

Czech University of Life Sciences
Faculty of Environmental Sciences
Department of Applied Ecology



Bachelor Thesis

**Experimental comparison of the suitability of filter materials for the removal of
selected micro-pollutants occurring in greywater**

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- The first phase of work on the thesis will be a literature search aimed at mapping studies that dealt with the use of different filtration materials, following by comparison of different design types of filtration devices used for the purification of greywater.
- The second phase of the thesis will be experimental part, where batch tests and then a physical model of infiltration trench will be used. Various filter materials will be tested to remove selected micropollutants from greywater.

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Honest proclamation:

I hereby declare that I have independently elaborated the bachelor thesis with the topic of: “Experimental comparison of the suitability of filter materials for the removal of selected micro-pollutants occurring in greywater” and that I have cited all of the information sources that I used in the thesis as listed at the end of the thesis in the list of used information sources. I am aware that my bachelor thesis is subject to Act No. 121/2000 Coll., on copyright, on rights related to copyright and on amendments of certain acts, as amended by later regulations, particularly the provisions of Section 35(3) of the act on the use of the thesis. I am aware that by submitting the bachelor thesis agree with its publication under Act No. 111/1998 Coll., on universities and on the change and amendments of certain acts, as amended, regardless of the result of its defense. I declare that I have used the AI tools in accordance with the internal regulations of the University and the principles of academic integrity and ethics. I refer to the use of these tools in the work in an appropriate way. With my own signature, I also declare that the electronic version is identical to the printed version and the data stated in the thesis has been processed in relation to the GDPR.

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Gratitude:

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Abstrakt

Cílem této studie bylo experimentálně porovnat vhodnost různých přísad do filtračních materiálů pro odstranění vybraných mikropolutantů běžně se vyskytujících v šedé vodě. Byly hodnoceny tři přísady filtračních materiálů včetně vápence, štěpky, biouhlu 5% s ohledem na výchozí strukturu. Účinnost odstraňování těchto materiálů byla hodnocena pro Celkový organický uhlík (TOC), celkový uhlík (TC), anorganický uhlík (IC), celkový dusík (TN), amonné ionty (NH_4^+), Bor (B), měď (Cu), nikl (Ni), zinek (Zn), benzotriazol (BTR) a ibuprofen (IBU) při různých rychlostech nasycení a kontaktních časech. Výsledky ukázaly, že biochar 5% byl nejúčinnějším filtračním materiálem při odstraňování TOC, TN a Ni, IBU a BTR ve všech experimentálních nastaveních. Vápenec vykazoval proměnlivou účinnost, ale fungoval lépe při 30% nasycení a 22 hodinách kontaktního času pro TOC, TN, Cu, Ni, BTR a vykazoval vynikající výkon a byl nejvíce dominantní při odstraňování Zn. Štěpka vykazovala nižší účinnost odstraňování ve srovnání s vápencem a biouhlem 5%, ale byla jediným filtračním materiálem, který do určité míry odstranil TC. Pokud jde o zbývající TC, IC, NH_4^+ a B, vybrané filtrační materiály se nedoporučují používat pro účely odstraňování těchto vybraných mikropolutantů. Zjištění této studie poskytují cenné poznatky o výběru vhodných filtračních materiálů pro systémy úpravy šedé vody, s důrazem na důležitost optimalizace kontaktních časů a rychlosti nasycení pro účinné odstraňování mikropolutantů. Je zapotřebí dalšího výzkumu, aby bylo možné lépe porozumět složitým interakcím mezi filtračními materiály, kontaktními časy, mírou nasycení a odstraněním kontaminantů, aby se vyvinuly účinnější a udržitelnější systémy úpravy šedé vody.

Klíčová slova:

Šedá voda, filtrace, štěpka, vápenec, biouhel 5%

Abstract

This study aimed to experimentally compare the suitability of different additives to filter materials for the removal of selected micro-pollutants commonly found in greywater. Three filter materials additives were evaluated including limestone, woodchips, biochar 5% with reference to default structure. The removal efficiency of these materials was assessed for total organic carbon (TOC), total carbon (TC), inorganic carbon (IC), total nitrogen (TN), ammonium ions (NH_4^+), boron (B), copper (Cu), nickel (Ni), zinc (Zn), benzotriazole (BTR), and ibuprofen (IBU) at varying saturation rates and contact times. The results demonstrated that biochar 5% was the most effective filter material in removing TOC, TN, and Ni, IBU, and BTR across all experimental setups. Limestone displayed variable efficiency but performed better at a 30% saturation and 22 hours of contact time for TOC, TN, Cu, Ni, BTR and showed excellent performance and was most dominant in Zn removal. Woodchips exhibited lower removal efficiencies compared to limestone and biochar 5% but was the only filter material to remove TC to some extent. Regarding the remaining TC, IC, NH_4^+ and B, selected filter materials are not recommended to be used for removal purposes of these selected micropollutants. The findings of this study provide valuable insights into the selection of suitable filter materials for greywater treatment systems, emphasizing the importance of optimizing contact times and saturation rates for effective removal of micro-pollutants. Further research is needed to better understand the complex interactions between filter materials, contact times, saturation rates, and contaminant removal to develop more efficient and sustainable greywater treatment systems.

Keywords:

Greywater, filtration, woodchips, limestone, biochar 5%

Table of Content

List of abbreviations	11
1. Introduction.....	1
2. Objectives of the thesis	2
3. Literature review	3
3.1. Production of greywater.....	3
3.2. Greywaters physiochemical parameters.....	3
3.2.1. Physical composition	4
3.2.2. Chemical composition.....	4
3.2.3. Biological composition	8
3.2.4. Criteria TC, TOC, IC, TN, NH_4^+ in GW	9
3.2.5. Metals occurring in GW (B, Cu, Zn, Ni)	14
3.2.6. Pharmaceuticals occurring in GW (IBU, BTR)	16
3.3. Greywater treatment.....	18
3.3.1. Treatment of GW on TOC, TN, NH_4^+	19
3.3.2. Treatment of GW from heavy metals Cu, Ni, Zn	21
3.3.2. Treatment of GW from pharmaceuticals (IBU, BTR)	21
3.4. Filtration materials	23
3.4.1. Woodchips	23
3.4.2. Biochar	24
3.4.3. Limestone.....	27
4. Methods	28
4.1. The experiment site and equipment	28
4.1.1. Physical models design	29
4.1.2. Synthetic greywater (SGW)	31
4.1.3. Field work on experimental site.....	32
4.2. Laboratory analyses	32
4.2.1. pH, EC, Turbidity analysis:.....	32
4.2.2. NH_4^+ analysis:	34
4.2.3. TC, TOC, IC, TN analysis:	36
4.2.4. Heavy metals analysis:.....	36
4.2.5. Pharmaceuticals analysis.....	37
4.3. Statistical evaluation of collected data.....	37
5. Results.....	38

5. 1. Efficacy in removal of total carbon (TC), total organic carbon (TOC), inorganic carbon (IC).....	39
5. 2. Efficacy in removal of total nitrogen (TN) and ammonia ion (NH ₄ ⁺)	41
5. 3. Efficacy in removal of Boron (B), Copper (Cu), Nickel (Ni), and Zinc (Zn).....	44
5. 3. Efficacy in removal of Benzotriazole (BTR) and Ibuprofen (IBU)	50
6. Discussion.....	52
6.1. Total carbon (TC), total organic carbon (TOC), inorganic carbon (IC)	52
6.2. Total nitrogen (TN) and ammonia ion (NH ₄ ⁺)	53
6.3. Metals removal (B, Cu, Ni, Zn)	54
6.4. Pharmaceuticals removal (IBU, BTR)	55
7. Conclusion	56
8. Bibliography	57
8.1. Scientific Publications	57
8.2. Internet Sources	66

List of abbreviations

B	Boron
BOD ₅	Biochemical oxygen demand
BTR	Benzotriazole
COD	Chemical oxygen demand
Cu	Copper
EC	Electrical conductivity
FM _s	Filter materials
GW	Greywater
HM _s	Hard metals
IBC tank	Intermediate bulk container tank
IBU	Ibuprofen
IC	Inorganic carbon
MBR	Membrane bioreactor
ME-(O/A)CW	micro-electrolysis - oxic/anoxic vertical constructed wetland
NH ₄ ⁺	Ammonia iont
Ni	Nickel
NTU	Nephelometric turbidity unit
SGW	Synthetic greywater
TC	Total carbon
TN	Total nitrogen
TOC	Total organic carbon
TP	Total phosphorus
TSS	Total suspended solids
TTri	Tolyltriazole
U-N	Urine Nitrogen
WWTP	Wastewater treatment plant
Zn	Zinc

1. Introduction

Greywater (GW) is a sewage water without fecal pollution which is relatively suitable for reuse due to its explicit origin from bathtubs, shower drain, bath sink and laundry outflow in comparison with blackwater (BW) which comes from toilet and other draining plumbs for more solid or greasy sewage. Nevertheless, GW is still considered as a polluted water which cannot be reused straight away nor discharged into the environment prior treatment.

It is a known fact, that developed regions of the globe run on essential for their community's wastewater treatment plants (Malik et al., 2015). In the case of developed regions both GW along with BW flushed in the same drain due to the way they were designed decades ago. Wastewater treatment plants represent a huge area with massive multiphase treatment systems where community sewage end its path before being treated and safely discharged into nearest body of water (US Environmental Protection Agency, 1988). However, most developing countries don't have a privilege of proper water sanitation which makes utilization of treatment plants mentioned above economically unavailable (Bouabid et Louis, 2021). Unfortunately, most regions of these countries have decentralized sewage systems. Basically, every household makes their own way of discharging wastewater. In the best-case scenario sewage from a households was relocated to septic tanks (Mandal et al., 2011). In the worst-case scenario decentralized sewage from a household may just flow out to soil if toilets represent improvised pit latrines with a cabin placed outdoors at the back yard where potential BW content ends up in fecal sludge (Garn et al., 2017). Meanwhile potential GW originated from kitchen, bathtubs etc. as mentioned above discharges straight to soil. Heavy reality in some cases like the last one already created GW separation leaving a window for implementation of cost-effective treatment for GW (Semiyaga et al., 2017). The pace of centralization of sewage systems is remarkably slow and introduction of proper wastewater treatment solutions is uncertain. Further pollution of topsoil and ground water by GW before the centralization may lead to devastating ecological imprint (Reichman et Wightwick, 2013). Meanwhile, the idea of filtering GW is worth considering as a solution for these regions. There is a way of using cost

effective filtering with the help of biodegradable materials for treating GW before it contacts ground water.

2. Objectives of the thesis

This experiment consists of a two-phase study of GW. The first phase was reviewing previous studies about GW and its treatment. The second phase included an experimental part which compared suitability of filter materials additives such as woodchips, limestone, and biochar 5% for the removal of selected micro-pollutants occurring in greywater.

To assess the efficiency of filter materials additives to remove selected micropollutants, those additives were tested under different saturation conditions (30%, 70%) and two different contact times (22 hours, 72 hours). Under these conditions objectives subdivide into:

- Assessment of individual additives removal efficiency
- Assessment of carbon (TOC, TC, IC), total nitrogen (TN), and ammonia ion (NH_4^+) removal
- Assessment of semi metal boron (B) and heavy metals zinc (Zn), nickel (Ni), and copper (Cu) removal.
- Pharmaceuticals benzotriazole (BTR) and ibuprofen (IBU) removal.

3. Literature review

3.1. Production of greywater

Production of GW varies from household to household. The quantity varies from 15 l to hundreds of liters a day from a single user (Oteng-Peprah et al., 2018). The generated amount of greywater shifts its true volumes from region to region according to inhabitants needs and taxation on water supply. This astonishing quantity comes mainly from bathtubs, sinks and washing machines after activities such as showering, washing dishes and laundry. Outcome of our daily activities involving use of freshwater is dumping great amounts of alternative water resource with a great potential for secondary use (Fountoulakis et al., 2016).

3.2. Greywaters physiochemical parameters

The composition of greywater reflects household array of chemicals used for daily activities on washing, cleaning, and personal hygiene. That is why it is fair to say that there is no certain formula in existence which would describe the composition of greywater as accurate as possible. Starting off with the chemicals, we may take a closer look into detergents, hygiene, and cleaning chemicals. These types of chemicals are referred to be alkaline on pH scale ranging from 7 to 14. The ability to dissolve dirt, oil and other polluting agents is caused by chemicals negative charge on hydrophilic ends (ACI, 2016). The ability to dissolve dirt plays a huge role in maintaining clean state inside a household but when untreated water with such chemicals discharges to sewage it will remain active in dissolving. Meaning, dissolving properties may cause soil degradation in case of untreated irrigation by greywater. Chemical compounds are not the single criteria of pollution in greywater as far as it is known. The composition of greywater is also rich for disease causing agents such as *E. Coli* and *coliforms* which are products of the human body originating in digestion system. These bacteria travel around a bathroom in many ways as we occasionally defecate, wash parts of our body, and often delay regular cleaning of bathroom. Wet and warm environment in bathtubs and sink create ideal conditions for bacteria reproduction (Rusin et al., 1998). These bacteria get flushed away adding up to GW.

3.2.1. Physical composition

Temperature (t), total suspended solids (TSS), electrical conductivity (EC) and turbidity (NTU) are used to describe physical state of GW (Shaikh et Ahammed, 2020). Greywater has a temperature of 18 – 35 °C due to the use of mainly hot water for cleaning purposes. High temperatures allow microbial life to flourish creating comfortable environment for disease causing agents. Total suspended solids or TSS is the parameter to describe foggy appearance of water. Range of TSS in greywater shift from 190–537 mg/L as has been reported (Oteng-Peprah et al., 2018; Delhiraja et Philip, 2020). These values come from washing activities where dirt is dissolved to smaller particles. Electrical conductivity (EC) of greywater ranges from 14 and 3000 $\mu\text{S}/\text{cm}$ (Shaikh et Ahammed, 2020). The shown value indicates high levels of EC due to dissolved inorganic compounds and leaching particles of poor-old plumbing (Fountoulakis et al., 2016). Turbidity ranges between 19 and 444 NTU, origin of the cause is related to the TSS of water and describes visible and non-visible change in color of the water (Fountoulakis et al., 2016).

3.2.2. Chemical composition

Greywater may contain a variety of chemicals which are introduced to wastewater by household activities. The specific chemicals occurring in greywater vary depending on the source and the products used in these activities (Henze et Curtis, 2007). Here are some examples of common chemicals that may be present in GW.

Detergents and soaps: GW can contain detergents and soaps used for washing clothes, dishes, and personal hygiene (Delhiraja et Philip, 2020). These products contain surfactants, which can persist in the soil for long period of time and can travel through to reach ground water (“Micropollutants and Challenges,” 2020). Detergents and soaps contribute significantly to accumulation of TOC and IC in GW. TOC is a critical parameter for evaluation of organic matter in GW that can potentially contribute to microbial growth or chemical reactions in stored water (Tsoumachidou et al. 2017). IC in GW mainly comprises carbonates, bicarbonates, and dissolved

CO₂. It's crucial to monitor IC because it affects the pH and alkalinity of water, influencing corrosion potential and the effectiveness of disinfection processes (Tsoumachidou et al. 2017). Some products also contain phosphates and other chemicals that can harm the environment. Both surfactants and phosphates cause mobilization of heavy metals present in the soil, which can lead to the transport of these metals into the groundwater (Johnson et al., 2021).

