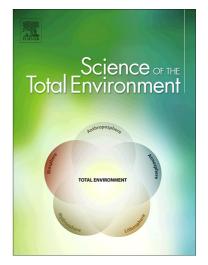
## Review

A review of nature-based solutions for greywater treatment: applications, hydraulic design, and environmental benefits

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## A review of nature-based solutions for greywater treatment:

## applications, hydraulic design, and environmental benefits

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## Highlights

- Nature-based solutions (NBS) are a viable option for greywater (GW) treatment
- Review of case studies provided numerical thresholds for hydraulic design of NBS
- Life cycle assessment studies demonstrated the benefits of NBS for GW treatment

## A review of nature-based solutions for greywater treatment: applications, hydraulic design, and environmental benefits

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## Highlights

- Nature-based solutions (NBS) are a viable option for greywater (GW) treatment
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Recognizing greywater as a relevant secondary source of water and nutrients represents an important chance for the sustainable management of water resource. In the last two decades, many studies analysed the environmental, economic, and energetic benefits of the reuse of greywater treated by nature-based solutions (NBS). This work reviews existing case studies of traditional constructed wetlands and new integrated technologies (e.g., green roofs and green walls) for greywater treatment and reuse, with a specific focus on their treatment performance as a function of hydraulic operating parameters. The aim of this work is to understand if the application of NBS can represent a valid alternative to conventional treatment technologies, providing quantitative indications for their design. Specifically, indications concerning threshold values of hydraulic design parameters to guarantee high removal performance are suggested. Finally, the existing literature on life cycle analysis of NBS for greywater treatment has been examined, confirming the provided environmental benefits.

**Keywords:** Nature-based solutions; Greywater treatment; Constructed wetlands; Green roofs; Green walls; Sustainable water management.

#### **1** Introduction

It is estimated that one third of the world's population does not have access to clean drinking water (Prüss-Üstün, et al., 2008; Ghaitidak & Yadav, 2013; World Health Organization, 2017). Global water shortage is due to a combination of population growth, economic development with extensive use of water in agriculture and industry, increasing standard of lifestyle, dietary changes, and climate change (Kummu, et al., 2010). Moreover, regulations for treating civil wastewater require progressively higher performances in removal of traditional and emerging contaminants (European Commision, 2012), increasing energy consumption and operating costs.

A sustainable management of water resource is therefore essential (Grant, et al., 2012). Greywater (GW) reuse can play a fundamental role, converting a significant fraction of wastewater (WW) from a waste to a valuable water resource (Friedler & Hadari, 2006). GW is defined as household wastewater made of all domestic WW with the exception of toilet flushes (e.g., WW produced in bathtubs, showers, and laundry machines) (Eriksson, et al., 2002). GW may represent up to 75% of total domestic WW, accounting for up to 100-150 L/PE/day in EU and high-income countries, and for smaller volumes in low-income countries

plants and to minimize the energy required for their treatment (Remy, 2008; Larsen, et al., 2009) because only the more polluted fraction of domestic WW is sent to the treatment plant (Otterpohl, et al., 2002). Additionally, reclaimed GW can be locally recycled for other uses (e.g., toilet washing, irrigation) which would otherwise employ high-quality water; in this way, a circular economy is promoted (Masi, et al., 2018).

Nature-based solutions (NBS) are defined as "actions to protect, sustainably manage, and restore natural or modified ecosystems, that address societal challenges effectively and adaptively, simultaneously providing human well-being and biodiversity benefits" (IUCN, International Union for Conservation of Nature, 2016) and also as "living solutions inspired by, continuously supported by and using nature, which are designed to address various societal challenges in a resource-efficient and adaptable manner and to provide simultaneously economic, social, and environmental benefits" (European Commission, 2015a). NBS are techniques that mimic natural processes in urban landscapes, including in WW management, with low inputs of energy and chemicals.

