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Life cycle performance and associated environmental risks of constructed wetlands used for micropollutant removal from municipal wastewater effluent

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A B S T R A C T

Wastewater treatment systems produce environmental impacts in their construction and operation, and nature-based treatment processes offer opportunities to reduce the environmental burdens. Constructed wetlands represent such a solution that can remove micropollutants from municipal effluent. This study evaluates life cycle impacts and environmental risk of constructed wetlands for improved treatment performance. The assessment of laboratory- and pilot-scale installation performance provides insights into sustainability of scaling fundamental research to technology demonstration. The normalised life cycle assessment showed that the laboratory installation generated higher environmental impacts than the pilot, due to the cooling tank and its associated electric power (~60% of the total burdens for five impact categories). The avoided environmental impacts through the micropollutants' elimination ranged from 50% to 99.9% (for freshwater ecotoxicity and human toxicity, respectively). A sensitivity and uncertainty analysis highlighted how the substrate and electricity demands represented the highest environmental impacts, thus extending lifespan of a full-scale system whilst maintaining treatment performance represents the most notable opportunity to improve the environmental performance. The findings support measures to enhance sustainability through design, procurement and operation stages of development. Constructed wetlands represent a sustainable nature-based form of wastewater treatment, and this study offers lessons to further enhance their environmental performance.

1. Introduction

The proper treatment of water is central to protecting the natural environment, besides other domains, increasing global population leads to higher production of sewage. Wastewater treatment faces a range of challenges, from large capital costs to introduce new treatment technologies that reduce environmental pollution, as well as possibly increasing operational costs and thus producing more greenhouse gas (GHG) emissions (Siatou et al., 2020). Considering that 3–4% of global energy consumption is associated with wastewater treatment, solutions must be innovative in terms of both their performance and sustainability credentials (International Energy Agency, 2018). In doing so, they will address the UN's Sustainability Development Goal #6 to '*Ensure availability and sustainable management of water and sanitation for all*' and deliver improvements in the percentage of wastewater treatment globally (International Energy Agency, 2018). Taking this into consideration, more sustainable wastewater treatment plant (WWTP) solutions are needed (Gallagher and Gill, 2021).

To deliver a sustainable water sector, energy efficient processes and embracing circular economy solutions are essential in the future of wastewater treatment (Galychyn et al., 2022; Okonkwo et al., 2023; Stefanakis et al., 2021). Passive treatment (i.e. processes that do not require human intervention or energy input while operating) systems offer an opportunity to directly reduce the operational energy demands of WWTPs (Martínez-Hernández et al., 2020). However, such treatment systems are notably larger in size, and require regular maintenance. Alternatively, recovering resources from wastewater (e.g., adsorbents from cellulose materials present in wastewater or production of biodiesel from lipids from sewage) can support a circular economy whilst indirectly having the potential to reduce the energy requirements associated with wastewater treatment. These measures have the potential to reduce the environmental impacts of WWTPs and address a

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reuse and recovery goal in the forthcoming revision of the Urban Wastewater Treatment Directive (European Commission (EC) 2022). In addition, this enhances water quality standards, which majority of the EU member states are failing to achieve (European Environment Agency, 2018).

One such reason that could be attributed to deteriorated water quality is the inability of the conventional mechanical-biological wastewater treatment processes to fully remove the emerging micropollutants (MPs) (Kim and Zoh, 2016). The negative impacts of MPs in water bodies on aquatic organisms is due to their high persistency, bioaccumulation and ecotoxicity (Szymańska et al., 2019). This creates a sector under pressure to meet water quality standards that are becoming more and more stringent (Link et al., 2016). As such, preserving a good ecological status in water surface bodies in the EU and in UN member states requires advanced treatment (EC, 2018). Ozonation and the adsorption of MPs on activated carbon (AC) represent the two most common technologies applied for treatment of municipal wastewater effluents designed for eliminations of MPs (Margot et al., 2013). Both methods have high installation and operational costs, and are typically only fitted in large WWTPs. Ozonation consumes up to 4 times more energy than MP removal through adsorption on AC, thus making the latter method a more sustainable treatment process regarding the energy consumption (Mousel et al., 2017). Connecting the two challenges together – sustainable wastewater treatment and preventing entrance of MPs into water bodies, constructed wetlands (CWs) were tested as a type of nature-based solution (NBS) in order to remove MPs from the municipal effluent (Matamoros et al., 2016).

NBS are presented as a passive treatment solution that also promotes ecosystem services in urban settlements. Such NBS are engineered systems, which mimic existing natural ecosystems and are designed in a way to operate in lowest possible dependence on mechanical parts (International Water Association Publishing, 2021). There are several types of NBS, e.g. different types of ponds (aerated, anaerobic, facultative), or the previously mentioned constructed wetlands. Taking CW as an example, they have been applied as a treatment process for various types of wastewater (domestic, urban run-offs, agriculture run-offs, stormwater, slaughterhouse run-offs, etc (Kadlec and Wallace, 2008)).

Appraising the sustainability of WWTPs can provide valuable insights into improved design and optimisation of these technologies and systems (Chen et al., 2020). Life cycle assessment (LCA) provides a means of quantifying the environmental performance of any treatment technology or an entire WWTP. LCAs have been applied in multiple studies to suggest general improvements to WWTPs' treatment efficiency (Corominas et al., 2020) or to evaluate e.g. the energy efficiency of WWTPs, the impact of the WWTPs' size and performance on the generated environmental factors or to compare different design solutions (Allami et al., 2023; Pasciucco et al., 2023; Tsangas et al., 2023). Purely focused on MPs' removal, the impacts of advanced wastewater treatment technologies were evaluated with freshwater ecotoxicity (EF) and human toxicity (HT) representing the commonly appraised impact categories (Li et al., 2019; Pesqueira et al., 2021; Postacchini et al., 2018). High EF and HT burdens were observed in LCA studies accounting for systems treating effluent with compounds reflecting personal care products as organic MPs in WWTPs (Li et al., 2019). Other investigations helped to identify the preferred components for advanced treatment technologies to reduce the environmental impacts and attain optimal operational standards (Igos et al., 2021). For example, Igos et al. (2021) found that using micro-grain (μGAC) was favourable to using a powder AC (PAC) in a fluidized bed process. A recent study by Pistocchi et al. (2022) evaluated the toxicity of 1337 compounds in European waters, yet for only 60 personal care products characterisation factors (CFs) exist (Li et al., 2019). Further characterisation and quantification of emitted MPs into water bodies are required to improve the identification of these compounds and to assess their environmental burdens. Several LCA studies stress the importance of circularity measures in CWs, e.g. usage of recycled materials or resource recovery (de Simone Souza et al., 2023; Hube et al., 2023). Limited applications of CW for targeted elimination of MPs have been undertaken to date (Venditti et al., 2022a,b), with no research tackling an environmental evaluation of this technology as part of the investigation.