Cleaning products: GW can contain products used for household cleaning, which can contain a variety of chemicals such as disinfectants, bleaches, and ammonia (Dwumfour-Asare et al., 2020). This source is also a main contributor to TC and metals such as Cu, Zn, Ni, and B in GW. The complex mixtures found in these products can introduce a variety of metals and carbon compounds, necessitating comprehensive treatment strategies. Vargeese et al. (2015) emphasize the importance of understanding the total carbon content for selecting appropriate greywater treatment solutions, ensuring the removal of both organic and inorganic constituents derived from cleaning products (Vargeese et al. 2015). Disinfectants can contribute to the development of antibiotic-resistant bacteria (Bragg et al., 2014). This is because some disinfectants can stimulate the growth of bacteria, creating a selective pressure that favors the development of antibiotic-resistant strains (Itzhari et Ronen, 2023). Bleaches can have a negative impact on soil quality and can reduce soil fertility and productivity (Eriksson et al., 2002). These chemicals can be harmful to human health and the environment if not properly treated. Ammonia (NH₃) may alter pH levels of soil when discharged. High soil pH levels can affect products due to excessive nitrogen accumulation in soil (Wardani et al., 2021). This impact may lead to plant diversity decreases causing changes in higher trophic levels.

Personal care products: GW can contain personal care products such as shampoos, conditioners, lotions, and deodorants. These products can contain fragrances, preservatives, and colorants that can cause harm to the environment. They are also found to be origins of Total Nitrogen (TN), ammonia ion (NH₄⁺), and certain metals in greywater (Tsoumachidou et al., 2017; Alrousan et al., 2020). Fragrances may contain various chemicals, some of which may cause indirect negative impact to the environment (Zhang et al., 2013). Phthalates occurring in fragrances are known endocrine disruptors and can damage soil microbiota (Gao et Wen, 2016). Preservatives such as parabens (Pritchett et al., 2015), formaldehyde releasers

(Brandão et al., 2018), quaternary ammonium compounds (Di Nica et al., 2017), and organic acids are widely used in personal care products to prevent the growth of harmful bacteria, fungi, and other microorganisms. The use of preservatives in personal care products is a subject of ongoing debate, as some preservatives have been associated with potential health risks (Nowak et al., 2021). These risks are related to primary use by inhabitants of households before their contact. In the matter of GW impact containing preservatives to the environment, both soil and groundwater pollution is possible (Brausch et al., 2011). For instance, formaldehyde is referred to as carcinogen posing risk to human health (Heshammuddin et al., 2023). Groundwater contamination by parabens preservative in the case study of Serra-Roig et al., 2016 did not show feasible risks for local species nor to humans. On the other hand, surface water contamination by parabens from households has proven rising endocrine disruption in aquatic life (Serra-Roig et al., 2016). Colorants show greater risks as they present immunogenic, carcinogenic, teratogenic, and mutagenic hazard to the environment when discharged to the bodies of water without treatment (Azari et al., 2019).

Pharmaceuticals: GW can contain common pharmaceuticals such as Ibuprofen (IBU) and Benzotriazole used for wide range of pain and anti-inflammatory treatment in both cases and further antiviral, antibacterial, and antihypertensive properties in Benzotriazole (BTR) shown in studies (Suma et al., 2011). Pharmaceuticals can occur in GW when people flush unused medications down the sink. In addition, medications mentioned above may enter GW through rinsing of medical equipment (“Nurofen For Children Orange With Dosing Syringe 150ml,” 2023). Pharmaceuticals in GW may have several harmful effects on the environment. BTR is not only a chemical compound used in pharmaceutical purposes but also used as common compound in dish-washing detergents (Careghini et al., 2014). Ibuprofen and Benzotriazole may have long-term ecotoxicity hazard on aquatic organisms such as fish, algae, and invertebrates due to their slow biodegradability (Liu et al., 2012). These compounds can have endocrine-disrupting effects on aquatic organisms affecting their growth and reproduction (Flippin et al., 2007).

It is important to note that not all the chemicals found in GW are harmful. For example, some of the chemicals can provide nutrients for plant growth (Rodda et al., 2011). However, it is important to identify the specific chemicals present in GW to

determine the appropriate filtering material and filtration method for effective treatment.

Table 1. The physicochemical characteristics of grey water with biological composition by different categories (Li et al., 2009).

	Bathroom	Laundry	Kitchen	Mixed
pH (-)	6.4–8.1	7.1–10	5.9–7.4	6.3–8.1,
TSS (mg/l)	7–505	68 – 465	134–1300	25–183
Turbidity (NTU)	44–375	50 – 444	298.0	29–375
COD (mg/l)	100–633	231 – 2950	26–2050	100–700
BOD ₅ (mg/l)	50–300	48 – 472	536–1460	47–466
TN (mg/l)	3.6–19.4	1.1 – 40.3	11.4–74	1.7–34.3
TP (mg/l)	0.11– > 48.8	ND – > 171	2.9– > 74	0.11–22.8
Total coliforms (CFU/100 ml)	10– 2.4×10^7	200.5– 7×10^5	$> 2.4 \times 10^8$	56– 8.03×10^7
Fecal coliforms (CFU/ 100 ml)	$0–3.4 \times 10^5$	50– 1.4×10^3	–	0.1– 1.5×10^8

Table 1 presents the findings of a study on greywater characteristics based on sources of wastewater (Li et al., 2009). These characteristics play a role in determining water quality. PH serves as an indicator to monitor water quality as it influences the solubility of nutrients and minerals in water as well as the efficiency of water treatment

processes. Total Suspended Solids (TSS) reveal the presence of pollutants that can harm ecosystems and human health. Turbidity, measured in turbidity units (NTU) indicates the clarity or cloudiness of water with higher levels suggesting suspended solids. Chemical Oxygen Demand (COD) measures the amount of oxygen needed to oxidize inorganic compounds in a water sample providing insight into the organic content present. Biochemical Oxygen Demand (BOD5) gauges the oxygen required by microorganisms to decompose matter in water samples highlighting potential negative impacts on aquatic life and human well being. Total Nitrogen (TN) encompasses both inorganic nitrogen forms serving as a comprehensive measure, for nitrogen content analysis (Li et al., 2009)..

Excessive nitrogen levels, similar to phosphorus (TP) can lead to eutrophication, in water bodies resulting in oxygen depletion and environmental changes. Fecal coliforms, a subgroup of coliform bacteria linked to fecal matter suggest the likelihood of harmful pathogens that may trigger illnesses (Li et al., 2009) (ChatGPT4 grammatical corrections).

3.2.3. Biological composition

Studies on the components of greywater often mention bacteria like *Escherichia coli*, known for their pathogenic nature and anaerobic fecal coliform bacteria. These bacteria are highlighted for their presence in greywater ranging from 11–13% (Eriksson et al. 2002). Coliforms, *E. Coli* are commonly associated with warm blooded animals. The temperature conditions in greywater around 35 ± 2 °C allow coliform bacteria to survive for up to 48 hours while *E. Coli* can persist at temperatures of 15–18°C for 4–12 weeks. If untreated these bacteria have the potential to cause illnesses such as diarrhea and fever. Additionally the presence of coliforms serves as an indicator that the aquatic environment may support other microbial life forms like viruses and protozoa (Halkman et Halkman., 2014). While fecal matter is typically absent in greywater since it originates from the intestine and is usually separated from sources like sinks and washing machines; there are instances where fecal matter could contaminate greywater systems. One such scenario is through cross connections,

between greywater and black water (sewage) systems resulting from plumbing installations or maintenance practices.

Sewage backflow can lead to the presence of matter in groundwater. When there is a blockage in the sewage system sewage can overflow into the groundwater system. Having an understanding of all aspects of greywater is essential, for this analysis. Improved comprehension of the chemical and biological makeup of greywater enables us to concentrate on effectively removing water pollutants and contaminants.

3.2.4. Criteria TC, TOC, IC, TN, NH_4^+ in GW

3.2.4.2. Total Carbon (TC)

Greywater's Total Carbon (TC) encompasses all carbon forms, including organic (TOC) and inorganic carbon (IC) such as carbonates, bicarbonates and free carbon dioxide. Understanding TC is crucial for gauging the carbon content in greywater and its impact on treatment processes and the environment. Managing TC effectively is essential for treating greywater to a level for reuse or environmental discharge. Evaluating TC levels aids in assessing the efficiency of greywater treatment systems. Elevated TC levels, from organic sources can spur microbial activity and chemical oxygen demand (COD) leading to lowered oxygen levels in water bodies and compromising water quality. On the other hand the inorganic segment of TC primarily comprising carbonates and bicarbonates helps maintain greywater pH balance vital for biological treatment processes and aquatic ecosystem health post treated greywater discharge. Various household activities contribute to TC in greywater with organic sources like food remnants, detergents, human waste and other organic material, from kitchen, laundry, and bathroom wastewater playing a role.

The presence of carbon in water is mainly influenced by the dissolution of carbon dioxide resulting in the formation of carbonates and bicarbonates. This process is impacted by the use of cleaning products and the hardness of the water supply (Ghaitidak & Yadav 2013). The variation in carbon (TC) concentrations

found in greywater reflects its diverse sources emphasizing the importance of understanding greywater to customize treatment processes effectively. Managing TC in greywater poses a challenge due to the need for treatment systems to adjust to changes in carbon concentrations and compositions. To optimize TC removal treatment methods must strike a balance between chemical processes to efficiently break down organic components while regulating inorganic carbon levels to control system pH and prevent scaling issues. Additionally extracting carbon from organic sources offers an opportunity, for recovering resources through biogas production supporting circular economy principles and sustainability objectives (Kadewa et al., 2010) (ChatGPT4 Consensus tools for source search).

3.2.4.1 Total Organic Carbon (TOC)

Total Organic Carbon (TOC) is a fundamental measure of the organic compounds present in water, representing a critical parameter in assessing greywater quality and its potential impact on both human health and the environment. TOC in greywater originates from a variety of sources, including food residues, detergents, and personal care products, and can significantly affect the efficiency of treatment processes and the suitability of treated water for reuse (Eriksson et al., 2002). The variability and complexity of TOC composition pose challenges in greywater treatment, necessitating a detailed understanding of its characteristics and behavior. TOC is considered a pivotal factor in greywater management due to its dual role in promoting microbial growth and in the formation of disinfection by-products (DBPs) during treatment processes. High TOC levels can lead to increased biological oxygen demand (BOD) in receiving waters, contributing to oxygen depletion and adverse ecological effects (Jefferson et al., 2004). Moreover, TOC serves as a precursor for DBPs when chlorination is used as a disinfection strategy, raising concerns over water safety for non-potable reuse applications (Ledin et al., 2001). The sources of TOC in greywater are diverse, encompassing a wide range of organic materials from household activities. Kitchen greywater, for example, is characterized by high TOC levels due to the presence of food waste and cooking residues. In contrast, bathroom and laundry greywater may contain lower TOC concentrations but with complex compositions due to the use of synthetic detergents, personal care products, and

pharmaceuticals (Ghaitidak & Yadav, 2013). This variability underscores the importance of source separation and targeted treatment approaches to effectively reduce TOC levels in greywater (ChatGPT4 Consensus tools for source search).

3.2.4.3 Inorganic Carbon (IC)

Inorganic Carbon (IC) in greywater, primarily composed of carbonates, bicarbonates, and dissolved carbon dioxide (CO₂), plays a crucial role in determining the chemical properties of greywater, including its pH and alkalinity. IC's significance in greywater treatment processes and its impact on the environment cannot be overstated, as it directly influences the efficacy of various treatment methodologies and the potential for reuse of treated water. The presence of IC in greywater is pivotal for maintaining a balanced pH, essential for the biological treatment processes and the health of aquatic ecosystems. High levels of IC can lead to alkalinity, which may inhibit certain microbial activities crucial for organic matter degradation in biological treatment systems. Conversely, low IC concentrations can result in acidic greywater, leading to corrosion in piping systems and adverse effects on microbial flora essential for effective greywater treatment (Ghaitidak & Yadav, 2013). Sources of IC in greywater include the dissolution of atmospheric CO₂ and the use of cleaning agents and detergents that contain carbonates and bicarbonates. The variability in IC levels in greywater can be attributed to the types of products used in households and the initial water supply's chemical composition. This variability underscores the need for adaptable and robust treatment processes capable of handling the fluctuating levels of IC to ensure the treated water meets the required standards for reuse or discharge (Eriksson et al., 2002). The management of IC in greywater presents both challenges and opportunities. One of the main challenges is the need for continuous monitoring and adjustment of treatment processes to accommodate the variability in IC levels and maintain an optimal pH for treatment and reuse. However, this challenge also presents an opportunity to develop innovative treatment solutions that are flexible and efficient in managing IC in greywater. Furthermore, understanding the dynamics of IC in greywater can lead to more sustainable water reuse practices, where treated greywater can be safely used

for irrigation, landscaping, or industrial processes, contributing to water conservation efforts (Morel et Diener, 2006) (ChatGPT4 Consensus tools for source search).

3.2.4.4 Total Nitrogen (TN)

Nitrogen levels in greywater consist of nitrogen forms like organic compounds, ammonia, nitrites and nitrates. It's important to measure Total Nitrogen (TN) in greywater to understand its content and impact on the environment when using or disposing of it. Managing TN effectively is crucial for preventing effects on water ecosystems and promoting sustainable water reuse practices. Much TN in greywater can cause eutrophication in water bodies leading to excessive growth of algae and plants that can harm aquatic life by reducing oxygen levels. Additionally certain nitrogen forms like nitrate can pose health risks if greywater is used for irrigation and contaminates food crops or groundwater. Sources of TN, in greywater include waste, food scraps and nitrogen based cleaning products. The concentration of TN varies depending on household habits, product usage and how greywater is separated from blackwater streams.

The diverse nature of greywater poses difficulties in its treatment calling for solutions to effectively lower TN levels to meet reuse or discharge standards (Jefferson et al., 2004). Balancing nitrogen removal efficiency with the complexity and costs of treatment systems is a challenge in managing TN in greywater. The varying composition of TN in greywater necessitates resilient treatment methods. Nevertheless these challenges also create opportunities for advancements in greywater treatment technology. By optimizing treatment processes for nitrogen removal we not safeguard environmental and public health but also enhance the potential for nutrient recovery from greywater facilitating its reuse in beneficial applications, like irrigation (Li et al., 2009) (ChatGPT4 Consensus tools for source search).

3.2.4.1 Ammonium (NH_4^+)

Ammonium (NH_4^+) plays a role in the overall nitrogen content found in greywater originating from different household sources like human waste, leftover food and the use of cleaning products containing nitrogen. The presence of NH_4^+ environmental well being. Having an understanding of how NH_4^+ behaves in greywater is crucial for creating effective strategies to manage and treat it reducing potential risks associated with its disposal or reuse. NH_4^+ directly contributes to the content in greywater, which can have negative environmental effects if not handled properly. In water environments high levels of NH_4^+ can lead to eutrophication by encouraging algae growth that depletes oxygen levels and harms aquatic organisms. Additionally NH_4^+ can transform into nitrate (NO_3^-) through nitrification processes in water bodies posing health hazards if it contaminates drinking water sources. Therefore treating greywater to eliminate or lessen NH_4^+ levels is vital for preventing environmental harm and safeguarding public health (Eriksson et al., 2002). The concentration of NH_4^+ , in greywater varies depending on household activities and the types of products used. Common sources include urine as a contributor and nitrogen containing ingredients found in detergents and cleaning agents.