Different NBS have been implemented, including constructed wetlands (CWs), green roofs, green walls/living walls, and urban green spaces (e.g., parks, street trees). NBS contribute to save energy through reduced cooling loads (Alexandri & Jones, 2008; Castleton, et al., 2010), reduce flooding risk (Rizzo, et al., 2018), and promote a sustainable economic growth (Maes & Jacobs, 2017). Several studies have demonstrated the positive effects induced by greening vacant urban land on the well-being of the people (Tzoulas, et al., 2007; South, et al., 2018; Ling & Chiang, 2018). Finally, NBS support biodiversity by providing habitats for different species, significantly (Francis & Lorimer, 2011).

Planning and design aspects should be taken into account when implementing NBS. The aesthetic appearance is essential for their integration with the surrounding landscape and acceptance from local communities. One of the most critical limits to the application of NBS in densely built urban areas is the lack of available space. However, the outer surfaces of buildings can provide unused spaces in densely inhabited areas. From this perspective, green roofs and green walls represent two examples of NBS that are well suited to urban areas. Moreover, the use of GW for the irrigation of NBS can be the solution to the main critique addressed to urban greening, i.e., the large amount of water to maintain it.

GW treatment by Li, et al. (2009), the only listed NBS are constructed wetlands. GW treatment using green walls and green roofs is a very recent application: the oldest study is from 2008 (Frazer-Williams, et al., 2008). GW treatment using green walls and green roofs can be considered an adaptation of subsurface flow constructed wetlands, since they are based on a vegetated porous medium where water flows either vertically or horizontally. Unfortunately, to our knowledge, there is currently a lack of well-established criteria to guide the design of green walls and green roofs as NBS for greywater treatment.

The present paper is complementary to recent reviews focused exclusively on constructed wetlands as a specific type of NBS for GW treatment (Arden & Ma, 2018), or on specific issues such as the mechanisms of contaminant removal by plants and growing media, system longevity, and socio-cultural implications (Pradhan, et al., 2019). In particular, (Pradhan et al., 2019) addressed: significant issues related to the design of green roofs and green walls for GW treatment; pollutants removal mechanisms (both considering the plants and the growing media); problems that may compromise the durability of the systems; reduction in produced treated GW due to intense evapotranspiration in arid climates; potential damages to buildings due to the roots growth in green walls; reluctance to the reuse of treated GW, which is seen as contaminated in some cultural contexts; potential health issues related to the reuse of treated GW; costs for maintenance and operation. The aim of this paper is to provide an extensive review of the application of nature-based technologies to GW treatment and the evaluation of their performances at pilot and full-scale. Compared to existing literature, our goal is to clarify the following research questions (RQs): RQ1) are NBS appropriate for GW treatment? RQ2) is there enough available information to guide designers in the full-scale application of NBS for GW treatment? RQ3) is it possible to assess the environmental benefits connected to the application of NBS to GW treatment compared to conventional WW treatment processes?

## 2 Methodology

A thorough analysis of the scientific literature was performed on the Scopus database using the keyword "greywater" (or "grey water" or "gray water" or "graywater") combined with "constructed wetlands" (or "treatment wetlands"), "green roofs", and "green walls" (or "living walls" or "green facades" or "vertical gardens" or "vertical green systems"). The meta-analysis included all NBS categories, resulting in respectively 105, 34, and 15 articles for the combinations "greywater - constructed wetlands", "greywater -

"Article" or "Review", the numbers reduced to respectively 80, 19, and 10, indicating that about 30% of research studies were mainly conference papers. Among the intersections between the different typologies of NBS and the keyword "greywater", we were specifically interested in the applications related to greywater treatment. Therefore, we performed a distinction among studies specifically related to greywater treatment and studies related to other applications (i.e., irrigation) through a careful qualitative analysis of the abstract of each work.

