figure 21) reduced the concentration, and after the 9th day, the concentrations were lower than 0.24 mg/l among the biofilters. For 30% saturation in the fall, the best performance was for biochar 10% and woodchip where both biofilters removed the phosphorus successfully, while biochar 5% showed the lowest performance with an average of 0.54 mg/l.

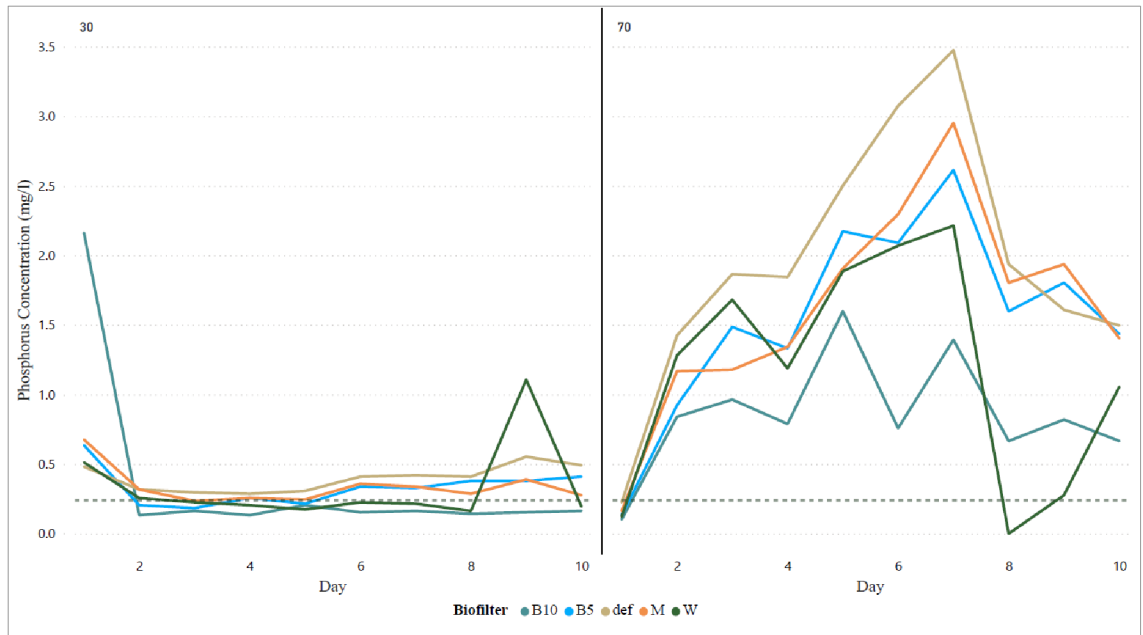


Figure 20: Phosphorus effluent concentration for all the biofilters in the fall for 30% and 70% saturation.