As MPs can induce toxic effects even though present at very low concentrations (ng/L or μg/L) (Gildemeister et al., 2023; Vymazal et al., 2017), environmental risk assessment (ERA) is included in this study to assess the potential risk of the studied chemicals for the environment. The ERA is a method used for monitoring of toxic compounds e.g., recently after COVID infection, in coastal areas, or for assessment of antibiotic resistance (Löffler et al., 2023; Lopez-Herguedas et al., 2023; Tovar-Salvador et al., 2023). The method is characterized by so-called risk quotients (RQ) of the targeted compounds (the MPs within the CWs), the knowledge about MPs' removal is then completed by investigation of their avoided impacts. As such, consideration of the ERA in parallel with LCA can offer a more robust form of evaluating passive CW systems in the treatment of wastewater.

This study aimed to undertake LCA and ERA of the CW systems to improve our understanding of the burdens of passive wastewater treatment systems. This was achieved by comparing the environmental impacts of conventional and innovative CW substrates, evaluating the burdens of laboratory and pilot-scale installations and assessing environmental risks linked to elimination of the MPs. A further sensitivity and uncertainty analysis support a set of lessons learned from the investigation. The study uniquely presents and justifies the value of CWs as a sustainable form of treatment, considering both the embodied and operational burdens of these systems.

2. Methods

2.1. Goal and scope

This work aims to quantify and compare the environmental risks and impacts of CW systems, a passive NBS wastewater treatment technology, by applying LCA and ERA methodologies. Specifically, different adsorbents and system configurations are evaluated for this NBS over a 1-year operational period, considering its ability to maintain MP removal. To effectively remove four MP compounds (atenolol, ciprofloxacin, diclofenac, carbamazepine), that were chosen based on their relevance and availability of their CFs, novel substrate with a 15% admixture of biochar was required as it has demonstrated its ability to remove MPs (Brunhoferova et al., 2022).

This analysis evaluated both laboratory- and pilot-scale installations, as they represent upscaling of fundamental research to technology demonstration of this NBS. These findings will help to evaluate if a scaling effect exists in CW systems used for MP removal, and the associated environmental sustainability of these systems can be improved, from experimentation to full-scale deployment. Quantifying the avoided impacts of the eliminated MPs and comparing this with the production of the biochar itself will provide additional insights into the preference of passive treatment systems compared to conventional WWTPs.

The attributional LCA methodology applied to quantification of the environmental impacts of CW for MP removal in WWTP is in accordance with ISO 14044 and ISO 14040 guidelines (ISO, 2006b; 2006a). The functional unit (FU) applied in this study to evaluate the substrates, and system performance of the CWs at different scales was selected as a hydraulic loading rate (HLR) of 150 L of treated wastewater per square metre per day ($l/m^2/d$), or 1 population equivalent (p.e.). The FU was chosen as it provides a fair metric for comparing the system's performance and associated environmental impacts with other forms of wastewater processes. The life cycle impact assessment will help evaluate the individual component and cumulative CW treatment system burdens.

The avoided environmental impacts and the ERA of the eliminated MPs will also amplify the value of the CWs to remove these compounds from municipal effluents and prevent their entry into surface water bodies.

A sensitivity and uncertainty analysis will be applied to ensure that the environmental performance of these systems are not affected by minor potential differences in system boundaries or operational factors.

2.1.1. Case study

2.1.1.1. Substrate. A comparison of conventional 100% sand substrate with novel homogeneous admixture (15% activated biochar (AB) with 85% sand) in the substrate used for the CWs was undertaken to assess the impact of improved process performance. The substrates were chosen based on their economic viability and lower environmental footprint as they were locally produced (Brunhoferova et al., 2022).

2.1.1.2. Laboratory scale experiment. The laboratory-scale CW installation (Fig. 1) is presented as two parallel plexiglass column units (inner diameter 29 cm, height 115 cm) filled with only sand or including a 15% AB admixture, and a bottom 10 cm layered fine/coarse (2–8/4-8 mm) gravel for drainage.

These vertical-flow laboratory CW systems (lysimeters) used a synthetic wastewater with a MP spike to imitate WWTP effluent percolated through the columns from the top to the bottom. The operating flow was intermittent, with three 30-min cycles per day, and maintaining an average hydraulic loading rate (HLR) of 100 L per square meter of surface area per day (l/m^2/d). An estimated 0.2 kWh of electrical energy was used daily for pumping. The units were planted with commonly used macrophytes *Phragmites australis* and *Iris pseudacorus*. The laboratory installation also required a cooling tank to maintain the wastewater at ca 4 °C. UV lamps provided adequate lighting (8 h per day).

2.1.1.3. Pilot-scale installation. The pilot CW installation (Fig. 2) was placed at the Reisdorf-Wallendorf WWTP in Germany which had a 4,600 p.e. capacity. It discharges effluent into river Sûre, which creates a geographical border between Luxembourg and Germany. The 1 m^3 vertical flow pilot acted as a reference size, using an intermediate bulk container (IBC) tank and had an intermittent HLR flow averaging 215 l/m^2/d (varying HLR between 100 and 300 l/m^2/d). The system required 0.28 kWh of electrical energy daily for pumping. The homogeneous sand and 15% AB admixture sat on top of a bottom gravel layer was created using a mix of coarse (8–16 mm) and fine (2–8 mm) layers. Two types of emergent macrophytes, *Phragmites australis* and *Iris pseudacorus*, were placed at the top of the CW, with woodchip used as an insulation layer and cover.

2.1.2. Micropollutant treatment conditions

The influent into both installations (real or synthetic) reflected typical wastewater effluent characteristics to allow for the removal the mixture of MPs with specific prominence in Luxembourg and Germany. Firstly, atenolol is a highly prescribed pharmaceutical (beta-blockers) in

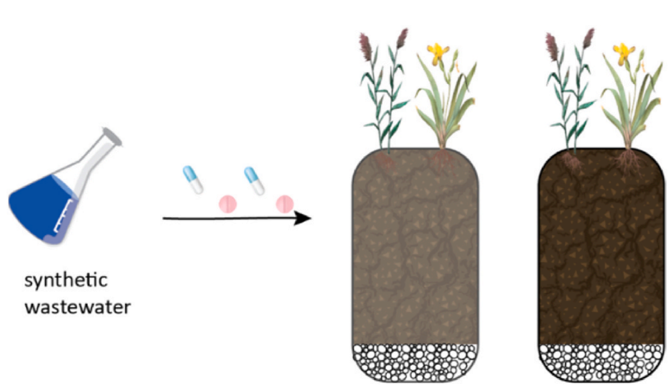

Fig. 1. Schematic of the laboratory constructed wetland installation.

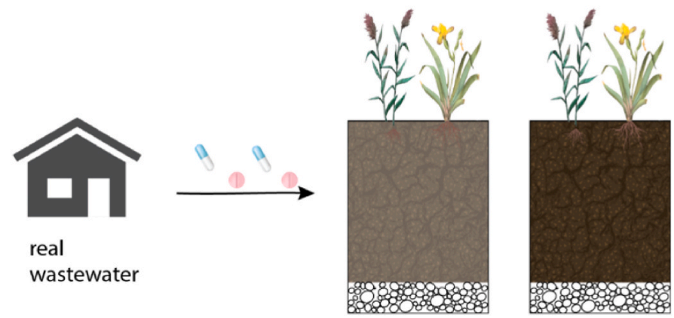

Fig. 2. Illustration of the pilot constructed wetland installation.