Figure 1a shows the results of the bibliographic search. The studies found through the keyword "constructed wetlands" were entirely related to greywater treatment, since constructed wetlands are specifically conceived for wastewater treatment. The results indicate that constructed wetlands are the most employed NBS for GW treatment (105 studies), followed by green walls (11 studies) and green roofs (7 studies), despite the total number of studies related to green roofs is higher than those on green walls. Many applications of green roofs focus on the use of GW for irrigation, investigating the impact of GW on the health, growth, and evapotranspiration rate of the plants and on the provided services such as thermal insulation (Ouldboukhitine, et al., 2014). It should be noticed that green walls and green roofs can sometime be considered as modified applications of traditional constructed wetland systems since all these systems are based on the same fundamental principle, i.e., the coupling of biological, chemical, and physical processes within porous media enhanced by plants and microorganisms. Therefore, some studies specifically focused on green walls and green roofs could actually be included also in the "constructed wetlands" category (e.g., Ramprasad, et al., 2017), although aesthetical considerations played a more prominent role for their design in terms of visual appearance and plant choice.

The temporal distribution of studies on NBS designed for greywater treatments is shown in Figure 1b. The interest in constructed wetlands for wastewater treatment started in the early 1980s, with a growth around the beginning of the 21st century (Masi, et al., 2018). Initial efforts were directed to the treatment of mixed domestic wastewater, while the attention to a proper separation of wastewater and thus on the specific treatment of GW emerged later. Overall, the term "nature-based solutions" first denoted new approaches to mitigate and adapt to climate change effects (Eggermont, et al., 2015; MacKinnon, et al., 2008). Even the interest in green roofs and green walls was not initially focused on greywater treatment. In fact, the original applications of green roofs concerned the achievement of some benefits in urban areas such as stormwater

elements of buildings, providing architectural and environmental benefits (Köhler, 2008; Alexandri & Jones, 2008; Weinmaster, 2009). In the last decade, a growing interest in NBS designed for GW treatment can be observed (Figure 1b), and it is expected to increase in the next years, as research on the topic is now specifically dedicated to the optimization of treatment efficiency.

Figure 1: (a) Relative contributions of the 154 studies regarding greywater use coupled with different nature-based solutions (105 on constructed wetlands, 34 on green roofs and 15 on green walls) and (b) number of articles about NBS based on greywater treatment published per year. Solid colours are related to NBS applications concerning greywater treatment and reuse, while hatched fillings indicate other types of applications.

#### 3 Greywater: characteristics, conventional treatment methods and critical issues

## 3.1. Origin and amount

GW is the amount of domestic wastewater that excludes toilet flushing, and it is usually divided into two types: light GW, whose sources are bathrooms, showers, tubs, hand basins, and sometimes laundry; dark GW, which includes laundry facilities, dishwashers, and in some studies also kitchen sinks (Jefferson et al., 2004; Hourlier, et al., 2010; Ghaitidak & Yadav, 2013; Leonard, et al., 2016; Fowdar, et al., 2017). Light GW has a high potential for local treatment and reuse due to its low pollutant concentrations (Fowdar, et al., 2017). The reuse of treated GW can produce economic benefits in those countries where water resources are expensive. GW can be reused as service water (Nolde, 2000), i.e., for irrigation and toilet flush. Reuse could potentially save from 9 to 47% of potable water (Hourlier, et al., 2010; Fowdar, et al., 2017). However, GW reuse requires attention in terms of pathogen proliferation and infection diffusion, especially in case of bad system maintenance or GW storage.

The volume of produced GW depends on the population habits; up to 75-80% of wastewater is GW, over 90% in case of vacuum toilet installation. In high-income countries where water consumption per capita is higher, the fraction of GW is lower than for countries with low total water consumption, even if the high-consume countries produce larger volumes of GW (e.g., in North America 52% of GW/WW corresponds to 117 L/PE/day, while in Yemen 87% of GW/WW corresponds to 35 L/PE/day) (Ghaitidak & Yadav, 2013; Leonard, et al., 2016; Kaposztasova, et al., 2014). These differences are attributed to the fact that toilet

household toilets in low-income countries.

mushing represents a large machen or m

Household sources contribute differently to GW composition. The average contribution of three main different sources to the composition of GW are: 49% bath tubs, basins and showers; 27% kitchen sinks and dishwashers; 24% laundry sinks and washing machines. These values were calculated from twenty studies referring to sixteen countries (Australia, Brazil, Burkina Faso, Denmark, England and Wales, Greece, Holland, India, Israel, Jordan, North America, Oman, Yemen) (Al-Jayyousi, 2003; Ghaitidak & Yadav, 2013; Edwin, et al., 2014; Maiga, et al., 2014; Vakil, et al., 2014; Noutsopoulos, et al., 2018).