The fluctuations in NH_4^+ levels create issues for treating greywater requiring technologies that can effectively eliminate NH_4^+ over various concentrations (Jefferson et al., 2004). Handling NH_4^+ in greywater brings about difficulties, including the necessity for monitoring and regulation of treatment procedures to ensure consistent removal efficiency. The variations in NH_4^+ concentrations and the risk of byproducts like nitrite during nitrification call for meticulous system planning and operation. Nevertheless these challenges also open up possibilities for advancements in greywater treatment offering the chance for recovering resources, such, as using nitrogen for producing fertilizers showcasing the benefits of a circular economy through efficient NH_4^+ management (Li et al., 2009) (ChatGPT4 Consensus tools for source search).

3.2.5. Metals occurring in GW (B, Cu, Zn, Ni)

Greywater has become a contributor of micropollutants, like Copper (Cu) Nickel (Ni) Zinc (Zn) and Boron (B). It is essential to comprehend their presence origins, destiny and possible environmental impacts to manage greywater resources efficiently.

3.2.5.1. Boron (B) occurring in GW

Boron is a trace element that, in small quantities, is essential for plant growth but can become toxic to plants and harmful to human health at higher concentrations (Gross et al., 2007). In domestic greywater, which includes wastewater from showers, sinks, and laundry, boron can originate from detergents, cleaning agents, and personal care products. The presence of boron in greywater poses challenges for reuse, especially for irrigation purposes, as excessive boron can lead to plant toxicity and soil degradation (Ghaly et al., 2021). Although high natural concentrations can occur in some areas, the risk to aquatic ecosystems from boron is generally low due to its low bioavailability and the adaptation of organisms to local conditions (Howe, 2007). The treatment of Boron (B) specifically was not widely considered in previous studies. However, findings of certain studies include this trace element among laundry effluents. The study notes that B is widely present in many detergents despite the regulations aiming to reduce B and substitute it with other compounds. The concentration of B in GW tested in this study averaged 1.3mg/L^{-1} . This concentration exceeded norms at some plants such as citrus trees (0.5mg/L^{-1}). Findings of this study also highlighted soil degradation by creating hydrophobic flow pattern of the soil reducing productivity (Wiel-Shafron et al., 2006) (ChatGPT4 grammar corrections).

3.2.5.2 Copper (Cu) occurring in GW

The presence of copper (Cu) as a micropollutant in greywater is a concern due to its potential environmental and health impacts. Copper originates from

plumbing materials, personal care products, and household cleaners. The treatment and removal of Cu and other micropollutants from greywater are crucial for safe reuse applications, such as irrigation and toilet flushing. Eriksson and Donner (2009) discuss the sources, presence, and potential fate of metals, including Cu, in onsite greywater treatment systems, highlighting that annual metal loads from bathroom greywater are relatively small but may still not always meet environmental quality standards for surface waters. The removal of Cu to acceptable levels is essential for minimizing risks to human health and the environment, particularly when greywater is used for irrigation, which could lead to the accumulation of Cu in soil and plants within 4 years (Turner et al., 2016). The management of Cu as a micropollutant in greywater requires comprehensive treatment strategies to mitigate its potential impacts. Advances in treatment technologies, such as activated carbon adsorption and ozonation, offer promising pathways for the effective removal of Cu from greywater, supporting sustainable reuse practices while protecting environmental and public health (Patel, Muteen, et Mondal, 2019) (ChatGPT4 grammar corrections)..

3.2.5.3 Nickel (Ni) occurring in GW

Nickel can enter greywater through the corrosion of nickel-containing fixtures and fittings, as well as from consumer products that contain Ni. While Eriksson and Donner (2009) discuss the sources, presence, and removal efficiencies of metals, including Ni, in greywater, the presence of Ni, even in small amounts, is significant due to its potential to cause allergic reactions and other health impacts in sensitive individuals. The presence of Ni in greywater emphasizes the importance of effective treatment strategies to ensure the safe reuse of greywater, particularly for applications that may involve human exposure or environmental release. The development and implementation of greywater treatment systems capable of efficiently removing Ni and other micropollutants are critical for mitigating health risks and environmental impacts associated with greywater reuse. While the research on Ni as a micropollutant in greywater is limited, the available studies underscore the need for comprehensive treatment solutions that address a wide range of contaminants (Turner et al., 2016) (ChatGPT4 grammar corrections).

3.2.5.4 Zinc (Zn) occurring in GW

The discussion and management of zinc (Zn) as a pollutant in greywater have drawn attention due to the widespread use of zinc in everyday products and its potential environmental effects. Zinc can find its way into greywater through means, such as the usage of personal care items containing zinc, detergents and the corrosion of zinc coated plumbing materials. Although zinc is a trace element for humans, plants and microorganisms excessive levels in the environment can harm aquatic life and ecosystems. Therefore regulating zinc levels in greywater is crucial to minimize impacts and support safe reuse practices. Ensuring removal of zinc and other pollutants from greywater is vital for its safe reuse especially in agricultural irrigation where the accumulation of heavy metals in soil can endanger crop health and soil quality. The advancement and optimization of greywater treatment systems that can efficiently eliminate zinc are key to expanding opportunities for greywater reuse and reducing the demand for freshwater resources. While research specifically targeting the removal of zinc from greywater treatment is limited exploring technologies like photocatalysis using ZnO nanoparticles shows promise, for innovative solutions to tackle this issue.

3.2.6. Pharmaceuticals occurring in GW (IBU, BTR)

The growing discovery of pharmaceuticals and industrial micropollutants such as Ibuprofen (IBU) and Benzotriazole (BTR) in greywater has sparked environmental worries. IBU, an used anti inflammatory medication has been detected in water bodies worldwide with research suggesting its ability to cause oxidative stress in unintended aquatic organisms. BTR, extensively utilized as a corrosion inhibitor along with its variations has been recognized as a pollutant particularly originating from sources like

dishwashers (Giger et al. 2006). Studies conducted by Pillard et al. (2001) on the toxicity of BTR to aquatic species showed varying levels of susceptibility emphasizing the environmental and ecological threats posed by these micropollutants. These results emphasize the requirement, for efficient wastewater treatment methods to reduce their release and safeguard aquatic life and water quality.

3.2.6.1. Ibuprofen (IBU) occurring in Greywater

The presence of Ibuprofen (IBU), a prevalent non-steroidal anti-inflammatory drug, in greywater and its potential environmental repercussions has become a focal point of research within the field of ecotoxicology. As active pharmaceutical ingredients (APIs) are increasingly recognized as emergent environmental contaminants, the distribution and impact of IBU in various water bodies, including influents and effluents of wastewater treatment plants (WWTPs), as well as surface, river, and public tap water across multiple countries, are of significant concern. Despite its widespread detection, the chronic effects and risks associated with IBU exposure to non-target organisms remain largely uncharted (Gonzalez-Rey et al., 2011). A notable study has embarked on evaluating the oxidative stress response in the sentinel species, mussel *Mytilus galloprovincialis*, by analyzing the activities of several antioxidant enzymes such as superoxide dismutase (SOD), catalase (CAT), glutathione S-transferase (GST), and glutathione reductase (GR), alongside levels of lipid peroxidation (LPO), upon exposure to environmentally realistic concentrations of IBU for two weeks. The findings reveal a substantial induction of SOD activity and LPO in exposed mussel gills, alongside a reduction in the antioxidant defenses attributed to CAT, GR, and GST when compared to controls. This differential biomarker integration effectively distinguishes between non-exposed and exposed groups, underscoring the disruption of the redox defense system and IBU's pro-oxidant capacity. The study advocates for further investigation into IBU's potential endocrine-disrupting effects on the reproductive fitness of mussels, given its role in inhibiting prostaglandin biosynthesis (Gonzalez-Rey et al., 2011) (ChatGPT4 grammar corrections).

3.2.6.2 Benzotriazole (BTR) occurring in Greywater

Benzotriazole (BTR) and its derivatives are recognized as significant micropollutants in greywater, attributed to their widespread use as corrosion inhibitors in various industrial applications, including coolants, deicers, surface coatings, cutting fluids, and hydraulic fluids. Pillard et al. (2001) conducted a study to assess the toxicity of benzotriazole and its derivatives, notably in the context of aircraft deicing fluids (ADFs), which have been identified as a major source of environmental toxicity. Their research utilized acute toxicity assays to evaluate the effects of benzotriazole (BT), two methylbenzotriazole (MeBT) isomers, and butylbenzotriazole (BBT) on three aquatic species: Microtox bacteria (*Vibrio fischeri*), fathead minnow (*Pimephales promelas*), and water flea (*Ceriodaphnia dubia*). The findings revealed that the toxicity response varied significantly across the tested organisms and compounds, spanning over two orders of magnitude. *Vibrio fischeri* demonstrated a higher sensitivity compared to *C. dubia* and *P. promelas* to all test materials, highlighting the varying degrees of susceptibility among aquatic organisms to BTR derivatives. Interestingly, the study found that 5-methylbenzotriazole exhibited greater toxicity than its unmethylated counterpart and 4-methylbenzotriazole, whereas BBT emerged as the most toxic derivative, inducing acute toxicity at concentrations as low as ≤ 3.3 mg/l to all tested organisms (Pillard et al., 2001). This research underscores the environmental and ecological risks posed by benzotriazole and its derivatives, present in greywater and runoff from industrial activities. The findings accentuate the need for developing effective wastewater treatment strategies to mitigate the release of these micropollutants into aquatic environments, ensuring the protection of water quality and aquatic life (ChatGPT4 grammar corrections).

3.3. Greywater treatment

GW treatment systems are not pioneers in our society. In the early stages of proposing acts against environmental pollution in 1980s around the world and specifically in EU, projects researching GW applications were estimating. After a decade of research and data gathered on GW parameters as written earlier, ecological engineering could deal with a more precise goal to build proper systems of treating

wastewater. Since the 1990s, over 200 schemes were introduced, where each one of them proposed a whole variety of treatment systems applications. The trend of GW research and applications hit Australia and California, USA (Radcliffe et Page, 2020). The new bloom in the industry of recycling of GW has been developing for decades and nowadays we may review most ambitious technologies in application that are left in market. The variety of GW treatment systems start with low budget solutions which show basic physical removal of dirt, grease, and other particles. Septic tanks with solids separation function were used in villages of Jordan as low income of household and lack of electrical communications would not meet operational requirements (Halalsheh et al., 2008). Most of the time, cheaper choice means safe discharge of accumulated water to the environment, often leaving the goal of reuse for medium and advanced high-end treatments systems. In most cases GW is collected from households. After collecting is made treatment involves removal of physical pollution by filtering and absorption. The following phases of the treatment go over chemical and biological pollution removal. These advanced approaches to water treatment include technologies such as: waste stabilization ponds, combination of microbes and UV in such systems like rotating biological contactors and membrane bioreactors to fully eliminate GW holding potentially hazardous chemical compounds and microbial life and viruses. Described approaches above demand more financial investments as they come close to capabilities of larger water treatment plants with less demand for space similar to technologies involving MBR (Fountoulakis et al., 2016). Even though principles have the same goal the approaches in GW management differ drastically. The goal of this study is to review cost effective approach where basic filtration principles and selected filtering materials as additives will be tested for their efficiency in filtering GW without further use.

3.3.1. Treatment of GW on TOC, TN, NH_4^+

Treatment of TOC was done by application of biologically activated carbon (BAC) in the study of Hess et Morgenroth, (2021) following with membrane

bioreactor (MBR) treatment. Biologically activated carbon (BAC) is derived from biomass such as wood which goes through pyrolysis leading into high surface area and high porosity serving as a suitable biofilter when applied in water treatment. The study lasted for over 900 days including observations on various conditions of the experiment such as: operational time, influent TOC concentrations, and empty bed contact time (EBCT). The mechanism of this treatment consisted of two processes. Absorption to biologically activated carbon (BAC) and biological degradation. Biodegradation in terms of TOC removal created conditions for microbial growth in BAC which led to pressure loss in filter bed increasing its efficacy. In the long term, EBCT did not affect the efficacy of BAC. Minor maintenance was needed as the system was backwashed only once after 800 days to relieve stress of clogging in the filter bed. The experiment could establish up to 60% removal of TOC concentrations in the effluent when BAC was used. The low-cost and low-tech properties of this greywater treatment system could be used widely to treat TOC high concentrations in greywater (Hess et Morgenroth, 2021).

Treatment of TN in research by Deng et al, (2021) in context of decentralized low-carbon greywater in rural areas introduced new approach by incorporating oxic/anoxic processes and Fe/C micro-electrolysis into vertical constructed wetland (novel system ME-(O/A)CW). This system was designated to operate at low temperatures (-11.5 to 8 °C) to effectively treat greywater. The ME-(O/A)CW system could reduce TN at 86.2% and ammonium nitrogen ($\text{NH}_4^+\text{-N}$) at 94.3%. The system had aerobic layers that facilitated effective nitrification. This layer would target specific micropollutant class (TN; $\text{NH}_4^+\text{-N}$) and following process of Fe/C micro-electrolysis of autotrophic denitrification process would lead to efficient removal of targeted micropollutants. The study identified specific microbial activities in the ME-(O/A)CW system including *Nitrosomadales* for ammonium oxidation and *Nitrospira* for nitrate oxidation, autotrophic denitrificans (e.g. *Thiobacillus*, *Hydrogenophaga*, and *Sulfurimonas*), heterotrophic denitrificans *Denitratisoma*. These microbial communities were key to confirming the reaction mechanisms within the system.

3.3.2. Treatment of GW from heavy metals Cu, Ni, Zn

The study set by Abbasi et al (2018) explores the utilization of the ornamental plant *Alternanthera ficoidea* (also known as *A. tenella*), a species widespread throughout the tropics and increasingly considered an invasive weed, as an effective agent in greywater treatment. The plant was integrated into a recently developed bioreactor system known as SHEFROL®, which demonstrated significant efficacy in purifying greywater with a wide range of pollution levels (250–1,300 mg/L COD - Chemical Oxygen Demand).

High Reduction in Pollutants: The system achieved a substantial reduction in greywater heavy metals: copper 44.8%, nickel 27.5%, manganese 38.2%, and zinc 43.2% efficiency rates. Remarkably, all these outcomes were achieved using a single pot, in a single step, and through a simple reactor operation that requires only 6 hours of hydraulic retention time. This highlights the process's efficiency and simplicity compared to more conventional treatment systems that rely on macrophytes in tanks or constructed wetlands. **Environmental and Economic Benefits:** The use of *Alternanthera ficoidea* in the SHEFROL® bioreactor presents a method of greywater treatment that is not only highly efficient in pollutant removal but also potentially less costly than traditional methods. This could offer significant environmental and economic benefits, particularly in areas where greywater recycling and reuse are critical for sustainable water management. The work demonstrates an innovative approach to managing an invasive species by harnessing its growth and adaptability for environmental remediation. This turns a problematic weed into a valuable resource for wastewater treatment (Abbasi et al., 2018).

3.3.2. Treatment of GW from pharmaceuticals (IBU, BTR)

The study conducted by Smook (2008) observed that more than 95% of ibuprofen was removed in the aeration tank of the wastewater treatment plant (WWTP), with aerobic biodegradation identified as the primary mechanism for its removal. This high removal efficiency underscores the effectiveness of aerobic processes in degrading ibuprofen within municipal wastewater treatment systems. The research also compared ibuprofen biodegradation rates between a conventional WWTP aeration tank and a membrane bioreactor (MBR) pilot plant using first-order kinetics. These findings indicate that the biodegradation rates in both systems were statistically similar, suggesting that MBR technology is as effective as conventional aeration tanks in the biodegradation of ibuprofen (Smook et al., 2008).