Luxembourg (Venditti et al., 2022a,b). Ciprofloxacin is an antibiotic present in a European Watch List (EC, 2018). Diclofenac is a compound for the control of the efficiency of the quaternary wastewater treatment in Switzerland and Germany (DIV_Recommendation, 2019). Lastly, carbamazepine is classified as a control for surface water bodies in Luxembourg since 2016. Three of the four targeted compounds, not atenolol, are commonly found in European waters in concentrations higher than ecotoxicity exceedances which effects including change in growth, reproduction and survival (Fekadu et al., 2019).

2.1.3. Boundary conditions

The system boundaries (Fig. 3) focus on substrate production, the embodied components of the system, and the operational energy demands for selected 1-year period as a lifespan for the baseline scenario for both scales. The geographical location considered was Luxembourg, as it presents a suitable location for full-scale deployment as an advanced treatment technology in small WWTPs. A cradle-to-grave boundary condition was defined for this system, which ensured that the embodied and operational life cycle of each treatment system was considered. However due to the uncertainties and complexities of the end-of-life stage of these systems, the management of waste and any potential recovery of resources was omitted from consideration in this assessment.

From Fig. 3, the foreground data is reflected by all components used in the laboratory- and pilot-scale CWs, with background data extracted from Ecoinvent for the materials and energy, to allow for the environmental burdens to be quantified for these systems.

2.2. Life cycle inventory

The life cycle inventory for the CW substrates (Table 1) and installation components (Table 2) was collated from purchase orders and online data sources. The inventory data for the gravel were obtained from gravel quarry in Switzerland, for the sand from bentonite the data were obtained from bentonite quarry operation in Germany. For biochar, data for wood charcoal was sourced from the Ecoinvent database (Ecoinvent, 2023), which only differs from biochar in terms of its application (Tenenbaum, 2013). All background data extracted from Ecoinvent applied the cut-off model as the end-of-life was omitted from the assessment, and thus waste or recovery pathways were not evaluated within the scope of this study.

The laboratory- and pilot-scale installations used similar pumps and controllers for dosing (with a 10-year lifespan), and both used high density polyethylene (HDPE) sampling tanks with 20-year lifespan. The laboratory installation also required a cooling tank from Lely Center, Greece (for tempering the inlet mixture to avoid biofilm formation in the influent), Marprene process and Tygon E−3603 tubing was used with corresponding connections, a polymethyl metacrylate (PMMA) plexiglass column as the storage tank all with 20-year lifespans. The laboratory installation also used a Megaman LED UV lamp with a 5-year lifespan. The pilot-scale installation required a HDPE storage IBC tank

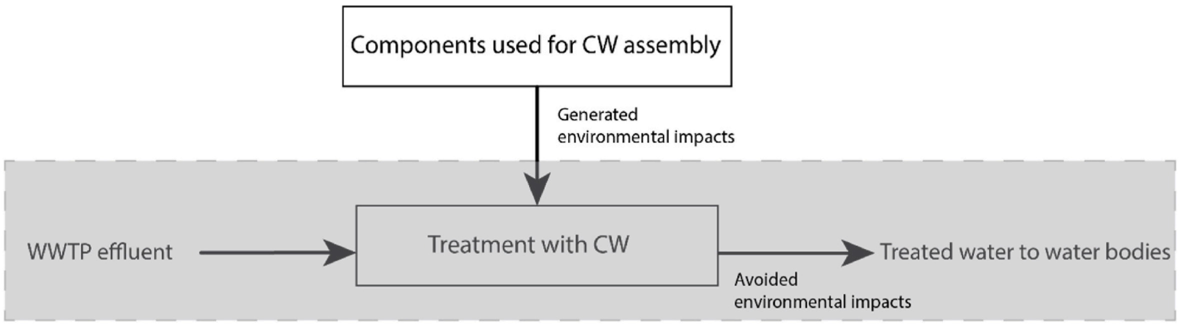

Fig. 3. Boundaries of the CW (constructed wetland) system (component details provided in Table 1 and Table 2).

Table 1
Substrate material quantities used in the laboratory- and pilot-scale CW installations (15% AB stands for 15% activated biochar).

Substrates	Material quantity per unit (kg)	
	Laboratory	Pilot
Gravel	11	433
100% sand	139	2,340
85% sand	132	2,227
15% AB	23	393

Table 2
Different component weights and energy demands of the laboratory- and pilot-scale CW installations.

Component	Weights		Energy	
	Laboratory (kg)	Pilot (kg)	Laboratory (kWh/yr)	Pilot (kWh/yr)
Pump	9.0	8.3	73.91[a]	102.2[d]
Controller	0.2	0.7		
Tubing	1.1	3.0		
Sampling tank	1.0	1.7		
Storage tank	13.2	65.0		
LED Lights	0.215	–	1.46[b]	
Cooling tank	1,013.0	–	5,694[c]	

[a] Operational annual energy demand for laboratory pump, averaging 1.5 h per day.
[b] Operational annual energy demand for 8 h of LED lighting for 365 days per year.
[c] Operational annual energy demand for cooling tank, 24 h per day, 365 days per year.
[d] Operational annual energy demand for laboratory pump, 21 min per day, 365 days per year.

and elastomer tubing with 20-year lifespans. The inventory data for the components of the studied installations and the substrates were obtained from the Ecoinvent v3.9 database. The grid electricity mix in Luxembourg consists of a mix of coal and renewable sources, with this mix being maintained in future year as a worst-case scenario for related environmental impacts (Luxembourg 2020 – Analysis - IEA). Direct transport related burdens of the substrate and components were omitted from this assessment as it was considered negligible in the scale of these CW systems. It was assumed that no unforeseen maintenance requirements were required during the project lifespan. Lastly, the inclusion of gaseous emissions from the wastewater were omitted from the assessment.

2.3. Impact categories and environmental assessment

Seven midpoint impact criteria were chosen including: freshwater ecotoxicity (EF); climate change (CC); land use (LU); human toxicity (carcinogenic (HT-C) and non-carcinogenic (HT-N/C)); water use (WU); and non-renewable energy resources (ER). These indicators were selected based on their appropriateness to the systems being evaluated and the impacts of concern in wastewater treatment. Thus, for stakeholders in the wastewater sector, a clear link between treatment outcomes with specific impacts of the system was deemed more valuable as outputs from the study.

These categories are chosen based on similar indicators used in related literature (e.g. Igos et al., 2021 which considered EF, CC, HT-C & HT-N/C as it focused on MPs and their impacts), and the authors' evaluation of other impact categories and their relevance. Therefore, ER addressed the energy demands of these systems, and LU and WU accounted for the need for natural materials and its relationship to water for the other indicators. The environmental burdens associated with each of these impact categories were examined for all of the studied components and substrates. CW is an extensive NBS technological solution, challenging in terms of area, yet are less demanding in terms of financial, energy and chemical requirements due to acting as a passive treatment process.