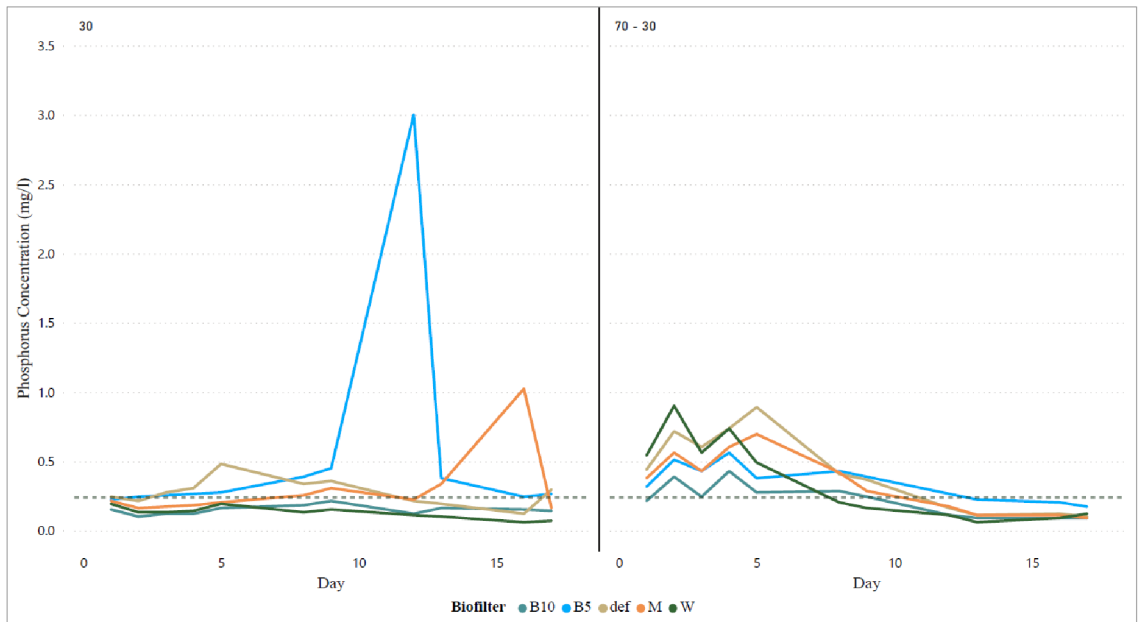


Figure 21: Phosphorus effluent concentration for all the biofilters in the fall for 30% and the decrease from 70% - 30 % saturation.

5.6 Organic Compounds Removal efficiency

Regarding the Organic compounds Removal efficiency and the biofilters' behavior, it was tested. The analysis was carried out 4 times during the experiment due to expenses and complexity reasons. Table 10 represents the removal efficiency averages for DEET and Diclofenac for two saturation levels, 70 and 30%.

According to the results of the DEET removal efficiency, the saturation level affected differently the removal efficiency of the different biofilters where biochar 5%, default, and mycorrhiza has higher removal efficiency under 30% saturation, while biochar 10% and woodchip has higher removal under 70%. Overall, the biochar 5% has the higher performance with 91.6 % and the lowest is default with 50.9%.

The diclofenac removal efficiency clearly shows that 30% saturation has higher removal efficiency than 70%. Moreover, in both saturations, the biochar 5% has the highest performance while biochar 10% has the lowest.

Table 29: Organic compounds removal efficiency after 1-day retention for all biofilters in two saturation levels.

		<i>DEET</i>	<i>Diclofenac</i>
	<i>Saturation (%)</i>	<i>Removal Efficiency %</i>	
<i>Default</i>	70	50.9	82.7
	30	81.3	84.3
<i>Woodchip</i>	70	66.2	83.5
	30	64.2	94.2
<i>Mycorrhiza</i>	70	54.4	78.8
	30	79.7	94.7
<i>Biochar</i> <i>5%</i>	70	71.5	85.2
	30	91.6	98.1
<i>Biochar</i> <i>10%</i>	70	85.2	71.8
	30	66.9	74.4

6 Discussion

This chapter is describing and discussing the results were recorded during the experiment and presented in the previous chapter. In purpose to have detailed discussion, elements such as rain and its consequences are being considered.

6.1 Summary of the data

Table 30 presents a summary for chemical analysis that contains pH, Electrical Conductivity (EC), Total organic carbon (TOC), Inorganic Carbon (IC), Total Carbon (TC), Total Nitrogen (TN), anionic surfactants, heavy metals (Boron is a trace element, Zinc, Copper, and Nickel), ammonium nitrogen ($\text{NH}_4\text{-N}$), and phosphorus for 5 different biofilters Default, Woodchip (W), Mycorrhiza (M), Biochar 5%, and Biochar 10%. The average pH values for all the biofilters lie within the optimal range of 6.5 – 8.4 of water quality for irrigation (EPA, 2012).

The highest EC value was reported for the effluent of Default barrels of 967 $\mu\text{S/cm}$. While the lowest value was 475 $\mu\text{S/cm}$ for Mycorrhiza. All the values of EC reported for all the samples through the experiment lie within the EC standards for water quality for irrigation (EPA, 2012).