Another study by Finn et al. (2023) conducted experiments on IBU removal with application of coconut, lignite, and blend of powdered activated carbon (PAC) pellets. The study observed rapid contact times (0.5 – 30 mins) of IBU in deionized water or synthetic urine with these selected filtering materials. For this study mentioned coconut and lignite were powdered and later palletized both separately and together to create blend for identification of potential improvements. Coconut-derived PAC demonstrated a higher BET surface area (1088 m²/g) compared to lignite-derived PAC (509 m²/g), suggesting a potential for greater adsorption capacity. Pelletizing the PACs resulted in a decrease in the total surface area and an increase in ash content due to the inorganic binder. Surface area reductions were 14% for the coconut-based pellet (P1-coco) and 10% for the lignite-based pellet (P3-lig). P1-coco pellets took about 150 seconds to dissolve back to a powder form, while P2-blend and P3-lig pellets dissolved in less than 30 seconds. Faster disintegration, particularly for P2-blend and P3-lig, was associated with a more rapid onset of ibuprofen adsorption due to the quicker reduction in particle size. Increased pellet dosage led to higher ibuprofen removal across all tested formulations. At a dosage of 10.0 g/L, P1-coco and P2-blend showed almost identical percent removals (87% and 88%, respectively). Kinetic adsorption studies revealed that ibuprofen removal efficiency reached equilibrium at 30 minutes, with the highest removal efficiency observed for P1-coco (61%), followed by P2-blend (51%), and P3-lig (40%). The research highlights the potential of using lignite- and coconut-derived PAC in pelletized form for the efficient removal of ibuprofen from greywater, with the design and composition of the pellets playing a crucial role in optimizing removal efficiency (Finn et al., 2023).

The study by Reemtsma et al. (2010) delivers crucial insights into the challenges of removing Benzotriazole (BTR) and its derivatives, such as tolyltriazole (TTri), from wastewater. Employed widely as corrosion inhibitors in industrial applications and household products, these compounds exemplify polar and poorly degradable trace pollutants. The research indicates that the elimination rates of these compounds in four wastewater treatment plants (WWTP) in Berlin vary significantly, with removal efficiencies ranging from 20 to 70% for 5-TTR, 30 to 55% for BTR, and being negligible for 4-TTri. The effluent concentrations observed were between 7-18 $\mu\text{g/L}$ for BTR, 1-5 $\mu\text{g/L}$ for 4-TTri, and 0.8-1.2 $\mu\text{g/L}$ for 5-TTri.

Furthermore, Reemtsma et al. (2010) documented the pervasive presence of BTR and 4-TTri in the surface waters of the rivers Rhine and Elbe, noting an increase in concentrations over a distance of 600-700 km. Despite several months of residence time in bank filtration systems, used for generating raw water for drinking water production, BTR and 4-TTri were still detected at concentrations of a few hundred ng/L.

3.4. Filtration materials

Filtration systems are widely used around the world due to its simplicity compared to complex advanced wastewater treatment systems. Materials used in filtrating systems are affordable. Saw-dust, activated charcoal, sand, gravel, and other filtering materials form layers of filtration and absorption. Main function of filtration is removal of physical pollutants such as solids, lipids, and sludge before discharge or further reuse.

3.4.1. Woodchips

Woodchips can be an effective filtering material for treating GW due to their ability to remove nitrates (NO_3^-) (Abusallout et Hua, 2017). More in depth, woodchips have high surface area, which allows for the accumulation of microbial populations

that further aid in the breakdown of nitrates (NO_3^-) by converting them into dinitrogen N_2 (WOODCHIP BIOFILTERS A Best Management Practice for reducing nutrient loss in drainage water, n.d.). In general, woodchips are an inexpensive and readily available filtering material that can be used in a variety of GW treatment systems. They can be used in constructed wetlands to effectively remove contaminants from the water (Kaetzl et al., 2018). It is important to note that woodchips should be properly sourced and prepared to ensure that they do not contain harmful chemicals or other contaminants that could pose a risk to human health or to the environment. According to research of Kaetzl et al (2018), woodchips could treat COD and TOC at 90% efficiency. Although COD and TOC were effectively eliminated from the effluent, studies did not prove woodchips to be a sole solution in treating GW. The use of woodchips as a filtering material should be combined with other treatment processes, such as disinfection and sedimentation, to ensure that GW is safe for reuse or discharge.

3.4.2. Biochar

Biochar is referred to anaerobic filtration material made by heating in low-oxygen environment with a low cost and outstanding parameters for filtering greywater. Specific surface area, low bulk density, and high porosity of biochar offer its unique suitability in removal of HMs due to high cation exchange capacity and COD reduction of 90% (Kaetzl et al., 2018). The heavy metals (HMs) absorbed on biochar through cation exchange of its own (e.g., H^+ , K^+ , Na^+ , and Ca^{2+}), which can be later used by plants (Chen et al., 2007). When GW passes through biochar, the organic matter in the water is absorbed onto the surface area of the biochar. In general, biochar is a sustainable and renewable filtering material that can be used in a variety of treatment systems. However, biochar should be combined with other filtering materials to reach GW reusability or discharge (Kaetzl et al., 2018).

There are variety of biochar types with different characteristics such as pore size, surface area, and absorption capacity, as well as any potential contaminants that may be present in biochar (Quispe et al., 2022). Nutshell biochar is a type of biochar which is made of shells of nuts, such as almond, coconut, or pecan shells. It has a high absorption capacity and can be effective at removing contaminants and nutrients from

GW (Ahmedna et al., 2004). Sewage sludge-based biochar is made of organic matter in sewage sludge. It can be effective at removing nutrients and contaminants from GW but may contain pathogens and HMs, which may be a concern in using this type of biochar for filtration (Xing et al., 2021). Bone char is made from the bones of animals, typically from the beef industry. It is highly porous and has the capacity to remove pollutants at higher volume. A bone char consists of calcium carbonate and calcium phosphate. It removes excessive amounts of fluoride in water through absorbing and replacement of carbonate with fluoride ion (Ali et al., 2017). Wood-based biochar is the most common biochar used for GW treatment. It is made of a variety of wood sources, including softwoods and hardwoods, and can be produced at different temperatures and for different durations resulting in different pore sizes and surface area. Wood-based biochar is recommended for filtering purposes due to its ability to reduce ammonia according to a critical review by Taghizadeh-Toosi et al., 2011. The review provides insightful observations on the impact of biochar addition to urine nitrogen (U-N) treatments on ammonia losses. It was found that incorporating biochar led to a significant reduction in cumulative ammonia losses, which were 45% lower after a span of 29 days. Specifically, in scenarios where 15U and 30U amounts of urine-N were applied, the ammonia losses were remarkably minimal, constituting only 0.79% and 0.78% of the applied urine-N, respectively. This is in stark contrast to the 0U treatment, where no urine-N was applied, and the ammonia loss was significantly higher at 1.42% of the applied urine-N. The analysis further reveals that within the initial 24 hours, the ^{15}N enrichment of volatilized ammonia-N was notably higher at 7.88 atom% in the 0U treatment, indicating an increased loss rate initially. However, this trend reverses after 52 hours, with the ^{15}N enrichment in volatilized ammonia-N being lower in the 0U treatment compared to the 15U and 30U treatments, suggesting a reduction in the rate of loss over time. This data underscores the efficacy of biochar in mitigating ammonia volatilization from urine-N and highlights the dynamic nature of ammonia loss rates depending on the presence of urine-N and the application of biochar.

When used as a filter material, biochar can be added to the filter bed in concentrations ranging from 1% to 10% by volume. Here are some characteristics of biochar when used at a 5% concentration in GW treatment:

Nutrient removal: Biochar 5% can be effective at removing nutrients, such as nitrogen and phosphorus, from GW. At a 5% concentration, biochar can remove up to 60% of nitrogen and 80% of the phosphorus in GW (Clough et al., 2013).

Absorption capacity: Biochar has a high absorption capacity, which allows it to trap and remove contaminants from GW. At a 5% concentration, biochar removes 70% of the organic matter in GW (Wang et Wang, 2019).

pH stabilization: Biochar has a buffering effect that can help to stabilize the pH of GW. Biochar 5% can help maintain neutral pH in GW to ensure healthy plant growth (Clough et al., 2013). Water retention: High porosity helps retain water and release it slowly over time. This can help the filter from drying out and maintain stable moisture content (Clough et al., 2013). Kaetzl et al. (2018), have constructed research on comparing of conventional and relatively new kinds of media for treating sewage water. Table 2 represents mean effluent concentration and removal rates of biochar over experimental time. Standard deviations are presented as variation range.

Table 2. results of using biochar as anaerobic filter media. Mean effluent concentration and removal rates. Different letters (a,b,c) indicate a significant difference of the mean (ANOVA, $p < 0.05$) between materials after post hoc analysis (Kaetzl et al., 2018).

Parameter	Biochar	
	Concentration	Removal
	($\text{mg}\cdot\text{L}^{-1}$)	(%)
COD	48 ± 19	$87^b \pm 2.6$
TOC	31 ± 9.1	$77^b \pm 3.6$
N_{tot}	61 ± 10	14 ± 8.1
P_{tot}	2.3 ± 0.3	13 ± 9.0

According to results of the study, biochar has shown effective treatment of medium-polluted wastewater, particularly in COD treatment. TN and TP did not show any outstanding removal. Further treatment is needed. However, for irrigation purposes high content of N and P may let avoiding of agricultural fertilizers (Kaetzl et al., 2018).

3.4.3. Limestone

Limestone is commonly used as a filter medium for GW treatment because of its chemical properties, including its ability to neutralize acidity due to its high alkalinity. Being inexpensive makes it widely available even in developing countries. Limestone is a natural material which found its use in sand filters, constructed wetlands, and gravel filters (Li et al., 2009). Limestone in sand filters is added to layer with sand to help neutralize pH of GW as it passes through (Imran et Aznam, 2023). In constructed wetlands, limestone can be used as a substrate material for the plants, which helps to maintain stable pH in the system. In gravel filters, limestone can be used as one of the layers of filter media, along with sand and gravel, to create a multi-layer system. When using limestone as a filter media it's important to consider what type and quality of limestone is used.

Dolomitic limestone is another choice. In addition to calcium carbonate content, it also contains magnesium carbonate, which can be beneficial for plants and soil health in addition to neutralizing pH in the system (Kamprath et Smyth, 2005). Crushed limestone is often used in gravel filters, as irregular shape of crushed limestone helps to achieve porous filter bed (Chen et al., 2009). Hydrated limestone is chemically treated to increase its alkalinity, making it even more effective at neutralizing acidity in GW (Ghaly et al., 2021). When selecting a type of limestone as a filter media, it is important to consider its particle size, uniformity, and calcium carbonate content for removal of selected micropollutants if known.

4. Methods

The course of the experiment took part from 26.06.2021 to 28.10.2021. with 3 weeks break (12.08.2021 – 2.09.2021) to decrease saturation volume from 70% to 30% in barrels used on the experimental site of CZU. The experiment consisted of preparation of the experiment site, mixing synthetic grey water (SGW), field work, laboratory analysis and statistical evaluation of collected data over the course of experiment.

4.1. The experiment site and equipment

Experimental site included in total of 40 barrels which represented physical model of infiltration trenches filled with filtering materials (additives: woodchips, limestone, biochar 5% and default structure with no additives) divided into groups by saturations (70% and 30%) to gather data on a difference in filtering efficiency. The total was divided into sets of 5 barrels and put on two logistics pallets to create elevation for convenience of gathering of GW effluent samples. Each set of 5 barrels had similar content (replica) and to exclude influence of external conditions which could potentially distort the results or result in data loss if barrel was damaged. The experiment site included 1000L tank for SGW influent and 600L tank for disposal of sample leftovers. Both tanks were equipped with suction pump system for irrigation (influent) and disposal (GW sample leftovers). Irrigation system with extended hoses installed in every barrel was introduced in later course of experiment replacing 2/3 of conventional way of irrigation by garden pots replacing heavy lifting and providing gradual irrigation of testing additives in barrel sets for more quality infiltration of influent SGW to the tested filtering belts.

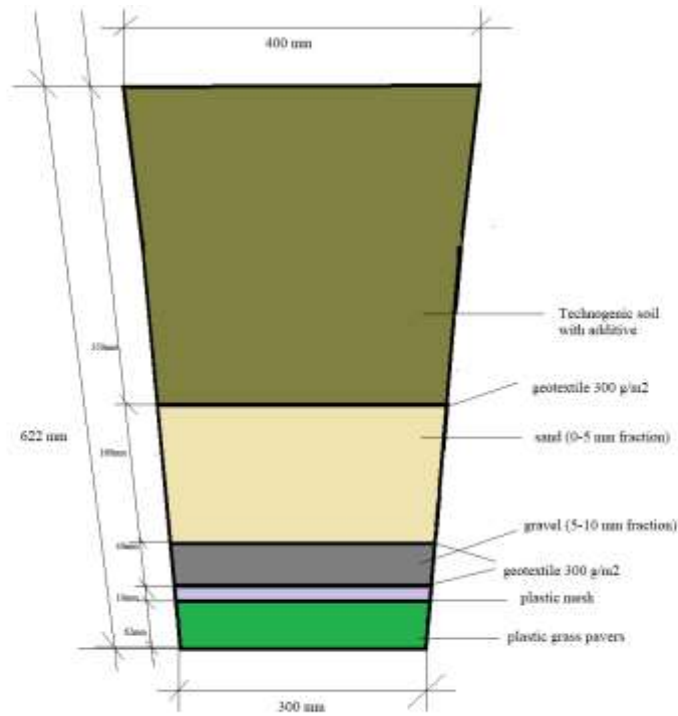
Figure 1: Experimental site, barrels with tested additives.



4.1.1. Physical models design

Barrels made of plastic, 60 cm high, 40 cm wide on the top and 30 cm wide on the bottom were used to construct the physical model of infiltration trenches. Geotextile was used to prevent preferential flow development. Geotextile was also used as a layering component for separation of absorption layers. As barrels were made of white plastic there was a necessity to cover them with black foil to avoid sun exposure which would lead to an uncontrollable growth of algae in the barrels. Each barrel was installed with a tap on the bottom for sample collection. There was a specific order in filling barrels which was in this order from bottom to top: starting with plastic grass pavers, plastic mesh, gravel, sand and technogenic soil with testing additives.

Figure 2: Physical model of filter material layers



Technogenic soil consisted of compost, sand, and topsoil. This content would be tested with the addition of filtering materials (additives) of interest such as woodchips, limestone, biochar 5%. Default content with no filtering additive was also on the site for having a reference of efficiency (Tab. 3). Meadow mix vegetation grew on top the soil serving as a bioindicator for upcoming changes in soil as well as protecting soil degradation.

Table 3: Filtering layers ratio of testing additives in barrels. Additives proportions mixed into natural soil.