#### 3.2. Characteristics

GW quality is conventionally assessed through the parameters summarized in Table 1; typically, most studies focus only on a subset of them (e.g., Eriksson, et al., 2002; Leonard, et al., 2016; Fowdar, et al., 2017). For pathogen assessment, different Faecal Indicator Bacteria (FIB) are employed – among which *Escherichia coli* is the most used – even though their correlation with enteric pathogens has been questioned (Benami, et al., 2016). Xenobiotic Organic Compounds usually include surfactants, fragrance flavours, preservatives, and solvents.

Table 1. Conventional parameters to characterize domestic greywater

Almost all literature agrees that quantity and quality of domestic GW exhibit seasonal variations and depend on the characteristics of users (number, age, habits, activities), composition of commercial cleaning products, wastewater collection system, house facilities (e.g., dishwasher, washing machine), area type (e.g., urban, countryside, seaside). Furthermore, source and mix play an important role in the compounds that are in greywater. Usually, bath and hand basin GW contains personal care products (e.g. soap, toothpaste, shampoo), human-derived components (e.g. shave residuals, hair, skin, sebum), and also traces of faeces and urine (Eriksson, et al., 2002; Ghaitidak & Yadav, 2013); laundry GW contains high concentrations of chemical products (e.g. soaps and oils) and non-biodegradable fibres (Morel & Diener, 2006; Ghaitidak & Yadav, 2013); kitchen GW (from sinks or dishwashers) contains food, oils, fats, and detergents (Noah, 2002; Ghaitidak & Yadav, 2013). Both baths and laundry are the most frequent origins for faeces, skin, and food residuals, especially if there are babies in the house (Leonard, et al., 2016). By definition, urine is not

of urine contamination (Eriksson, et al., 2002; Leonard, et al., 2016). Storage time has also an impact on greywater characteristics: quality improves within the first day of storage but drastically decreases after 48 h (Eriksson, et al., 2002; Liu, et al., 2010).

An extensive comparison of the properties of GW from different sources was proposed by Eriksson et al., (2002). According to this study, it is hard to find in literature comparable data about parameter ranges or average values: some authors split different sources, while others consider a mixed GW; sometimes average values are given (with or without standard deviation), while other times ranges are provided. Most of the published papers organize data on either country or source bases. Table 2 adopts the country-based approach, comparing physico-chemical properties from published reviews and articles that analysed real domestic GW from 28 countries. None of the parameters exhibits a trend that allows grouping countries according to a geographic criterion. An income criterion has often been used to group countries (Ghaitidak & Yaday, 2013) but the comparison of the collected studies in Table 2 does not support this choice. Some parameters have similar ranges in almost all countries. For instance, pH fluctuates around the neutral value, and typical values of total suspended solids (TSS) are below 500 mg/L with peaks of the order of 1000 mg/L. Chemical Oxygen Demand (COD) and Biochemical Oxygen Demand (BOD<sub>5</sub>) values are usually of the order of some hundreds mg/L, although a few values larger than 1000 mg/L have been reported (e.g., Israel, Malaysia, Jordan, United Arab Emirates). Total nitrogen (TN) and total phosphorus (TP) generally exhibit concentrations below a few tens mg/L and a few mg/L, respectively, with only a few exceptions (e.g. Palestine and Jordan). Faecal Indicator Bacteria (FIB) instead present a huge variability both among and within countries, with variations in concentration spanning many orders of magnitude.