The lowest values of the analyzed chemical parameters TOC, IC, TC, TN, $\text{NH}_4\text{-N}$, anionic surfactants, Boron, Zinc, Copper, Nickel, and phosphorus were 17.1, 62.7, 79.8, 0.98, 0.14, 0.46, 0.96, 0.02, 0.009, 0.009, and 0.38 (mg/l) respectively refers to biochar 10%.

Table 30: Summary of the effluent data of the different filter's media (average \pm standard deviation)

<i>Parameter</i>	<i>Unit</i>	<i>SGW</i>	<i>Default</i>	<i>Woodchip</i>	<i>Mycorrhiza</i>	<i>Biochar 5%</i>	<i>Biochar 10%</i>
<i>pH</i>		7.344 \pm 0.116	7.00 \pm 0.09	6.9 \pm 0.13	6.9 \pm 0.12	7.1 \pm 0.1	7.13 \pm 0.09
<i>Conductivity</i>	$\mu\text{S/cm}$	391 \pm 18.82	775.24 \pm 107.3	653.1 \pm 105.70	675.5 \pm 93.3	730.5 \pm 94.3	684.8 \pm 62.6
<i>TOC</i>	<i>mg/L</i>	69.19 \pm 12.88	32.13 \pm 14.06	30.7 \pm 11.7	29.2 \pm 10.01	23.4 \pm 6.2	17.1 \pm 3.9
<i>IC</i>	<i>mg/L</i>	13.64 \pm 1.52	73.83 \pm 18.9	59.7 \pm 19.2	62.3 \pm 16.7	69.6 \pm 14.8	62.7 \pm 10.4
<i>TC</i>	<i>mg/L</i>	82.82 \pm 12.36	105.97 \pm 29.08	90.4 \pm 29.1	91.6 \pm 25.1	93.07 \pm 19.4	79.8 \pm 12.3
<i>TN</i>	<i>mg/L</i>	8.155 \pm 1.17	2.01 \pm 0.7	1.96 \pm 0.9	1.79 \pm 0.7	1.61 \pm 0.64	0.98 \pm 0.14
<i>NH₄⁺ - N</i>	<i>mg/L</i>	0.065 \pm 0.03	0.27 \pm 0.4	0.35 \pm 0.64	0.23 \pm 0.44	0.26 \pm 0.41	0.14 \pm 0.24
<i>Anionic Surfactants</i>	<i>mg/L</i>	90.08 \pm 24.69	0.932 \pm 0.17	0.83 \pm 0.77	0.59 \pm 0.55	0.42 \pm 0.32	0.46 \pm 0.28
<i>Heavy Metals</i>							
<i>Boron</i>	<i>mg/L</i>	0.95 \pm 0.09	1 \pm 0.12	0.95 \pm 0.2	0.99 \pm 0.17	0.98 \pm 0.11	0.96 \pm 0.15
<i>Zinc</i>	<i>mg/L</i>	0.22 \pm 0.62	0.01 \pm 0.01	0.013 \pm 0.016	0.01 \pm 0.013	0.01 \pm 0.018	0.02 \pm 0.02
<i>Copper</i>	<i>mg/L</i>	0.19 \pm 0.04	0.009 \pm 0.006	0.012 \pm 0.008	0.01 \pm 0.001	0.01 \pm 0.008	0.009 \pm 0.006
<i>Nickel</i>	<i>mg/L</i>				0.009 \pm	0.007 \pm	
		0.18 \pm 0.03	0.009 \pm 0.004	0.011 \pm 0.014	0.004	0.004	0.009 \pm 0.002
<i>Phosphorus</i>	<i>mg/L</i>	0.3 \pm 0.1	0.74 \pm 0.81	0.48 \pm 0.58	0.63 \pm 0.67	0.68 \pm 0.71	0.38 \pm 0.44

6.2 TOC removal

Organic carbon is such an important parameter for greywater treatment that needed to be considered to ensure safe reuse of the water. Reducing organic carbon to low levels is so important to; first, prevent the regrowth of pathogens during storage and distribution. Second, to ensure reliable disinfection performance. And third, to prevent the negative aesthetic effects of carbon degradation after treatment (Hess & Morgenorth, 2021).