Testing additive:	Proportions:
	sand : compost : natural soil : additive
Default	5 : 3 : 2
Woodchips	5 : 2 : 2 : 1
Limestone	5 : 3 : 2 : 1
Biochar 5%	10 : 5 : 4 : 1

4.1.2. Synthetic greywater (SGW)

Greywater in the whole experiment was synthetically made each time before irrigation. Formula of the synthetic greywater (SGW) was created by the lab staff based on prior research on GW content with proximity due to the known fact that GW content varies from household to household. Despite the range of parameters that differ, prior online survey handled by CZU FES personnel determined brands of home and hygiene appliances which were used the most around the Czech Republic. According to calculations and research, the following recipe for SGW was created with adaptation of Abed et Scholz (2006) and Diaper et al (2008) recipe for the experiment by the laboratory of Faculty of Environmental Science (Tab. 4). The following hygiene products listed in the table below consisted in the SGW to simulate organic and inorganic pollution for further studies during the experiment (tab. 4).

Table 4: Recipe for synthetic greywater per 1000 L tap water. Adapted recipe from Abed et Scholz (2006) and Diaper et al (2008) was used to recalculate measures of individual contaminant sources to fit ratios needed for the assessment with respect saturations (30% and 70%).

Contaminants:	Specifications:	The amount for 1000L of tap water (70% saturation):	The amount for 600L of tap water (30% saturation):	Units
Shampoo	Head & Shoulders	72	43.2	g
Soap	Dove	648	388.8	g
Body shower gels	Nivea	10	6	g
Toothpaste	Colgate	32.5	19.5	g
Deodorant	Nivea	10	6	g
Laundry detergents	Ariel	150	90	g
C ₆ H ₅ N ₃ .	Benzotriazole (BTR)	0.05	0.05	g/L
C ₁₃ H ₁₈ O ₂	Ibuprofen (IBU)	0.01	0.01	g/L
CuSO ₄ ·5H ₂ O	Copper	0.2	0.2	g/L
ZnSO ₄ ·7H ₂ O	Zinc	0.2	0.2	g/l
NiSO ₂ ·6H ₂ O	Nickel	0.2	0.2	g/L
H ₃ BO ₃	Boron	1	1	g/L

4.1.3. Field work on experimental site

The experiment began on June 28 and comprised three distinct phases. Initially, tap water was used to flush the filtration materials in all barrels, removing any built-up residues and impurities that typically accumulate, followed by the collection of composite samples from every group. The second phase involved a kinetic study designed to assess the effectiveness of Greywater permeation at varying retention intervals: 2 hours, 4 hours, 24 hours, and 4 days, with individual samples gathered from each barrel for lab analysis. This phase served purely as a control. In the third phase, effluent was filtered, and samples were taken bi-weekly; on Mondays after a 72-hour retention period and on Thursdays after 22 hours of operation. Subsequently, synthetic greywater (SGW) was used for irrigation at two different saturation levels, 70% and 30%. Post-interval, there was an adjustment in the hydraulic load volume, decreasing from 70% to 30% to investigate the impact of reduced hydraulic load on infiltration efficiency.

4.2. Laboratory analyses

End of an each procedure of watering and GW samples collection were followed by laboratory water quality tests which included determining of pH, EC, turbidity, heavy metals (HM) of interest such as Copper (Cu), Nickel (Ni), Zinc (Zn), and semimetal Boron (B), total organic carbon (TOC), total carbon (TC), inorganic carbon (IC), total nitrogen (TN), ammonium ion (NH_4^+), selected benzotriazole (BTR) and Ibuprofen (IBU).

4.2.1. pH, EC, Turbidity analysis:

Tests always started with pH measurement with “Lab pH meter inoLab® pH 7110” (Fig. 3). EC analysis follows next with “Conductivity benchtop meter InoLab®

Cond 7110” (Fig. 4). Turbidity analysis was next before filtration of samples to ensure proper data using “HI-93703 Portable Turbidity Meter” (Fig. 5). The devices mentioned above were easy to use and no prior sample adjustments were needed.

Figure 3: Lab pH meter inoLab® pH 7110



Figure 4: Conductivity benchtop meter InoLab® Cond 7110



Figure 5: HI-93703 Portable Turbidity Meter



4.2.2. NH_4^+ analysis:

The measurement of ammonium ions (NH_4^+), the filtration of samples was a critical step to prevent any large particles from interfering with the precision of the measurements. This was achieved by using a 10 ml syringe fitted with a "Millipore Millex – GP Hydrophilic PES filter unit, having a pore size of $0.22 \mu\text{m}$ and a diameter of 33 mm," directing the samples into designated tubes prepared for the NH_4^+ analysis. Next step involved indophenol blue method for revealing of NH_4^+ values in the tested content. 4 ml of tested sample was mixed with 0.4 ml of coloring agent (NH_4^+), 0.4 ml of alkaline solution (N- NH_4^+), and 0.2 ml of distilled water (tab. 5). Blank sample which contained 4 ml of distilled water instead of SGW sample with respect to ratio of additives according to indophenol blue method was also analyzed along with other samples to have better overview of the results. The mixtures were thoroughly homogenized using a "Vortex RX3 VELP® Scientifica". After thorough mixing, the samples were allowed to stand for 1 hour away from sunlight, facilitation the required chemical reactions. Once prepared, the GW samples for the NH_4^+ were analyzed

measuring the absorbance by “Cary 60 UV-Vis Spectrophotometer” set to wavelength of 190-1100 nm (Fig. 6).

For the next steps of analysis filtering of samples was essential as devices were extremely sensitive to any large particles which would disturb precise measurement. Filtering of samples were handled using 10 ml syringe with attached filter unit “Millipore Millex – GP Hydrophilic PES pore size 0.22 μm , diam. 33 mm” right into prepared sets of empty tubes for ammonium ion (NH_4^+), total organic carbon (TOC), total carbon (TC), inorganic carbon (IC), and total nitrogen (TN).

Table 5: GW sample preparation for measuring for NH_4^+ with “Cary 60 UV-Vis Spectrophotometer.”

GW sample:	Coloring agent ammonium ion (NH_4^+):	Alkaline solution ammonia nitrogen (N-NH_4^+):	Distilled water:	Total volume:
Volume in [ml]				
4	0.4	0.4	0.2	5

Figure 6: Cary 60 UV-Vis Spectrophotometer



4.2.3. TC, TOC, IC, TN analysis:

Measurements of Total Organic Carbon (TOC), Inorganic Carbon (IC), Total Carbon (TC), and Total Nitrogen (TN) were carried out using a Skalar Formacs^{HT} TOC/TN Analyzer, into which a 15ml test tube containing the filtered sample was introduced (fig. 7). The Formacs analyzer employs a method of high-temperature catalytic combustion to quantify the levels of nitrogen and carbon fractions within the liquid samples. TOC is determined by first measuring TC and IC, where TC is derived through catalytic oxidation at high temperatures, transforming both organic and inorganic carbon in the sample to Carbon Dioxide (CO₂). The amount of CO₂ generated is then quantified following acidification, allowing for the calculation of TOC as the difference between TC and IC (TOC = TC – IC). In this method employed by Skalar Formacs^{HT} converted all forms of nitrogen present in samples, including organic nitrogen compounds, ammonia, nitrate and nitrite, into nitrogen oxides. These nitrogen oxides are further reduced and quantified as elemental nitrogen or nitrogen dioxide (NO₂), allowing for the accurate measurement of TN in the sample.

Figure 7: Formacs^{HT} TOC Analyzer by Skalar



4.2.4. Heavy metals analysis:

Analysis for heavy metals Cu, Zn, Ni and semi-metal B were held alongside. Separately collected samples with unfiltered GW (10 ml) were mixed with 0.25 ml of

nitric acid (HNO₃). Boron, and heavy metals (copper, nickel, and zinc) were analyzed by an inductively coupled plasma optical emission spectrometry (ICP-OES, Agilent 730, Agilent Technologies, USA) in the laboratory of the Department of Environmental Geosciences CZU. Collected data over the course of experiment were inserted in Excel tables for further statistical analysis.

4.2.5. Pharmaceuticals analysis

IBU and BTR measurements were carried out in an external laboratory. The samples were purified by solid phase extraction (SPE) with Oasis Prime HLB cartridges (200mg, 6 ml) from Waters (Milford, MA, USA) conditioned with 5 ml of methanol and 5 ml of water. After sample loading (50 ml), the cartridges were washed with 2 ml of water and further eluted with 5 ml of methanol and 5 ml of MeOH:H₂O with 0.1% formic acid. The elutes were analyzed by liquid chromatography with tandem mass spectrometry “Agilent 1290 Infinity II” (LC-MS/MS).

4.3. Statistical evaluation of collected data

Data gathered during the laboratory tests were recorded in Microsoft Excel. Tables with data were rearranged into raw data to evaluate them further in statistical software RStudio (2022.12.0 Build 353 © 2009-2022 Posit Software, PBC).

Data for most important parameters including micropollutants were reviewed individually in RStudio. To do so data had to go through Shapiro-Wilk test of residuals normality to define other assumptions. P-value in Shapiro-Wilk normality test was an indicator to decide between parametric and non-parametric calculations. If P-value was above indication of 0.05 meaning normal distribution of residuals, ANOVA parametric test method was used. Tukey HSD test processed statistically significant

differences between groups. In cases where residuals did not show normal distribution non-parametric Kruskal-Wallis test was used in addition with Dunn test.

The removal was calculated using the following formula: $E = \frac{C_{in} - C_{total}}{C_{in}} * 100$.

Where: E – removal efficiency, C_{in} – influent concentration (mg/L), C_{total} – effluent concentration (mg/L). Results were also converted from mg/L to removal efficiency percentages where it was necessary using following formula: $E_{per} = 100 - \left(\frac{E * 100}{1}\right)$.

Where E_{per} – concentration in percentages (%).

5. Results

SGW content of total organic carbon (TOC), total carbon (TC), inorganic carbon (IC), total nitrogen (TN), ammonia (NH_4^+), heavy metals (HMs): copper (Cu), nickel (Ni), zinc (Zn), semi metal boron (B), and pharmaceuticals ibuprofen (IBU) and benzotriazole (BTR) reduction in effluent by selected additives to infiltration trenches are presented individually. Results were divided data tables and figures with respect to contact time (22 hours and 72 hours) and saturation (30% and reduction of saturation from 70%-30%) and represented 4 experimental setups (a. results after 72 hours of contact time at 70 -30 % saturation; b. results after 72 hours at 30% saturation; c. results after 22 hours at 70 – 30% saturation; d. results after 22 hours at 30 % saturation).

5. 1. Efficacy in removal of total carbon (TC), total organic carbon (TOC), inorganic carbon (IC)

The experiment on TC removal from effluent using woodchips, limestone, biochar 5% and default structure with no additives did not achieve sufficient removal rates across 4 experimental when dosed with 76.29 mg/L TC in influent. It must be noted that over the course of the experiment, it was observed that TC concentrations generally decreased across all setups but did not decrease below influent TC dose. This reduction was particularly noticed after 72 hours of contact time, with initial saturation levels starting at 70 % and later dropping to 30%. Woodchips emerged to be more successful among other tested filter materials including default with removal efficiency of 41.16% Despite this, when evaluating mean efficiencies of all tested groups, none were reliable in consistently removing TC. A decrease in saturation (70 -30%) was linked to stabilization of TC levels in all 4 experimental setups, suggesting that lower saturation could prevent TC concentrations from increasing again. When looking at TC concentrations in 6 months timeframe across all setups, TC levels tended to decrease by the end of experiment in most tested filter materials (FMs) particularly after 72 hours of contact time (See appendix No 1-5.).

TOC removal was conducted with an initial dose of TOC 66.4 mg/L across all experimental setups. Findings from TOC removal had positive results in all tested FMs including default. The decrease in saturation in the middle of the experiment (70 – 30%) did not show significant affect the overall decreasing trend (Fig. 8). This suggests that TOC removal was effective regardless of both high and low saturations. The range of mean efficiency in TOC removal varied among the tested FMs, with woodchips moderate removal efficiency of 53.67 % and biochar 5% achieving the highest removal efficiency of 66.12 % after 72 hours of contact time and 70 – 30% saturation (Fig. 9). Similar trends were observed in samples collected after 72 hours of contact time and 30% saturation, TOC levels generally decreasing below the initial dose. Biochar 5% consistently showed the highest removal efficiency across all experimental setups. A notable finding was an unexpected exceedance of TOC concentrations in default structure samples, and it is suggested that it occurred due to technical errors as other tested FMs across all experimental setups did not show similar changes in trend (Fig. 8).

Figure 8. Concentration of TOC after application of filtering materials during the experiment. A. 70-30% saturation after 72 hours. B. 30% saturation after 22 hours. Red timeframe indication. C. 70-30% saturation after 22 hours. D. 30% saturation after 22 hours. C. 70-30% saturation after 22 hours. D. 30% saturation after 22 hours.: 3 weeks break (12/08/2021-2/9/2021).

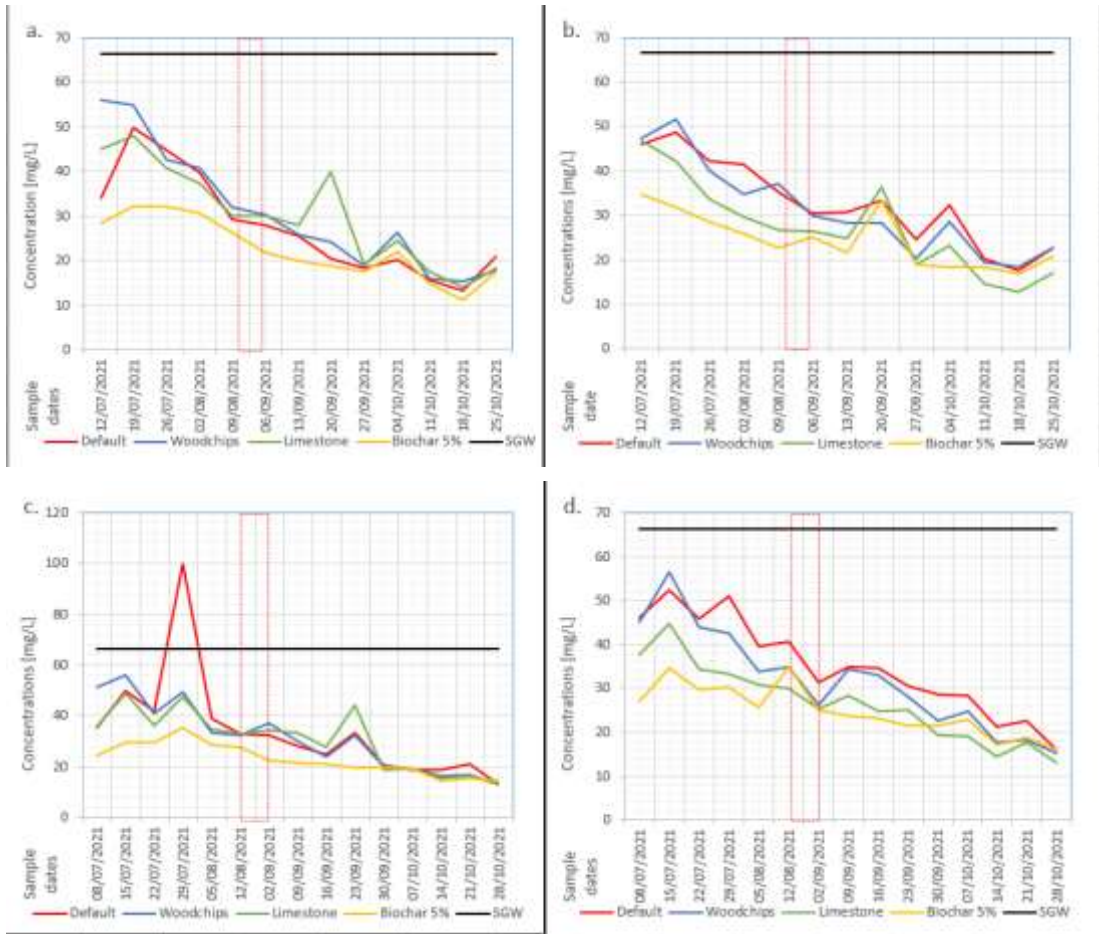
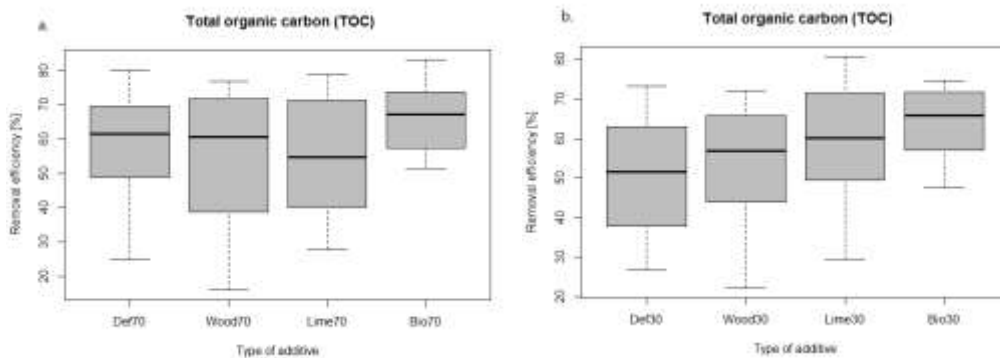
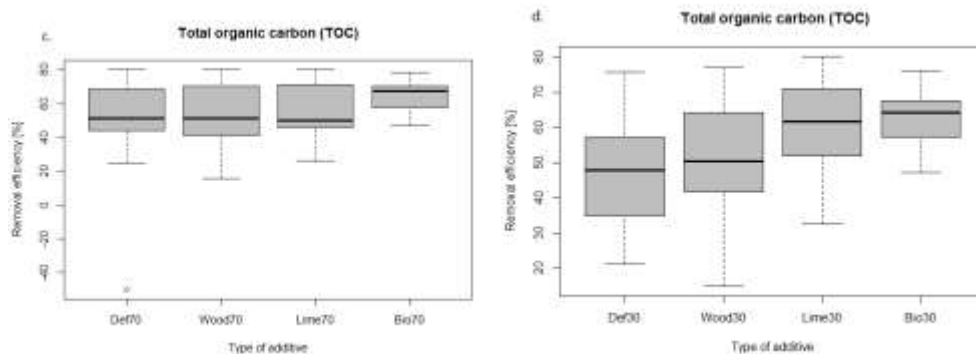