The impact assessment for EF v3.0 was compared in terms of ecosystem impacts (comparative toxic units or CTUe), and the CC potential (kilograms of CO_2 equivalents or kg CO_2 eq.). For evaluation of impact assessment for LU was used soil quality index, which has no dimension. The impact assessment of HT is represented by comparative toxic units for human (CTUh). The WU impact assessment reflects the deprivation-weighted water consumption in the world (m^3 world eq. dep.). Finally, ER was represented as MJ (net calorific value), reflecting the abiotic depletion potential of fossil fuels.

For the selected MPs is evaluated freshwater ecotoxicity and both carcinogenic and non-carcinogenic human toxicity given the accessibility of the CFs, which were obtained from Igos et al. (2021). Furthermore, ERA is evaluated based on comparison of worst-case predicted no effect concentrations (PNEC) and the predicted environmental concentrations (PEC) or measured environmental concentrations (MEC), respectively. PNEC values are generally estimated from standard toxicity tests and the values used are taken from Norman database (Norman Network) and they represent the lowest values obtained in freshwater (conservative approach). As such, values are aligned with those the Swiss competence centre for applied, practice-oriented ecotoxicology (Ecotox Centre, 2023). In this case, the RQ is calculated from MEC/PNEC and based on its value following risk quotient can be assessed: very high risk (RQ $>$ 1), high risk (RQ $=$ 1), medium risk (0.1 $<$ RQ $<$ 1.0), low risk (0.01 $<$ RQ $<$ 0.1), negligible risk (RQ $<$ 0.01).

Lastly, global normalisation factors prepared by Crenna et al. (2017) are applied to provide a balanced global scale to the environmental impacts, consider a per person or p.e. output to compare both systems in terms of environmental performance.

2.4. Sensitivity & uncertainty analysis

Some aspects of the CW installations have associated sensitivities or uncertainties associated with them. The sensitivity and uncertainty analysis evaluates the impact of several factors on overall results (Table 3) and includes (i) evaluating different hydraulic rates into the pilot-scale installation, (ii) limiting boundary conditions to exclude the cooling tank in the laboratory-scale installation, (iii) accounting for the biomass produced from the plants growing in the wetland as an offset measure, and (iii) quantifying the impact of different component life cycles on the life cycle performance of the pilot CW system.

3. Results & discussion

3.1. Impact of the substrates used in a constructed wetland

Initially focusing on the substrates, which govern MP removal efficiency in the wetlands, the EF impact category was prioritised to compare the production value of 1 kg of AC and 1 kg of biochar as admixtures in the substrate are compared (Fig. 4).

The initial freshwater ecotoxicity (EF) impacts for AC were approximately 30 times higher (265.1 CTUe) as compared to biochar (8.6 CTUe), suggesting the environmental benefits of biochar. Table 4 provides the complete set of environmental burdens and provides a comparison between the substrates in terms of all impact categories.

The high EF burden for AC was similarly high for ER, with a burden over 39 times greater (109.1 MJ) than for biochar (2.8 MJ). CC and HT-C impacts of producing 1 kg of AC were 4.5 and 6.0 times higher (7.9 kg CO_2 eq. and $1.9{\cdot}10^{-9}$ CTUh, respectively) than its equivalent quantity of biochar (1.8 kg CO_2 eq. and $3.2{\cdot}10^{-10}$ CTUh respectively). The higher CC and ER impacts are associated with the energy demands of AC production through process steps requiring high temperature thermal decomposition and activation.

In contrast, the LU demands to produce 1 kg of the substrate were circa 12 times higher for the biochar (158.4 versus 13.8) because the biochar is produced from lignocellulose coming from wood, and naturally occupies more space than coal. This is reflected by the carbon content of hard coal (86–97%) as opposed to lignocellulose (44–65%) (EIA, 2022). HT-N/C and WU both favoured the production of AC over biochar to minimise environmental burdens, with increased impact for HT-N/C associated with its composition, containing toxic polycyclic aromatic hydrocarbons (PAHs) (Kuśmierz and Oleszczuk, 2014). The demands on WU for production of biochar were approx. 1.5 times higher than for production of AC, due to greater water demands of trees as wood is used for biochar production.

In summary, the results suggest that the AC has on average 11.66 times larger environmental impacts than the biochar. Four of the seven impact categories presented higher burdens for AC, ranging from as significant as ER being 39.1 times higher to CC being 4.5 greater than associated impacts from biochar. The remaining three impact categories presenting values for AC that ranged from between 9% (LU) and 73% (WU) of its equivalent quantify of biochar. The results demonstrate that the selection of a substrate for a CW can influence the overall life cycle performance, and findings depend on the impact categories considered in the investigation.

Table 3
Sensitivity and uncertainty analysis scenarios for the pilot-scale CW installation.

Scenario	Justification
Adjusting Hydraulic loading rate (HLR)	A higher HLR of 300 $l/m^2/d$ s was applied in the pilot-scale installation to evaluate the environmental impacts of the system whilst maintaining system performance.
System boundary excluding cooling tank	The cooling tank is an external component of laboratory-scale installation and could be considered outside the system boundary.
Offset from biomass production	Examining the potential offset impacts of producing 5 kg of macrophyte as biomass in the constructed wetland over a 1-year period.
Extending system lifespans	5-, 10- and 20-year system lifespans were compared to assess system performance and hotspots in the pilot-scale installation over time.

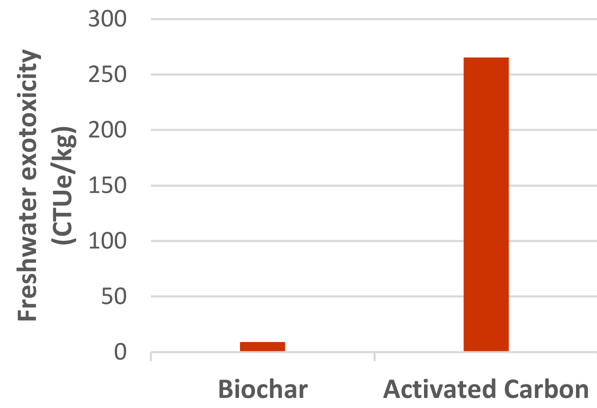

Fig. 4. Comparison of freshwater ecotoxicity generated from 1 kg of biochar and 1 kg of activated carbon.

3.2. Laboratory and pilot scale CW installations

3.2.1. Cumulative environmental burdens

An initial assessment of the cumulative burdens was adopted to evaluate the differences that may exist between the laboratory- and pilot-scale CW installations, to guide further assessment of components representing hotspots within either installation. This comparison was undertaken by normalising the calculated burdens for both installations with respect to the FU (150 $l/d/m^2$). A summary of the normalised cumulative environmental burdens of both installations are presented in Fig. 5.

This provides an overview of the comparative performance of the systems for the range of impact categories evaluated. In both cases, the 15% activated biochar admixture with sand substrate was accounted for in both installations.

Five of the seven impact categories observed greater normalised values for the laboratory installation, with differences ranging from 6.7 times higher for HT-C ($2.05{\cdot}10^{-6}$versus $3.06{\cdot}10^{-7}$ CTUh/p.e.) to a marginal 1.08 times larger WU burden (346.3 versus 319.3 m^3 world eq./p.e.) for the laboratory installation. The two impact categories presenting higher burdens attributed to the pilot installation were HT-N/C and LU, which were 3.1 ($6.32{\cdot}10^{-5}$ versus $2.06{\cdot}10^{-5}$ CTUh/p.e.) and 5.0 (45,702 versus 9,134.0/p.e.) times greater than the burdens for the laboratory installation.