According to the results in Figures 9 and 10, the decrease in the saturation level affected positively the system and allowed better performance for the biofilters. During the fieldwork, it was noticeable that the barrels with 70% have flooding in some of the barrels when the SGW were added to them. By decreasing the saturation from 70% to 30%, it created a more aerobic environment that allows more oxygen to penetrate and oxidize the organic carbon to inorganic carbon such as CO₂ as mentioned by Hess et al, (2021) in their study. However, according to Gomez, (2021), the decrease in the hydraulic load did not affect positively the TOC removal efficiencies by the different biofilters.

Moreover, the increase in the removal efficiency of TOC for 30% in the fall can be explained by the rain according to the data, the summer of 2021 was identified as a rainy summer which also caused more floods in the barrel and clogging which enhances the anaerobic environment in the system. As mentioned by Hess et al, (2021) in their study about the TOC removal by adsorption and biodegradation that the removal of the TOC affects by the volume of the inlet where the adsorption capacity is exhausted, and more biodegradation by bacteria is required for TOC removal.

6.3 Nitrogen compounds removal

The system has achieved a high TN removal rate in the summer for both saturation of 70% and 30%. When compared to the fall season of the experiment, the removal efficiency was higher in the fall, especially when in the barrels where the saturation has been decreased from 70 to 30%. For 30% saturation in the summer and the fall, the efficiency has slightly increased in

the fall. In the summer, the nitrate effluent concentration of 70% and 30% were similar where the concentration ranged between 0.02 – 1.1 mg/l and 0.07 – 1.05 mg/l respectively. It is important to mention that according to Figure 14 in 70% saturation it is hard to observe a trend throughout the experiment or to determine the highest or the lowest biofilter performance. While in 30% saturation it is noticeable that biochar 10% has the highest performance and the concentration has been decreased from 0.35 mg/l on the first sample (July) to 0.15 mg/l in the last sample (mid-August).

While during the fall samplings, the drop from 70% to 30% saturation has been followed by a significant increase in the nitrate effluent concentration (Figure 15), the drop on the second day and the increase after can be explained by adaptation of the system to new hydraulic conditions.

The ammonium effluent concentration is inverse with the nitrate effluent concentration. In summer for 70% (Figure 16), the ammonium concentration was higher than the nitrate, while in 30%, the ammonium concentration was lower than the nitrate concentration, and the lowest concentration is of biochar 10%. In the fall (see Figure 17), the decrease of the saturation level has been followed by a decrease in the ammonium effluent concentration; however, the nitrate concentration has increased which can be explained by a nitrification process that oxidizes the ammonium to nitrate to the oxygen penetration the system after decreasing the saturation. The highest performance is of biochar 10% with 0.01 mg/l. Furthermore, for 30% saturation in the fall the ammonium concentration is lower than the nitrate, where the ammonium concentration ranges between 0.001 and 0.14 mg/l.

The nitrite concentration is under the limit detection which can indicate of a nitrification process that occurred in the barrel. Further, the nitrate concentration can be explained by the nitrification process which the nitrite is an intermediate product of the process, the reason that the nitrite is under the limit of detection in the effluent. During the summer, it was observed rain during July and August which is reflected in the high moisture in the barrels, the reason where some of the barrels were flooded and clogged. Consequently, these conditions enhanced the anaerobic environment in addition to 70%

saturation in the barrels. Therefore, it is clearly in Figure 14 and Figure 16 for 70% saturation, that the ammonium effluent concentration is slightly higher compared to the nitrate which means that nitrate is converted to ammonium in a process called nitrate ammonification.

As discussed, (Yang et al., 2022) in their study about the nitrogen removal from rural domestic wastewater, under anaerobic conditions, organic nitrogen is converted into ammonium under the action of ammoniating bacteria. The decrease from 70% to 30% saturation in figures 14 and 16 can show an inverse concentration between nitrate and ammonium. Through aeration and intermittent operation to increase the oxygen in the soil, thereby promoting nitrification, the $\text{NH}_4^+ - \text{N}$ is sequentially converted into NO_2^- and NO_3^- using nitrifying bacteria.