Some studies considered synthetic GW (Diaper, et al., 2008; Hourlier, et al., 2010) due to its reproducibility and consistency in composition. Synthetic GW is usually prepared combining specific amounts of laundry soap, shampoo, oil, hand soap, etc. Different recipes have been used in terms of kind and quantity of products. In order to ensure the consistency of synthetic GW recipe, it is necessary to confirm the availability of the specific products (brand or exact composition). For this reason, Hourlier, et al., (2010) suggested a composition that uses only chemical products of technical quality. Najem & Scholz (2016) provided a comparison between different recipes: some of them use commercial products, others chemical ones. There

Chrispim & Nolasco, 2017).

## 3.3. Quality standards

Quality requirements for GW reuse are available, as regulations and guidelines, in a number of countries. Quality requirements and reuse limits depend on type of reuse, on origin of GW, and on possibility of human contact with recycled water (Eriksson, et al., 2002; Hourlier, et al., 2010; Abu Ghunmi, et al., 2011; Ghaitidak & Yadav, 2013; Leonard, et al., 2016). Country reuse standard comparison has been provided in literature (Edwin, et al., 2014; De Gisi, et al., 2015), but a more widespread regulation has been highly recommended in recent studies (Al-Ismaili, et al., 2017).

Table 3 shows the mandatory standards for Great Britain (BSI, 2011), USA (NSF/ANSI 350 – 2011, 2011), New South Wales – Australia (NSW-DEUS, 2007; Vuppaladadiyam, et al., 2018), Italy (D.M. 185, 2003), and Japan (JSWA, 2016; Vuppaladadiyam, et al., 2018). The Table also lists the guidelines voluntarily adopted in Germany (USEPA, 2012; Jokerst, et al., 2011), Slovenia (Sostar-Turk, et al., 2005), Jordan (Abu Ghunmi, et al., 2011), China (Zhu, et al., 2016), India (Hosseinzadeh, et al., 2015), and Canada (Chaillou, et al., 2011). Even though the parameters in Table 3 are not fully consistent among different countries, standard limits have been proposed by the existing guidelines for pH, BOD<sub>5</sub>, COD, suspended solids, total phosphorus, and faecal bacteria. Values of these standard limits in Table 3 exhibit considerable variations, with differences among countries that are larger than one order of magnitude.

Table 4 offers an overview of the guidelines for GW reuse in Europe and the world. These guidelines have been developed to encourage and enhance the reuse of GW and provide recommendations for a proper reuse, with a specific focus on managing health risks related to the use of non-treated GW.

1	Journal Pre-proofs
2	(average), depending on available data.
3	
4	Table 3: Overview of regulations and guidelines for GW reuse
5	Table 4: Proposed guidelines and reports for GW reuse (no standards or limits are available)

## 7 **3.4.** Conventional treatment processes

Table 5 summarises the main conventional GW treatment options in the last 20 years, while a comparison between these treatment processes and GW treatment through NBS is presented later (see Section 6). GW treatment processes are classified according to: type of removed contaminants and adopted process. It is of interest to underline that in general the treatment efficiency depends on operational conditions and on GW origin and composition. Bathroom and washbasin GWs contain soap up to 90% of loading mass and low concentrations of biologically-essential macro- and micro-nutrients (phosphorus, nitrogen, potassium) with respect to carbon, which may not be effectively removed through a biological process.

Among chemical GW treatment processes, the most adopted are coagulation and flocculation, which achieved the following removal percentages: 85-89% BOD<sub>5</sub>, 64% COD, 13% total nitrogen, >99% TOC and >99% *E. coli* (Ghaitidak & Yadav, 2013).

Among physical GW treatments, filtration is the conventional technique to remove turbidity, colloids and residual suspended solids. The removal efficiencies are reported in Table 5. Generally achieved removal efficiency of filtration systems are (Noutsopoulos, et al., 2018): 53–93% TSS, 89–98% BOD<sub>5</sub>, 37–94% COD, 5–98% total N, 17–73% NH<sub>4</sub><sup>+</sup>-N, 0–100% total P, 12-99% MBAS, 100% *E. coli*, 100% Calcium, 100% Magnesium, 47% Sodium and 56.2% Potassium.