3.2.2. Contribution analysis

A breakdown of quantitative values and percentage contributions of the environmental burdens attributed to all components within in the laboratory and pilot-scale installations are presented in Fig. 6 and Table 5. Dominant contributions were evident across the seven impact categories for the laboratory and pilot installations. For the laboratory scale installation, the cooling tank itself and associated electricity consumption represented over 60% of the total burdens for five impact categories, with the substrate representing over 66% for five impact categories in the pilot installation. Components such as tubing, controller and sampling tanks represent only negligible environmental burdens for both installations.

When looking at Table 5, it is evident that the electricity demands, representing the sole operational impacts of the laboratory installation, produced an average of 36.1% of the total environmental burdens across all seven impact categories, ranging from 7.6% ($1.56{\cdot}10^{-7}$ CTUh) for HT-C to over 66% for ER (11,835 MJ). For the cooling tank, with the exception of LU (6.8% or 622), the inverse contribution to the total

Table 4
Environmental impacts of 1 kg of activated carbon or biochar, and the magnitude of difference between the environmental burdens of both substrates (freshwater ecotoxicity (EF); climate change (CC); land use (LU); human toxicity (carcinogenic (HT-C) and non-carcinogenic (HT-N/C)); water use (WU); and non-renewable energy resources (ER)).

Impact category	EF	CC	LU	HT-C	HT-N/C	WU	ER
	CTUe	kg CO_2 eq.	–	CTUh	CTUh	m^3 w x eq. dep.	MJ
Biochar	8.7	1.8	158.4	$3.2 \cdot 10^{-10}$	$2.2 \cdot 10^{-7}$	0.9	2.8
Activated carbon	265.1	7.9	13.8	$1.9 \cdot 10^{-9}$	$1.0 \cdot 10^{-7}$	0.6	109.1
Difference	30.7	4.5	0.1	6.0	0.5	0.7	39.1

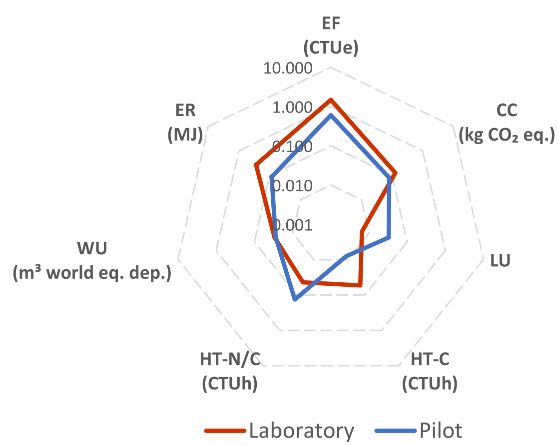

Fig. 5. Global normalised burdens (1 p.e.) for laboratory and pilot CW installations (freshwater ecotoxicity (EF); climate change (CC); land use (LU); human toxicity (carcinogenic (HT-C) and non-carcinogenic (HT-N/C)); water use (WU); and non-renewable energy resources (ER)).

environmental impacts were observed for HT-C and ER, representing over 81% ($1.67 \cdot 10^{-5}$ CTUh) and less than 12% (2,108 MJ) respectively. This significant HT-C burdens was attributed to the large chromium steel tank.

For the pilot-scale installation, the substrate represented an average of 71% (range 27.3% for ER to 99.4% for LU) of the total burdens across all seven impact categories, as compared to an average of 17.5% (0.8% for HT-C to 63.5% for LU) for the laboratory installation. The high burden associated with the substrates was linked to LU in both installations. High HT-NC burdens of 37.4% ($7.7 \cdot 10^{-6}$ CTUh) and 95.4% ($6 \cdot 10^{-5}$ CTUh) for the laboratory and pilot installations may be caused by the toxic compounds of biochar in combination with the presence with the sand.

Electricity for the cooling tank in the laboratory installation represented 98.6% of the total energy demands for this installation, as the LED lights and pump dosing reflected negligible contributions. The maximum impact related to electricity demands for the pilot installation equalled a 7.6% for any impact category (ER of 423 MJ).

The environmental burdens attributed to the pump, which contained heavy metals and plastics, and the HDPE storage tanks, represented the remaining components of both installations that presented notable burdens for some impact categories. However, it should be noted the pump was oversized in the laboratory installation. This allowed the equipment to be used in other experimental projects, and ensured it had the capacity for larger projects. It reflects an important legacy of short-term experimental research, as to ensure that equipment life cycles go beyond the project. The impacts of the plastics used in manufacturing the storage tanks led to 13–14% of the CC burdens (90–152 kg CO_2 eq.) in both installations, with ER impacts much larger for the pilot installation (58.2% or 3,294.7 kg CO_2 eq.) than the laboratory installation (13.4% or 2,392 kg CO_2 eq.).

The pilot installation demonstrated its ability to be a wastewater treatment system with an improved environmental performance, as advancing the technology readiness level (TRL) of CW treatment can reduce infrastructural and energy demands with increasing the scale of the system. To further improve environmental performance of the pilot installation, delivering a passive means of distributing effluent throughout the CW can remove the need for embodied (pump) and operational (electricity) burdens. For the experimental laboratory installation, the selection of components and materials that have lower environmental impacts e.g. PMMA over plexiglass columns, can provide the same system requirements.

Resende et al. (2019) indicated that operation phase was in case of pilot CWs (vertical vs. horizontal flow, decentralized domestic wastewater treatment) responsible for more than 90% of the total impact for CC. This reflects the results of the laboratory scale environmental evaluation, where the unit operation (represented by electricity for pumping, LED lights lightning and cooling tank operation) represented majority of the generated burden for CC (583 kg CO_2 eq. out of 1071 kg CO_2 eq. in total). Pasciucco et al. (2023) mentioned the positive correlation between the increasing size of the WWTP and better treatment performance in terms of environmental impacts. However, due to the lack of studies of similar character (lab and pilot scale CWs for post-treatment) it is intricate to set proper result comparison.

3.3. Micropollutant removal and environmental assessment

The avoided environmental impacts are assessed for four MPs: atenolol, carbamazepine, ciprofloxacin and diclofenac. The CFs used in the assessment are presented in Table 6 based on Igos et al. (2021). Ciprofloxacin has the highest CF for ecotoxicity of freshwater from the studied compounds and as a relatively cheap antibiotics it is thus largely produced in Europe, but also in Africa (UNICEF, 2002). Diclofenac, which has second highest CF, belongs to one of the most excreted drugs into surface waters in the world (Sathishkumar et al., 2020). Carbamazepine and diclofenac don't have proven direct carcinogenic effects on human health, so their CF are 0. The CF for atenolol and ciprofloxacin are not known. Fig. 7 presents the avoided ecotoxicity impacts for the laboratory and pilot installations.