According to the results, the TN removal efficiency of biochar 10% and biochar 5% were the highest. In the summer for 70% and 30% saturation the removal efficiency of biochar 10 is 80.8% and 84.8%, while biochar 5% 69.3% and 80.7% respectively. Both biochar biofilters are affected by the saturation decrease where the performance has improved to 88.3% and 81.2% for biochar 10% and biochar 5% in the fall. And for 30% in the fall, they performed 90.6% for biochar 10% and 85.2% for biochar 5%. Biochar 10% showed promising performance in ammonium and nitrate concentration in the greywater effluent in 30% saturation in the summer and fall. And it's considered as the lowest ammonium and the only case where the biofilters succeed to filtrate the ammonium from the greywater. Therefore, there is compatible discoveries between this research and Dalahmeh et al., (2018) research. In their study, they detected the capacity of biochar of ammonium adsorption and nitrification process in low hydraulic load and aerobic environments, especially within the biochar filters. A discovery that proves the theories in (Yang et al., 2022) about the nitrification process as mentioned previously.

Regarding woodchip filters, the results are compatible with Saliling et al, (2007) where the woodchip showed promising performance in reducing the nitrate, nitrite, and nitrogen effluent concentration from the greywater except

the ammonium effluent concentration that increased during the summer and during the fall where the saturation decreased from 70 to 30%. In fall 30% saturation the concentration is decreased to an average of 0.04 mg/l. Unlike Gomez, (2021) that indicated a high performance of ammonium removal efficiency of woodchip filter.

The mycorrhiza showed a similar performance with Gomez, (2021) study of nitrite, nitrate, and TN removal efficiency. The mycorrhiza did not show a high removal efficiency of ammonium where the concentration has increased throughout the experiment apart from 30% saturation in the fall where the low saturation affected positively, and the concentration is decreased to 0.04 mg/l.

6.4 Metals Removal

In this study, boron, copper, zinc, and nickel were analyzed for the purpose to examine the infiltration removal efficiency of the metals with different types of biofilters. The results revealed that the biofilters efficiently removed the Zn, Cu, and Ni from the influent. The removal efficiency is > 90% of the different heavy metals with no significant difference between the biofilter's performances.

With a small difference, the biochar 10%, biochar 5%, and mycorrhiza stood out. These results enhance those of Zhou et al., (2017) which revealed that biochar can efficiently remove Zn, Cu, Cr, and Mn from wastewater. In addition, Chen et al, (2018) explained in their study the properties of biochar and the adsorptive capacity of biochar for heavy metals. The mycorrhiza results come in line with Poor et al, (2018) findings that the addition of mycorrhiza to the plants in the soil can increase the absorption of the zinc. According to the results, the boron concentration is increased in the system, which can be explained by leaching from the biofilters into the system.

6.5 Organic Compounds Removal

The results revealed that DEET is removed successfully, and the removal efficiency ranged between 51 and 92% for both saturation levels.

According to Table 29, 30% saturation has higher removal than 70% unless for woodchip and biochar 10%. Bimova et al, (2021) mentioned in their study that a higher amount of biochar does not particularly increase the pharmaceuticals removal efficiency, which is compatible with this study.

It is noticeable that biochar 5% has the higher removal efficiency of DEET. This finding comes in line with Sui et al, (2010) where the DEET removal efficiency is 69%. Moreover, Ren et al, (2021) indicate 66.8% removal efficiency of DEET at low hydraulic load (8 cm/d) using CW.

The biofilters showed high efficiency in the diclofenac removal from greywater. Moreover, for 30% saturation, the removal efficiency was higher than 70% saturation among all the biofilters. The highest removal efficiency is 98.1% of biochar 5% in 30% saturation. The results are compatible with Bimova et al, (2021) that reported one of the highest sorption efficiencies by biochar is of diclofenac due to the biochar properties and capacity of pharmaceuticals sorption.