Figure 9. TOC removal efficiency. after application of filtering materials during the experiment. A. 70-30% saturation at 72 hours. B. 30% saturation at 22 hours. C. 70-30% saturation at 22 hours. D. 30% saturation at 22 hours.





IC concentrations across the experiment had a decreasing trend in all tested FMs. However, none of the filters including default were able to reduce the IC concentrations below the influent dose of 13.19 mg/L, indicating overall insufficiency in the removal process. A decrease in saturations (70 – 30 %) was observed to cause an increase in IC levels across all tested filter materials, notably after 3 weeks of break during the experiment. This suggests that lower saturation levels combined with interruptions in the process negatively affected the ability of FMs to remove IC from effluent (See Appendix No. 11-14).

5. 2. Efficacy in removal of total nitrogen (TN) and ammonia ion (NH_4^+)

The removal of TN was highly efficient across all tested filter materials, showing better performance compared to TC and IC. This indicates a strong capability of the materials to decrease TN concentrations under the conditions tested. At 70% saturation, biochar 5 % was dominant compared to other tested FMs with efficiency of 77.48 %, closely followed by default (no additives) at 75.47 %, limestone at 74.15% and woodchips at 71.04 % (Fig. 10., a.; Fig. 16., c.). This demonstrates that even without applied FMs TN concentrations could drop below the influent dose (7.76 mg/L).

Figure 10. Concentration of TN after application of filtering materials during the experiment. A. 70-30% saturation after 72 hours. B. 30% saturation after 22 hours. Red timeframe indication: 3 weeks break (12/08/2021-2/9/2021).

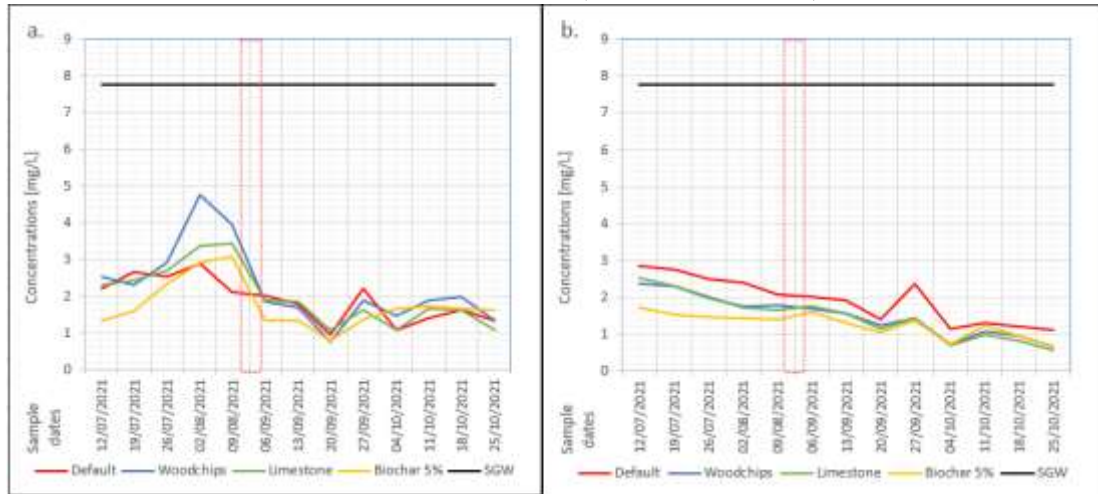
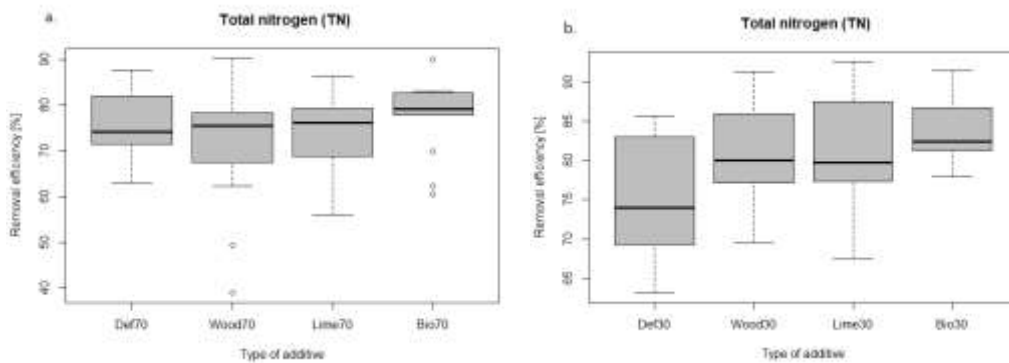


Figure 11. TN removal efficiency. after application of filtering materials during the experiment. A. 70-30% saturation at 72 hours. B. 30% saturation at 22 hours.



When saturation was reduced to 30%, biochar 5 % again showed the highest removal efficiency, this time increasing to 83.76 %. Woodchips and limestone also showed improvement in lower saturation with efficiencies close to each other (80.6 % and 80.99 %, respectively) (Fig. 11., b). This suggests that lower saturations may enhance the TN removal efficiency of certain FMs. A general decreasing trend in TN concentrations was observed across all tested materials when saturations changed from 70 – 30 %, possibly indicating TN stabilization by the end of the experiment (Fig. 10., a; Fig. 12., c). Tested FMs improved efficiency in TN removal by 5.89% - 9%, indicating only slight improvement in treatment when such additives are used.

Figure 12. Concentration of TN after application of filtering materials during the experiment. C. 70-30% saturation after 22 hours. D. 30% saturation after 22 hours. Red timeframe indication: 3 weeks break (12/08/2021-2/9/2021).

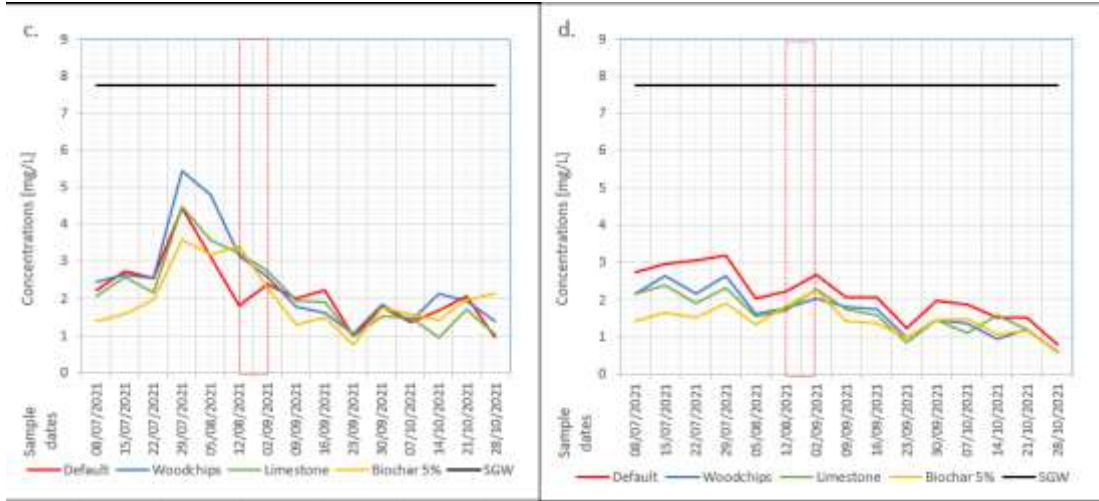
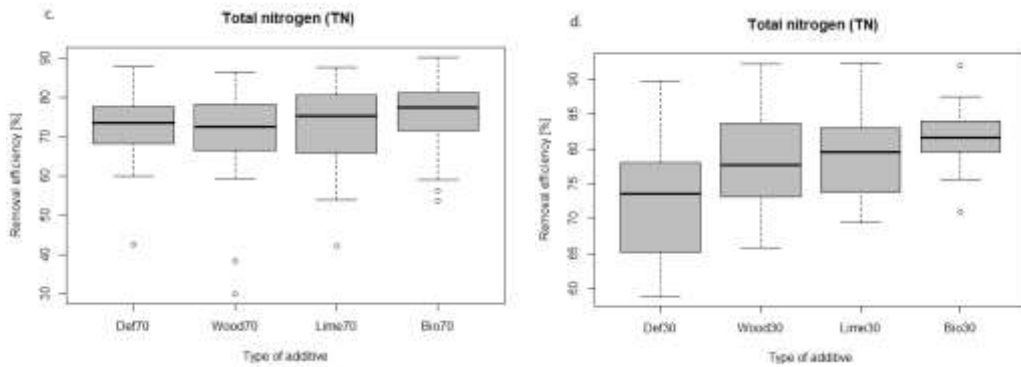


Figure 13. TN removal efficiency. after application of filtering materials during the experiment. C. 70-30% saturation at 22 hours. D. 30% saturation at 22 hours.



5. 3. Efficacy in removal of Boron (B), Copper (Cu), Nickel (Ni), and Zinc (Zn)

The experiment showed inefficacy in Boron (B) removal across all experimental setups as there was no significant success in reducing B concentrations below the influent dose of 0.97 mg/L. The tested FMs did not demonstrate effectiveness as concentrations generally maintaining or exceeding the influent dose. The initial phase of the experiment witnessed a rapid increase in B concentration in all tested FMs within the first 2.5 weeks, reaching and sometimes exceeding the dose. This trend was consistent across all experimental setups, suggesting that all tested FMs along with default support the presence of Boron in the effluent rather than effectively removing it. Moreover, 3 weeks of break could lower B concentrations below the dose as no boron was constantly adding physical models, suggesting that longer contact time is effective but not sufficient. In summary, these selected FMs at designated experimental setups were not suitable for effective B removal (See appendix No. 25 – 29).

The results from Cu removal showed significant efficacies across all experimental setups in all tested FMs, with concentrations dropping from the initial influent dose of 0.21 mg/L. This trend was consistent in all observed groups, indicating a high efficiency range from 99.02 % (woodchips) - 99.25 % (biochar 5%) in copper removal. However, this also stated that no FMs needed to effectively remove Cu from effluent as default structure, without any additives, also accomplished effective Cu removal over the course of the experiment with efficacy of 99.13 % (Fig. 15., b.). Conditions of the experiment with different setups showed smooth progression of Cu concentrations in effluent in all tested FMs and default at lower saturation (30 %) and shorter contact time (22 hour) (Fig. 14., d.).

Figure 14. Concentration of Cu after application of filtering materials during the experiment. A. 70-30% saturation after 72 hours. B. 30% saturation after 22 hours. C. 70-30% saturation at 22 hours. D. 30% saturation after 22 hours. Red timeframe indication: 3 weeks break (12/08/2021-2/9/2021).