The results demonstrated high avoided impacts of greater than 96% for all studied compounds in the laboratory installation. This is based on stable elimination rates of the parent compounds reached in the laboratory installation thanks to the well-conditioned system and stable operational conditions (constant loads of macropollutants and MPs, absence of weather fluctuations, etc.). In case of the pilot installation, which was operated in real conditions, with real wastewater effluent, fluctuating concentrations of nutrients and MPs, changing weather conditions, etc., the difference in avoided toxicity impacts of atenolol was 49% for sand and 45% for activated biochar admixture. In the case of ciprofloxacin, the difference was 19% for sand and 15% for AB admixture. For carbamazepine was the difference 21% for sand and 7% for AB admixture. Removal of diclofenac resulted in similar avoided toxicity impacts, above 97%. These results confirm the results of (Venditti et al., 2022a,b), where the wetland unit with 15% AB admixture resulted in better overall performance demonstrated by higher pollutant

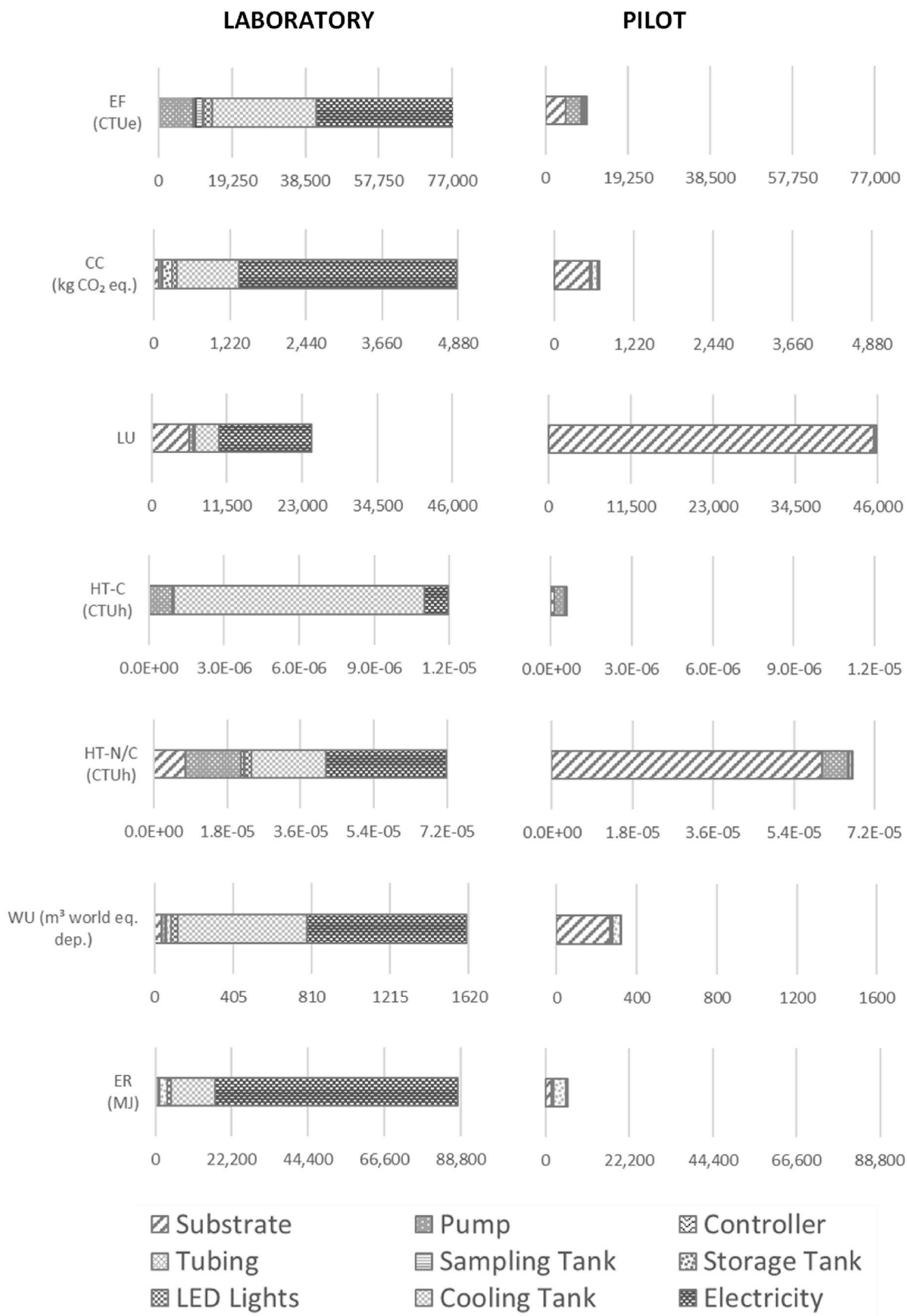

Fig. 6. Comparison of environmental burdens for seven impact categories for the a) laboratory- and b) pilot-scale CW installations (freshwater ecotoxicity (EF); climate change (CC); land use (LU); human toxicity (carcinogenic (HT-C) and non-carcinogenic (HT-N/C)); water use (WU); and non-renewable energy resources (ER)) (Notation for HT-C and HT-N/C expressed as E−05 equal to 10^{-5}).

Table 5
Percentage contributions of the individual components to the generated impacts for the seven studied categories of the laboratory- and pilot-scale CW installations (highlighted contributions for components representing greater than 5% of the overall impact) (freshwater ecotoxicity (EF); climate change (CC); land use (LU); human toxicity (carcinogenic (HT-C) and non-carcinogenic (HT-N/C)); water use (WU); and non-renewable energy resources (ER)).

Scale	Component	EF	CC	LU	HT-C	HT-N/C	WU	ER
Laboratory	Substrate	3.4%	6.5%	63.5%	0.8%	37.4%	9.7%	1.1%
	Pump	8.0%	0.8%	0.9%	7.4%	11.0%	1.0%	0.6%
	Controller	3.0%	0.3%	0.5%	0.6%	3.6%	0.4%	0.2%
	Tubing	0.2%	0.1%	0.0%	0.1%	0.1%	0.2%	0.2%
	Sampling Tank	10.0%	0.3%	0.0%	0.0%	0.1%	0.4%	0.6%
	Storage Tank	1.1%	14.2%	0.0%	0.6%	1.3%	6.9%	13.4%
	LED lights	13.7%	7.9%	2.4%	1.6%	7.8%	9.5%	6.0%
	Cooling Tank	25.9%	15.5%	6.8%	81.3%	14.8%	32.0%	11.8%
	Electricity	34.7%	54.4%	25.8%	7.6%	24.0%	39.8%	66.2%
Pilot	Substrate	66.7%	80.6%	99.4%	42.5%	95.4%	82.6%	27.3%
	Pump	16.9%	1.1%	0.2%	42.7%	3.1%	0.9%	1.5%
	Controller	1.6%	0.0%	0.0%	0.2%	0.0%	1.0%	0.0%
	Tubing	3.1%	1.4%	0.1%	3.8%	0.3%	1.5%	3.9%
	Sampling Tank	0.2%	0.3%	0.0%	0.2%	0.0%	0.3%	1.5%
	Storage Tank	7.3%	13.4%	0.2%	8.8%	0.9%	12.1%	58.2%
	Electricity	4.2%	3.1%	0.2%	1.8%	0.3%	1.5%	7.6%

Table 6
Characterization factors for assessment of the avoided environmental impact for freshwater ecotoxicity and human toxicity (carcinogenic and non-carcinogenic) (freshwater ecotoxicity (EF); human toxicity (carcinogenic (HT-C) and non-carcinogenic (HT-N/C)).