6.6 Surfactant's removal efficiency

Removal of surfactants has been one of the crucial tasks in wastewater treatment of many industries such as cosmetic, homecare, pharmaceuticals, and paper industries (Ogawa & Kawase, 2021). According to the results of this study, the biofilters showed high efficacy, and the surfactants were removed successfully, where the removal efficiency ranged between 97 and 100%. This indicates that the system performed well regarding the removal of the surfactant from the greywater. Moreover, Figure 18 and 19 are not showing a significant difference between the infiltration results in 70% saturation and those in 30% saturation. Even though, for 30% the removal efficiency is slightly higher than in 70% saturation. The surfactants removal by degradation option was eliminated in this study since as Ramprasad & Philip, (2018) described in their study about the surfactants and PCPs removal by CW that anionic and nonionic surfactants are found sensible to photo-degradation, and in this experiment, the barrels system was covered by a black plastic bag to prevent the sunlight to penetrate.

6.7 Phosphorus removal Efficiency

Regarding the phosphorus removal efficiency, the results revealed that the different biofilters showed a challenging removal of phosphorus.

According to Figure 20, the phosphorus effluent concentration is affected by the hydraulic load. Consequently, in 70% saturation after the first sample that showed a lower effluent concentration than the influent, the concentration was much higher and the biofilters failed to reduce the concentration. While for 30% the phosphorus was successfully removed in the effluent of the biochar 10% and the woodchip filters in 30% saturation.

The reduction of the saturation from 70 to 30% (Figure 21) showed a lower concentration than 70% saturation (yet until the 8th sample the concentration was higher than the influent one). As 30% in the summer, during the fall biochar, 10%, and woodchip have removed the phosphorus successfully and kept the concentration below the influent concentration throughout the experiment. While for biochar 5%, default, and mycorrhiza the phosphorus concentration has increased in the effluent of 70% saturation in summer and the decrease of the saturation from 70% to 30% in the fall.

According to the literature, Pang et al, (2020) detected that hydraulic load has a negative effect on total phosphorus removal. Where total phosphorus removal efficiencies decreased with the increase of the hydraulic load. These findings are compatible with the results of this study. The high efficiency of the surfactant's removal can be explained by plants uptake, which is in agreement with Ramprasad and Philip, (2018) that indicates anionic surfactants are found mostly in the root system and the nonionic surfactants are found in the leaf. In addition, the removal of the surfactant can be explained by adsorption by the biofilters. according to the results, all the biofilters showed a high efficiency, which is compatible with the results of Ramprasad and Philip, (2016) that indicates the high adsorption of the anionic and nonionic surfactants by the soil.

7 Conclusions

This study was conducted in purpose to examine the efficiency of the greywater infiltration treatment under two different hydraulic loads; 70 and 30%, and different biofilters. The biofilters chosen for the study are, default, woodchip, mycorrhiza, biochar 5%, and biochar 10% which are mixed in different ratios with soil, sand, and compost, and the mixture were added into 75 L barrel and 50 duplicates.

The results revealed a dependency between the biofilter type and the removal efficiency of each parameter. In addition, the analysis and graphs of the results proved that there is an effect of the hydraulic load on the efficiency of the treatment and the biofilters' performance. For the TOC and TN removal efficiency, the biochar 10% owned the highest performance in both saturations. In addition, all the biofilters showed higher performance in 30% during the fall specially biochar, mycorrhiza, and woodchips. Therefore, I would recommend the application of low saturation and further study of the biochar for chemical load removal in greywater. Similarly, the highest removal of TP by the biofilters was in 30% in fall where biochar 10% and woodchip showed the highest performance.

The metals were absorbed successfully by the biofilters except for the boron that leached from the filters. Additionally, DEET and diclofenac removal efficiencies are higher in 30% than 70%. Also, the diclofenac is found in lower concentrations than the DEET in the greywater effluent. Yet, according to the literature, their existence in the treated greywater raises the risk of contamination in case of reuse the treated water.

This study indicates the efficiency of the greywater treatment under different conditions such as the biofilters types and the hydraulic load. Since the biofilters and the hydraulic load affect differently the infiltration efficiency, I would recommend further research on the topic under a field condition to simulate a real situation to optimize and maximize the benefits of using a sustainable material such as biochar and treat the greywater to minimize the

water demand. Furthermore, I would recommend using an advanced computational model for future forecast and data analysis.

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