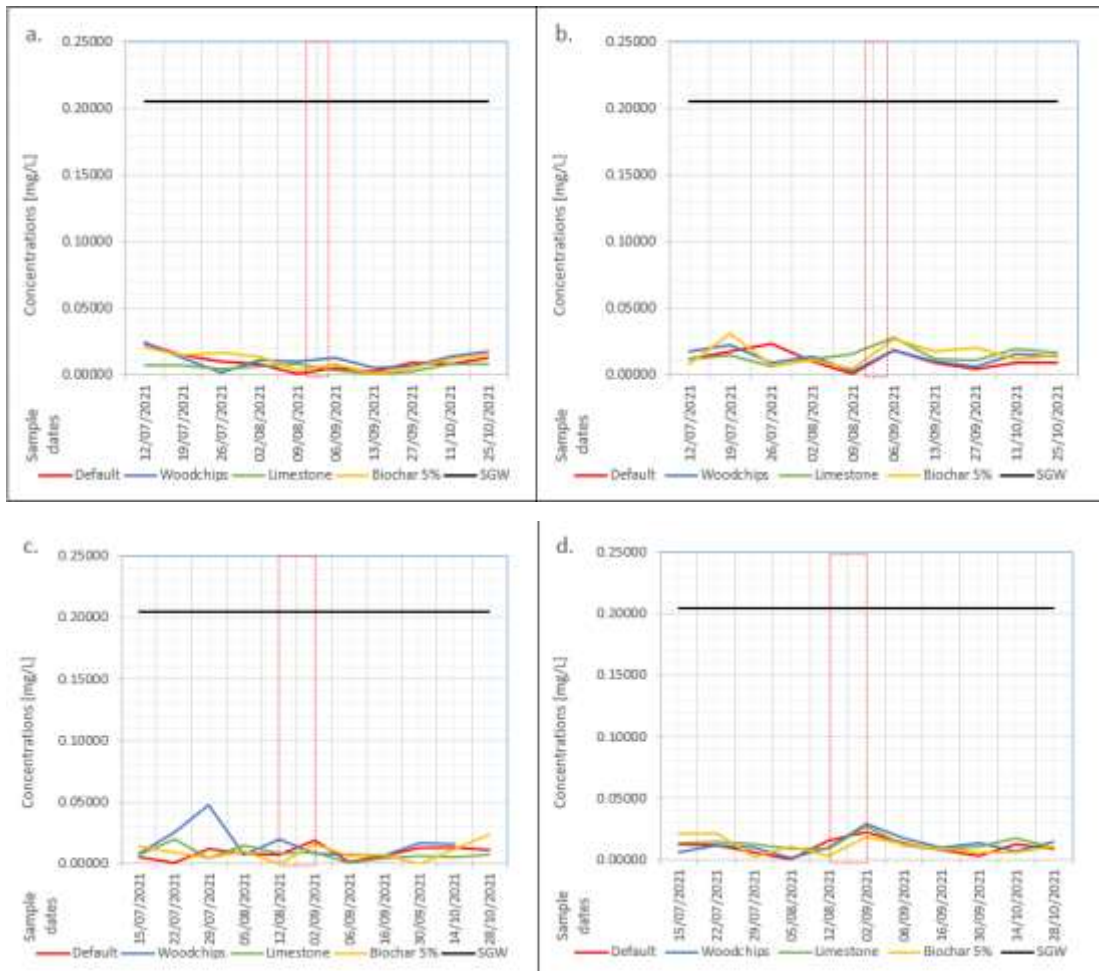
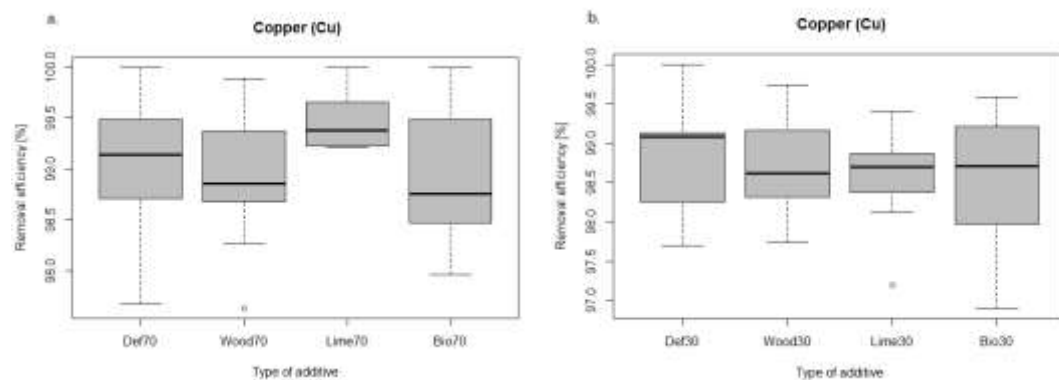
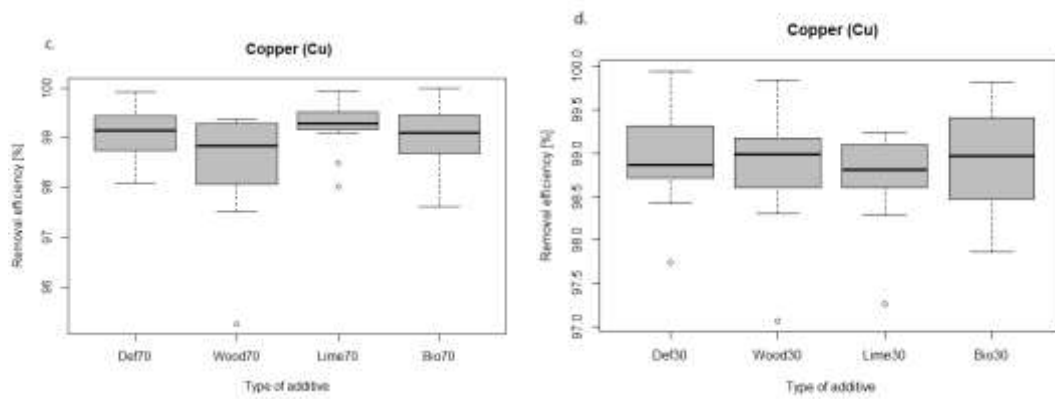


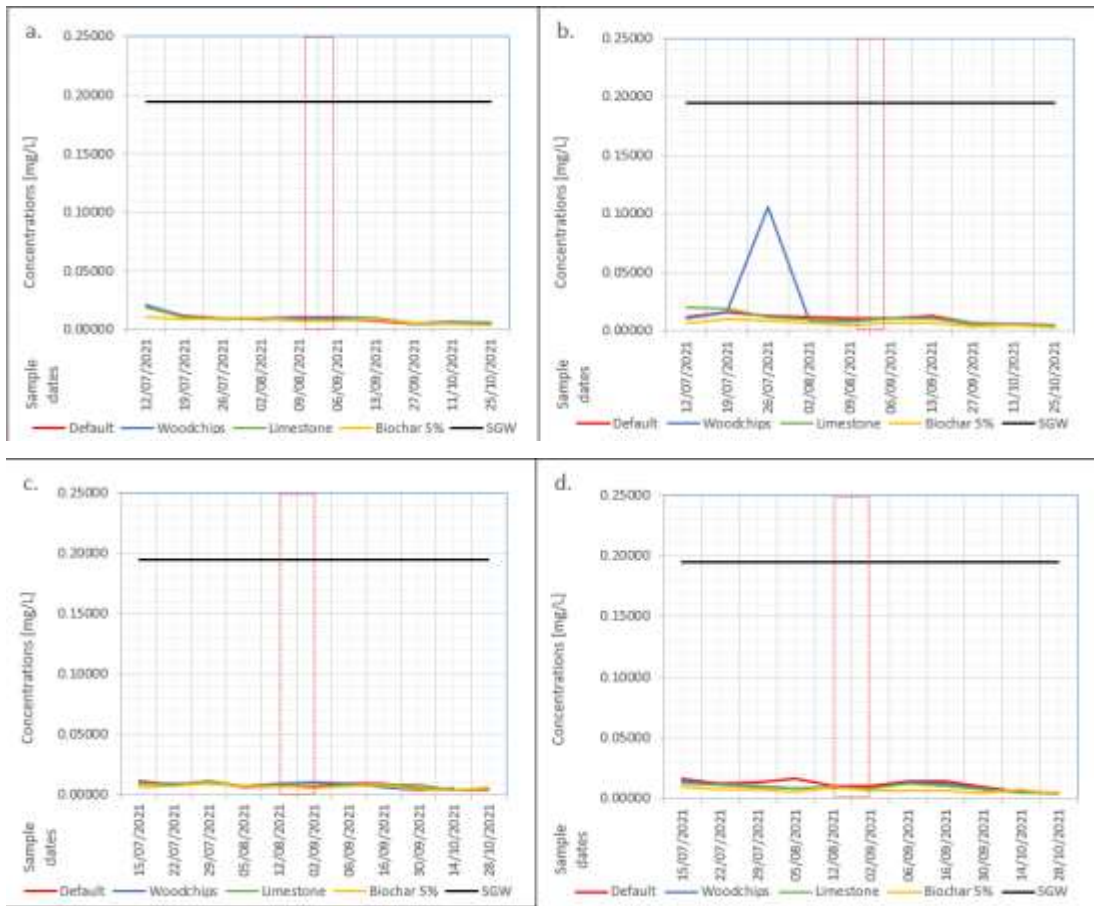
Figure 15. Cu removal efficiency after application of filtering materials during the experiment. A. 70-30% saturation after 72 hours. B. 30% saturation after 22 hours. C. 70-30% saturation at 22 hours. D. 30% saturation at 22 hours.





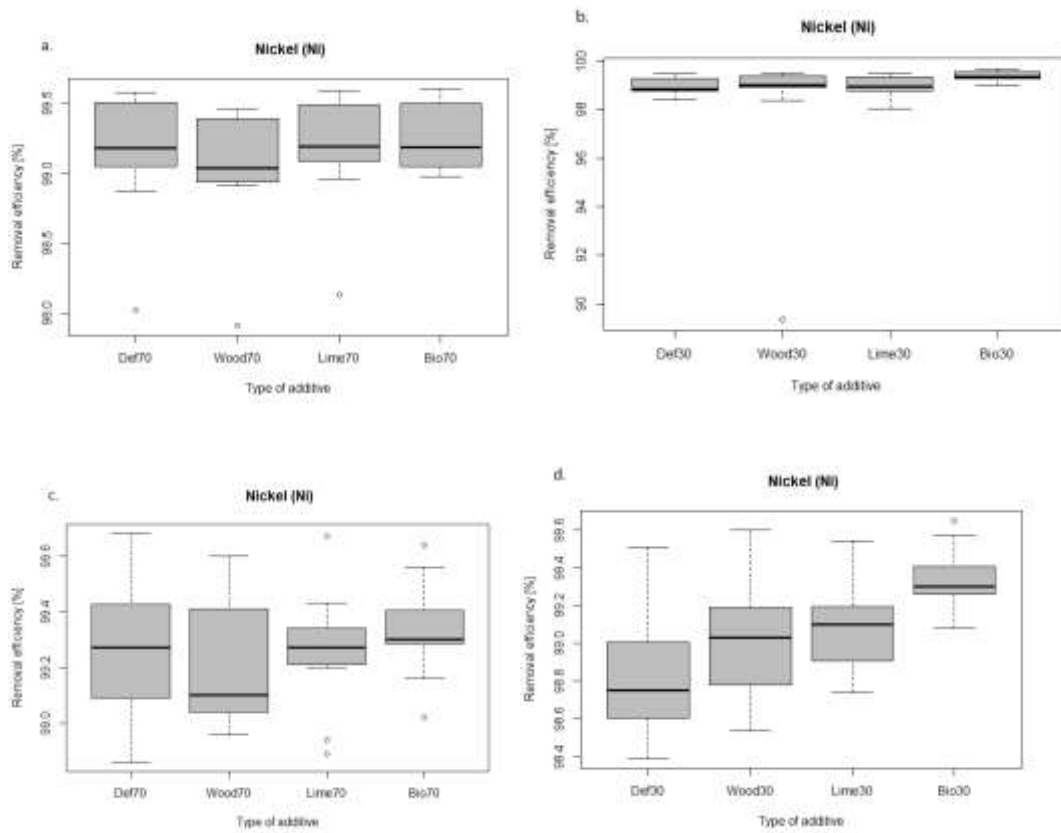
The results on nickel (Ni) removal stated significant efficiency in reducing Ni concentrations below influent dose of 0.19 mg/L in all tested FMs across all experimental setups. A sudden increase in Ni concentrations occurred in the woodchips material at 30% saturation after 72 hours of contact time, potentially indication data anomaly in the 4th week as similar changes were not observed in remaining cases. Aside from that, all tested FMs showed smooth removal (Fig. 16., b.).

Figure 16. Concentration of Ni after application of filtering materials during the experiment. A. 70-30% saturation after 72 hours. B. 30% saturation after 22 hours. C. 70-30% saturation at 22 hours. D. 30% saturation after 22 hours. Red timeframe indication: 3 weeks break (12/08/2021-2/9/2021).



Limestone had the highest removal rate among other FMs (99.48 %). However, remaining FMs were close to limestone where woodchips had the least efficiency of 98.88 %, biochar 5% of 98.93 %, and default with 99.07 %. Results stated that filter materials are not needed for effective treatment of Ni from effluent as default structure with no additives showed high removal rate (Fig. 17).

Figure 17. Ni removal efficiency after application of filtering materials during the experiment. A. 70-30% saturation after 72 hours. B. 30% saturation after 22 hours. C. 70-30% saturation at 22 hours. D. 30% saturation at 22 hours.



Results on Zn removal demonstrated success in all tested groups including default structure as Zn concentrations fell below the influent dose of 0.22 mg/L across all experimental setups. In the first half of the experiment concentrations remained remarkably low, close to 0 mg/L in all groups. However, an increase in Zn concentrations occurred from 5th week, continuing until the end of the experiment. The growth coincided with 3 weeks break (no influent), suggesting an impact of this break on Zn levels but changes occurred below the influent dose (Fig. 18). Limestone had the highest removal rate of 99.16 %. Other groups also showed high removal rates ranging from 98.49 % to 98.99 %, proving that default structure with no additives was able to remove Zn concentrations at significant rates (Fig. 20).

Figure 18. Concentration of Zn after application of filtering materials during the experiment. A. 70-30% saturation after 72 hours. B. 30% saturation after 22 hours. C. 70-30% saturation at 22 hours. D. 30% saturation after 22 hours. Red timeframe indication: 3 weeks break (12/08/2021-2/9/2021).