Compound	Characterization factors (PAF·m^3·day/kg)		
	EF	HT-C	HT-N/C
Carbamazepine	$7.80 \cdot 10^{2}$	0	$2.33 \cdot 10^{-6}$
Diclofenac	$1.94 \cdot 10^{3}$	0	$1.58 \cdot 10^{-4}$
Atenolol	$9.70 \cdot 10^{1}$	N/A	$1.50 \cdot 10^{-5}$
Ciprofloxacin	$2.09 \cdot 10^{4}$	N/A	$9.30 \cdot 10^{-6}$

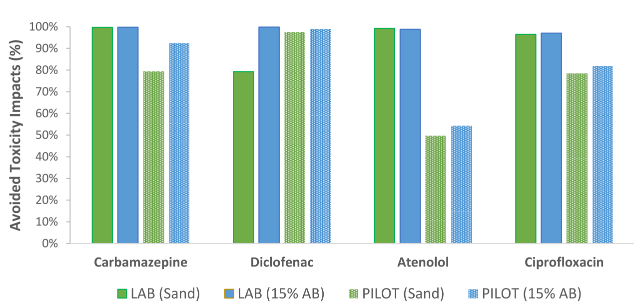

Fig. 7. Comparison of the avoided toxicity impacts of the removed MPs in the a) laboratory- and b) pilot-scale CW installations for sand and 15% activated biochar (15%AB).

removals, than the one with sand.

For the MPs' ERA, the PNEC values for the four targeted compounds were obtained from the Norman database as the lowest values in fresh water aligned with the Ecotox Centre. The RQ values are calculated for the inflow into the CW installations (which represents outflow of WWTPs) – RQI and for outflow from the installations – RQE. Based on the RQ values, the ERA revealed that for the laboratory installation the presence of the compounds in the inflow presented very high risk for diclofenac (104.3) and ciprofloxacin (98), followed by high risk for carbamazepine (1.4) and medium risk for atenolol (0.3). By applying CW in the laboratory this risk decreased to very high risk for ciprofloxacin (3.5 for 15%AB and 2.9 for sand), medium risk for diclofenac (0.3 for 15%AB and 0.1 for sand) and negligible risk for carbamazepine and atenolol (Fig. 8).

For the pilot installation, the RQ values at the inflow suggested a very high risk for diclofenac (19.9) and ciprofloxacin (1.7), followed by carbamazepine with low risk (0.07) and atenolol with negligible risk. After the treatment the environmental risk decreased to medium low risk for diclofenac (0.5 for sand and 0.2 for 15%AB) and ciprofloxacin (0.4 for sand and 0.3 for 15%AB) and negligible risk for carbamazepine and atenolol (Fig. 9).

For the MP removal evaluation, it is important to mention the experimental data management. For the actual MP concentration analysis, only parent compounds are considered, no further focus is given to the transformation products. The removal rates are calculated from the measured absolute concentrations, which are constant for laboratory installation (synthetic wastewater as influent) and fluctuating for the pilot CW system (real wastewater as influent). The final measured concentrations are influenced by the uncertainties of the analytics instruments, number of the methods and compounds' recovery rates, (e.g. recovery rate 1% for atenolol from wastewater).

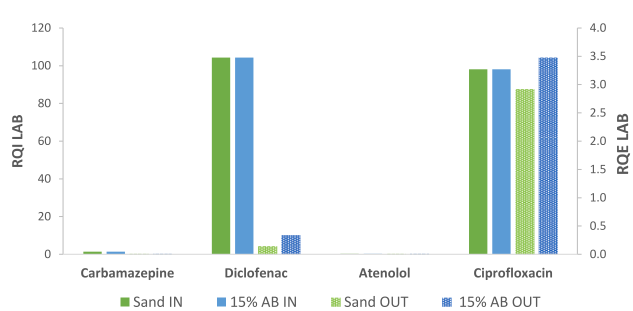

Fig. 8. Comparison of RQs for the four studied compounds in the laboratory installation at the inflow and outflow of the CW (RQI and RQE, IN and OUT respectively) for sand and 15% activated biochar (15%AB).

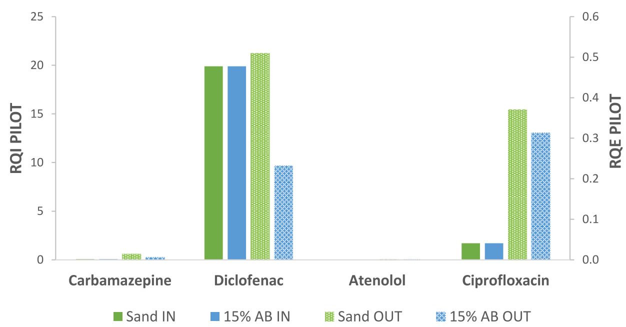

Fig. 9. Comparison of RQs for the four studied compounds in the pilot installation at the inflow and outflow of the CW (RQI and RQE, IN and OUT respectively) for sand and 15% activated biochar (15%AB).

3.4. Sensitivity and uncertainty analysis

Because the application of CW for removal of MPs from municipal effluents has not been object of many studies yet, an uncertainty and sensitivity analysis was undertaken to examine the potential impacts on the environmental burdens associated with the pilot installation.

3.4.1. Increased hydraulic loading rate

To evaluate changes in the overall environmental impacts of adjusting the HLR and increasing the loading on the pilot installation, the HLR was increased from 215 to 300 $l/m^2/d$. The purpose of increasing the HLR at the pilot installation was to experience the limitation of the CW with the load of real dissolved organic carbon and any type of organic matter. The normalised burden results are presented in Fig. 10.

This demonstrates a 28% reduction in the environmental burdens across all impact categories due to improved performance and capacity of the system with the same cross-sectional area. This highlights the importance of optimising the HLR to reduce the overall burdens of CW, whilst maintaining system performance for MP removal.

3.4.2. Revised boundary to exclude cooling tank

As the cooling tank is not directly part of the laboratory-scale installation, this scenario proposes a revised system boundary to exclude the cooling tank and quantify the change to the LCA results. The results show an average 26.9% reduction across all impact categories, with the HT-C burden reduced most significantly by 81.3% ($1.67{\cdot}10^{-6}$ CTUh). Among the six other impact categories, reductions in the environmental burdens of the laboratory installation ranged from 6.8% for LU (622.9) to 32.0% for WU (110.8 m^3 world eq. dep.). This highlights the sensitivity of including a component within the system boundary that is not directly attributed to the system, but still plays an important role in the experimental process.