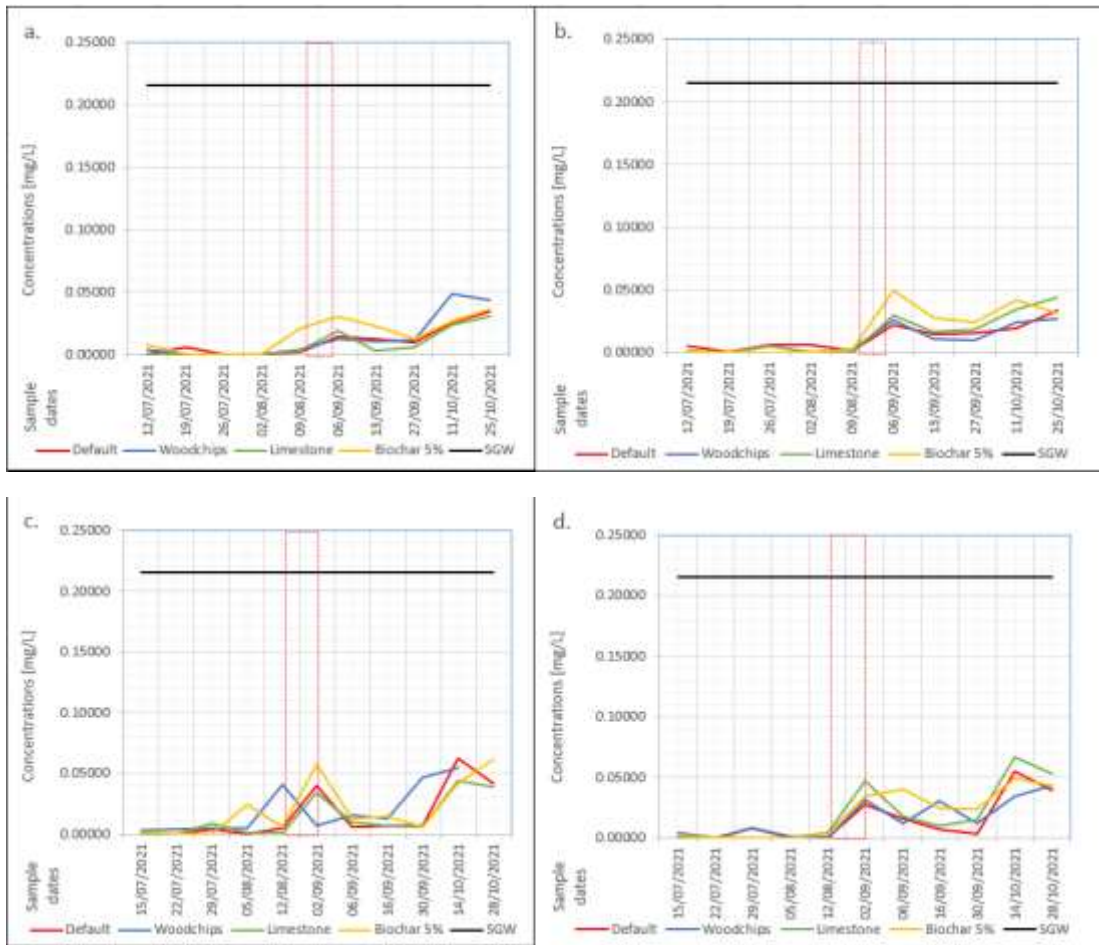
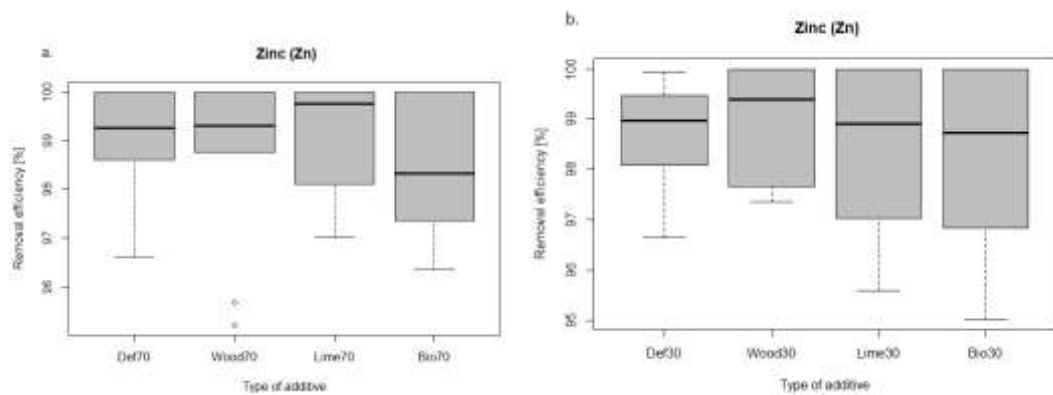
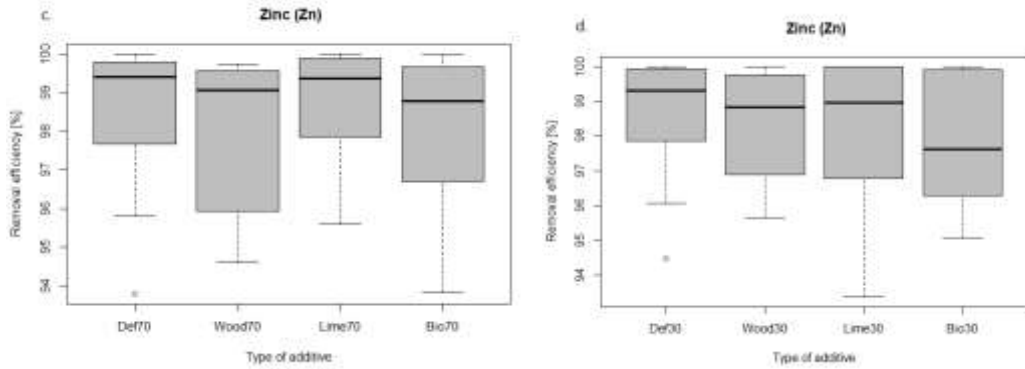


Figure 20. Zn removal efficiency after application of filtering materials during the experiment. A. 70-30% saturation after 72 hours. B. 30% saturation after 22 hours. C. 70-30% saturation at 22 hours. D. 30% saturation at 22 hours.





5. 3. Efficacy in removal of Benzotriazole (BTR) and Ibuprofen (IBU)

Removal of BTR and IBU was not so extensive as in previous micropollutants and was carried out only 3 times during the experiment. These limitations are related to the complexity and limited funds on the project. Table 6 presents mean removal efficiencies for both observed organic compounds.

Influent dose for BTR was $11.8 \pm 49.2 \mu\text{g/L}$. BTR removal efficiencies at 70% varied at ranges in between 75.14 % (woodchips) - 89.58 % (biochar 5%) across the tested FMs. The highest removal of BTR was observed at biochar 5%. The default structure had the lowest rate but at moderate 75.14%. Application of FMs increased BTR removal efficacies up to 14.44 % respectively (Tab. 6).

BTR removal efficacies at 30% saturation showed positive impact on FMs and default structure performance under lower saturation. The dominant FMs in this experimental set up was biochar 5% with 98.34 % efficacy in BTR removal. Remaining FMs had slightly lower values (limestone – 93.2 %; woodchips – 90.92 %). The default structure had the lowest efficacy in BTR removal of 87.19 %, indicating positive impact of FMs application as efficacy could be increased up to 11.15 %. Lower saturation in comparison to higher proved to boost efficacies in all tested FMs including default up to 18.37 % (Tab. 6).

Table 6. Removal efficiency of individual filter materials (FMs) in two different saturations (70%; 30%).

FMs	Saturations	BTR	IBU
	[%]		
default	70	76.71	26.05
	30	87.19	84.30
woodchips	70	75.14	51.54
	30	90.92	73.93
Limestone	70	87.80	-17.48
	30	93.20	40.15
Biochar 5%	70	89.58	81.46
	30	98.34	91.71

IBU concentration were $0.56 \pm 3.59 \mu\text{g/L}$. Regarding IBU removal at 70% saturation biochar 5% had significant efficacy (81.46 %) compared to other tested FMs including default (26.05 %). Woodchips had moderate removal efficacy of 51.54 %. Limestone is not recommended to be used against IBU removal as results stated possible support of this micropollutant in its concentrations (Tab. 6).

Removal of IBU at 30 % saturation showed superiority of biochar 5% with higher removal rate of 91.71 % among other tested FMs with boost of 10.25% compared to its performance at higher rate of saturation. The default structure showed a significant removal rate of 84.3 %. When compared to other tested FMs (woodchips 73.93 % and limestone 40.15 %) default had higher success in effective IBU removal. This evidence hints at support of IBU concentrations in effluent by limestone ones more. In summary, lower saturations showed an increase in FMs efficacies against IBU (Tab. 6).

6. Discussion

This thesis explored the efficacy of biochar 5%, woodchips, and limestone in the filtration of greywater (GW) with reference to default structure (no additives), focusing on the removal of total organic carbon (TOC), total carbon (TC), inorganic carbon (IC), total nitrogen (TN), ammonia (NH_4^+), heavy metals (Cu, Ni, Zn), semi-metal boron (B), and pharmaceuticals (BTR and IBU). Combinations of four different experimental setups were used to identify impact of different saturations and contact times to compare efficacy of selected FMs including default in removal of micropollutants of interest. The findings were juxtaposed with existing literature to identify concordances and areas of potential controversy, providing a nuanced understanding of GW treatment complexities.

6.1. Total carbon (TC), total organic carbon (TOC), inorganic carbon (IC)

Prior research by other authors did not contribute to TC concentrations occurring in greywater when treatment is discussed. The findings of this study conducted under combinations of different experimental setups stated general inefficiency of selected FMs in treatment of total carbon. It is important to note that woodchips was the only FM which could lower the TC concentrations with 41.16% efficacy. Lack of consistent irrigation (3 weeks break) during the experiment showed overall TC levels stabilization (See appendix No 1-5.). The total carbon consists of TOC and IC and when water quality for potential reuse is discussed in research chapter, prior studies highlighted the importance of compartmentalizing these criteria as they indicate specific conditions which represent greywater pollution. Total organic carbon (TOC) concentrations at high levels support microbial growth and deplete COD in bodies of water (Ledin et al., 2001).

In this experimental study TOC levels were successfully decreased particularly at woodchips (53.67%) and at biochar 5% (66.12%). The removal efficiency of biochar 5% is similar to findings of Hess et Morgenroth, 2021 where biologically activated carbon (BAC) was used. These two filtering materials share similar properties when the production is compared as both are made using biomass in pyrolysis leading into high surface area and high porosity. However, BACs properties in sorption are higher as additional process of carbon activation is usually

involved in production. Nevertheless, when efficacies of these materials are compared, they share the same range in TOC removal efficacy. Another highlight from the compared prior study suggests that longer operation leads into better removal as their study comprised over 900 days while this experiment lasted up to 6 months only (Hess et Morgenroth, 2021). The data on TOC concentrations in this experiment supports the finding as general decreasing trend in TOC concentrations were observed as well (Fig. 8).

IC concentrations did not decrease after application of tested FMs. Adequate literature describing IC removal processes in context of greywater treatment was not found. This experimental study found that all tested FMs did not prove to be sufficient in removal of IC as its concentrations did not reduce below the initial influent dose. However, overall IC concentrations tended to decrease by the end of the experiment. This overview suggests that temperature decrease by the end of the experiment may have lead to this outcome but do not prove it as lengths of the experiment did not exceed 6 month (June – October 2021) (See Appendix No. 11-14).

6.2. Total nitrogen (TN) and ammonia ion (NH_4^+)

This study showed that all tested FMs could remove TN concentrations in high ranges 71.04 % (woodchips) – 77.48 % (biochar 5%). Compared to studies by Clough et al (2013) which stated biochar's mean efficiency at 60%, this study showed higher performance of this filter material. When saturation was reduced to 30%, biochar 5% maintained the highest removal efficiency, which increased to 83.76%. Biochar 5% comparison with prior studies highlight nitrogen immobilization and mineralization processes to soil biochar combinations used (Clough et al., 2013). Both woodchips and limestone also showed improved removal efficiencies at lower saturation levels, with efficiencies close to each other (80.6% and 80.99%, respectively). This suggests that lower saturation levels may enhance the TN removal efficiency of certain filter materials. The tested filter materials

showed an improvement in TN removal efficiency by 5.89% to 9% compared to the control setup. This indicates that while the use of additives like biochar, woodchips, and limestone does lead to an improvement in TN removal, the increase in efficiency is relatively slight. A general decreasing trend in TN concentrations was observed across all tested materials as saturation levels were reduced from 70% to 30%. This might indicate TN stabilization by the end of the experiment, suggesting that the filter materials not only remove TN but may also contribute to its stabilization in the water (Fig. 10; Fig. 16).

Regarding ammonia ion (NH_4^+) removal prior studies proved efficacy of vertical constructed wetlands (CWs) with rework in structure with application of oxic/anoxic (ME-(O/A)CW) processes to boost removal effectiveness of such CWs with rate of 85.3%. The efficiency at these rates connected to the process of nitrification which was enhanced by aeration in the upper layer of CW. Aeration into top layers of the CW was a significant condition for feasible removal of ammonia (Deng et al., 2020). In comparison, this experiment applied physical models which did not have aeration. This difference potentially could dismiss the efficacy of tested FMs. A remarkable point in the discussion of ammonia ions concentration trend was the impact of 3 weeks break. All the experimental setups showed enormous increase in NH_4^+ concentrations before irrigations stopped for this break. This suggests NH_4^+ accumulation in all tested groups under constant influent dosing. 3 weeks of break stabilized the NH_4^+ levels and could even decrease its values below the influent dose. However, it was not sufficient to consider selected filter materials removal efficacy in NH_4^+ as a success (See appendix No. 20 – 24).

6.3. Metals removal (B, Cu, Ni, Zn)

This experiment in boron (B) removal was not successful as tested filter materials did not prove to be efficient in given experimental setups. Moreover, these FMs showed a sign of B support in all experimental setups as influent dose and effluent concentrations of B consistently correlated across the duration of the experiment. Lower concentrations of B at the start of the experiment and at 3-week

break support this hypothesis as active irrigation with B content continued to support B concentrations at influent dose levels on the contrary (See appendix No. 25 – 29). This finding could be potentially useful for maintaining B presence in soil where B is deficient. However, concentrations of B have to be controlled as excessive levels lead into plant toxicity and changes to hydrophobic properties in soil (Wiel-Shafron et al., 2006)

Regarding remaining heavy metals (HMs) their efficacies could be overviewed as a set as their overall trend of concentration across the experiment along with FMs efficiency in successful removal of Cu, Ni, and Zn were similar. Removal efficiencies ranged from 99.02% (woodchips) to (99.25 %) in Cu concentrations from the start to the end of the experiment. However, default structure with no additives had similar top efficiency, indicating no need for application of any FMs for successful removal of Cu. Similar findings were presented in results of Ni and Zn removal (Fig. 14-23). Prior study by Abbasi et al. (2018) provided data on the same set of HMs but with different approaches in removal of Ni, Zn, and Cu. The goal of the research was to test short hydraulic retention time and invasive plant (*Alternanthera ficoidea*) as a biofilter bed to determine its value under this setup. When efficiencies compared in two different experiments, significant improvement in removal efficacies was observed: increase in removal of copper up to 54.3 %, nickel 74.63 %, manganese 60.93 %, and zinc 55.93 % respectively (Abbasi et al., 2018; Fig. 14-23).

6.4. Pharmaceuticals removal (IBU, BTR)

The consistently high removal efficiencies of BTR by biochar 5% (89.58 % - 98.34 %) across different saturations highlight its effectiveness as a filtering material for this specific micropollutant. This could be attributed to its high surface area and porosity, which are beneficial for adsorption processes. The significant increase in removal efficiencies at lower saturation levels suggests that operational conditions, such as water content in the system, play a critical role in optimizing the performance of FMs for micropollutant removal (Tab. 6). Influent dose for BTR was 11.8 ± 49.13 $\mu\text{g/L}$ which was substantially higher than the influent dose 10.15 ± 26.1 $\mu\text{g/L}$.

Limited data from this experiment suggests that biochar 5% in this physical model was significantly dominant when compared to MBR treatment with removal rate of only 55% (Reemtsma et al., 2010).

Prior research on IBU removal with PAC coconut-based pellets, lignite-based pellets and blend of these two in form of pellets resulted in high removal (up to 87%) in coconut-based pellets. However, this study involved shorter contact time with synthetic urine and deionized water with content of IBU for up to 30 minutes (Finn et al., 2023). Given the differences of this experimental setup in comparison to the results of the experiment conducted at CZU, hardly any data can be cross-referenced with precision in statements. Nevertheless, insight of both studies discuss the matter of IBU removal importance as it is an endocrine disruptor at lower trophic levels, and what is more important they discuss methods in removal of IBU with different filter materials (Gonzalez-Rey et al., 2011).

7. Conclusion

This study aimed to assess the effectiveness of filter materials – such as limestone, woodchips, biochar 5% and a standard structure – in eliminating specific micropollutants from greywater (GW). The research evaluated the reduction of organic carbon (TOC), total carbon (TC), inorganic carbon (IC), total nitrogen (TN), ammonium ions (NH_4^+), boron (B), copper (Cu), nickel (Ni), zinc (Zn), benzotriazole (BTR) and ibuprofen (IBU) at different saturation levels and contact time.

The findings revealed that biochar 5% proved to be the most efficient filter material in removing TOC, TN, Ni, IBU, BTR across all test scenarios. Limestone showed varying effectiveness performing better at a 30% saturation level and with 22

hours of contact time for TOC, TN, Cu, Ni, BTR and was the most dominant at Zn removal. However, limestone should not be used for IBU removal. Woodchips was the only FM which could remove TC to some extent compared to limestone and biochar 5%. Regarding the remaining TC, IC, NH₄⁺ and B, selected filter materials are not recommended to be used for removal purposes of these selected micropollutants.

These findings underscore the significance of choosing the right filter materials and tuning contact durations and saturation levels for efficient greywater treatment. They offer guidance for creating greener and more effective greywater treatment setups.

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8.1. Scientific Publications

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