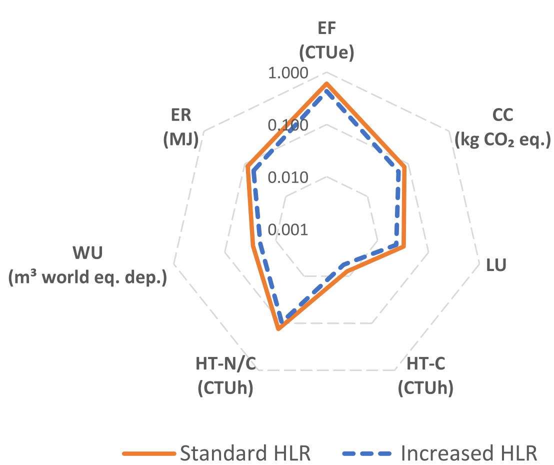

Fig. 10. Global normalised burdens (1 p.e.) comparing standard (215 $l/m^2/d$) and increased (300 $l/m^2/d$) hydraulic loading rate (HLR) for the pilot-scale installation (freshwater ecotoxicity (EF); climate change (CC); land use (LU); human toxicity (carcinogenic (HT-C) and non-carcinogenic (HT-N/C)); water use (WU); and non-renewable energy resources (ER)).

3.4.3. Offset from biomass production

To account for the potential impacts of including plant species in the LCA, the potential to plant macrophytes as a biomass was considered. The production of 5 kg of biomass from 1 m^2 pilot installation could offset the annual operational impacts related to electricity for pumping. A reduction in the total environmental burdens for all seven impact categories was negligible, with maximum reductions of 0.37–0.52% for EF, HT-C and LU impact categories, with less than 0.1% for the remaining four impact categories.

However, given the production of biomass relates to operational performance, its capacity to offset the operational energy demands of pumping. The results suggest a near three-fold offset for LU burdens (238.3 and 84.2) with notable reductions for EF and HT-C of 9.9% (30 and 301.9 CTUe) and 20.1% ($1.12{\cdot}10^{-9}$ and $5.56{\cdot}10^{-9}$ CTUh), respectively. The offset for the remaining four impact categories was negligible with a maximum difference of 2.3% was observed for these burdens.

3.4.4. Extended system lifespan

As the operational lifespan of most pilot-scale installation components was greater than one year, the system was examined over 5-, 10- and 20-year periods to evaluate the embodied versus operational burdens more critically. The results still present the substrate as the dominant component within the pilot installation, with an average contribution of 65.5% for a 5-year period (ranging from 21.0% for ER to 98.7% for LU) to 47.1% over the 20-year period (ranging from 8.8% for ER to 95.4% for LU). The extended lifespan increased the operational electricity contribution of the overall system, represents an average 11.3% at 5-years and rising to 22.8% over 20-years, with the greatest impact related to ER as it increased from 29.0% to 48.5% over this change in lifespan. The pump and storage tank represented two additional components with notable remaining burdens, with averages of 7.8–15.3% and 9.9–11.9% over these lifespans. The largest burdens for the pump were attributed to HT-C (36.7–61.8% depending on the system lifespan and need to replace the pump after 10-years) with ER represented the greatest impact category affected by the storage tank (34.6–44.7%). Changes for all other impact categories and for remaining components were marginal (<4% across all lifespans) in comparison to these key impacts.

3.5. Limitations

Some limitations exist within this study based on the constraints of the system boundary conditions and the assumptions made regarding assumed system performance over an extended period of time. This includes the omission of details relating to construction and excavation activities of a full-scale installations, the consideration of any unforeseen operational impacts, and the addition of gaseous emissions from the wastewater itself. The substrates were locally produced, thus the exclusion of transport burdens, but this would not fully reflect a full-scale system. Although this was outside the focus of the study, a full-scale installation would produce significant burdens related to the construction process and from gaseous emissions (N, organics) that were not present in the laboratory and pilot scale installations due to the use of synthetic wastewater preparation. Further research is required to capture the potential changes in environmental burdens between the pilot and full scale.

3.6. Sustainability in experimental and applied research: Lessons learnt

The results from this study identify several factors for considerations to support improved sustainability with the advancement of similar research from the early experimental phase in the laboratory-to pilot-scale installations and eventual full-scale deployment. This supports initiatives like My Green Lab, 2023, which aims to accelerate sustainability in experimental research settings, and this study proposes the following basic lessons learnt:

- Optimising experimental setup: scientifically justify the need for the size or and need for parallel experiments to avoid unnecessary replication of experiments and use of excess resources (e.g. the cooling tank's size was selected so that it can be used in subsequent experiments as well).
- Essential and green procurement: equipment and instrumentation should be acquired within the scope of green procurement guidelines and should be selected based on its function in multiple research studies (e.g. use of locally produced substrates reduces environmental burdens).
- Reducing operational energy demands: review experimental design procedures to reduce duration and timing of energy demands with aim of identifying passive alternatives to achieve similar requirements (e.g. accurate calculation of operational pumping demands and scheduling of the system, such as duration of resting periods allowing the wastewater to percolate through the unit).
- Informed experimental scheduling and setting: a suitable location and natural environmental conditions should be provided with the aim to reduce artificial controls during experimental research (e.g. the lab scale was placed at the University site and the pilot nearby).

4. Conclusions

The LCA and ERA methodologies helped quantify the impacts of constructed wetlands – substrates, embodied components and operational requirements – as a form of nature-based solution for passive wastewater treatment and MP removal from municipal effluent. Insights into more sustainable research are also addressed based on research findings.

The aggregated environmental burdens for seven impact categories suggested that a substrate with 15% activated carbon was 11.66 times greater as compared to a substrate with 15% biochar.

A broader comparison of the laboratory- and pilot-scale installations identified that the laboratory installation generated higher environmental impacts for majority (five) of the studied categories. This was attributed to a specific component, a cooling tank, which was for storage and required energy, cumulatively representing over 60% of the total burdens for these five impact categories). For the pilot installation, the component exploiting highest environmental demands was the substrate as it equalled two-thirds of the total burdens for five impact categories.

Regardless of these life cycle impacts, the CW environmental performance to support MP removal helps reduce emissions, with freshwater ecotoxicity and human toxicity impacts reduced by between 50% and 99.9%. The ERA revealed the decrease in MPs in the laboratory installation was greater than for the pilot installation, and risk ranging from very high for ciprofloxacin to negligible for atenolol in both systems.

A sensitivity and uncertainty analysis showed that increasing the HLR represented the greatest opportunity to reduce (28%) all environmental impact associated with CWs for effluent processing, with the capacity to ensure treatment efficiency. Offsetting the environmental impact through biomass production, and optimising the lifespan of a CW, presents other clear opportunities to reduce annual operational and embodied burdens.

Improving the sustainability of this type of research have briefly been outlined, with evident opportunities to apply principles and measures through design, procurement and operation. The findings from this study highlight areas of concern and opportunities for nature-based innovations whilst in parallel demonstrating how further improvements in environmental outcomes can be achieved throguh sustainable design of these systems.

CRediT authorship contribution statement

Hana Brunhoferova: Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Silvia Venditti:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition. **Joachim Hansen:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition. **John Gallagher:** Writing – review & editing, Writing – original draft, Visualization, Validation, Supervision, Software, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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