Biological methods for GW treatment are divided in aerobic and anaerobic. Generally, partially submerged rotating biological contractors (RBCs) are used for BOD<sub>5</sub> removal and combined carbon oxidation/nitrification of secondary effluents, while completely submerged RBCs are used for the same applications with additional de-nitrification (Wu, 2019). Moving bed biofilm reactor (MBBR) represents a widely applied wastewater and GW treatment technology, based on organic matter oxidation and nitrogen removal. According to origin of GW, the removal efficiencies of BOD<sub>5</sub> and COD were 59% e 70%

29	Journal Pre-proofs
30	optimised to reach the GW reuse standards. Generally achieved removal efficiencies of membrane bioreactor
31	(MBR) systems are: 98–99.9% turbidity, 100% TSS, 93–97% BOD <sub>5</sub> , 86–99% COD, 52–63% TN, 6–72%
32	NH4+-N, 19% TP and 10-40% PO43-P. MBR systems satisfy most standards for water reuse, as pH,
33	turbidity, BOD <sub>5</sub> , COD, TSS and NH <sub>4</sub> <sup>+</sup> -N (Chrispim & Nolasco, 2017). Sequencing batch reactor (SBR)
34	systems are characterized by a great flexibility of operation and time-controlled sequence, and they are
35	considered an effective nutrient removal technology (Rodda, et al., 2011). As a whole, BOD <sub>5</sub> removal varied
36	from 80 to 98% and a similar range of COD removal values is observed. Effluents from SBR treatment of
37	GW from showers satisfied NH <sub>4</sub> <sup>+</sup> –N, BOD <sub>5</sub> and COD standards for wastewater reuse (Lamine, et al., 2007).

Table 5: Conventional technologies for GW treatment and reuse.

## Journal Pre-proofs **4 Nature-based technologies for greywater treatment**

40 Constructed wetlands (CW) were the first NBS applied to GW treatment (see section 2). GW remediation 41 through green walls and green roofs involves the simultaneous presence of a wide range of biological and physico-chemical processes, according to the operation mode of CW (Kadlec & Wallace, 2009). The 42 43 efficiency of these systems in WW treatment is due to a strong interaction among plants, biofilms, substrate, 44 atmosphere, and nutrients from wastewater. The contact among roots, substratum, and biofilm favours 45 different fundamental mechanisms of pollutant and pathogen removal, such as sedimentation and filtration as 46 physical processes, precipitation and adsorption as chemical processes, and microbiological degradation and 47 plant uptake as biological processes. The large amount of involved mechanisms entails a huge variability in 48 removal efficiency (Arden & Ma, 2018). Despite the growing interest on CW application to GW over the last 49 decade, most literature refers to pilot systems with only a few full-scale applications.

50 The application of green walls and green roofs for GW treatment, despite being much more recent, has been reported by a comparable number of studies. Therefore, the present section was divided into: pilot-scale 51 52 constructed wetlands (section 4.1.1); full-scale constructed wetlands (section 4.1.2); green roofs and green walls (section 4.2). Given the low number of literature case studies, a comparison was also established with 53 lightly loaded constructed wetlands (LL-CWs), namely studies performed with secondary or tertiary treated 54 wastewater. Among the papers related to GW treatment using NBS identified through the literature survey 55 56 (see Section 2), only the studies concerning experimental applications were included as references. Specifically, we analysed 30 datasets as pilot constructed wetlands, 15 studies as full scale CWs, and 10 57 studies as green roof and green wall applications. Compared to the studies shown in Section 2, we here 58 analyse the subset of studies of NBS where treatment performances were quantified. 59

Figure 2 shows the relationships among the main operational factors of the treatment systems considered in 60 the meta-analysis, distinguishing among CWs treating GW (pilot and full scale), CWs treating lightly loaded 61 WW, green roofs and green walls. Specifically, the upper panel represents the relationship, where available, 62 63 between hydraulic retention time (HRT) and hydraulic loading rate (HLR), while the lower one represents 64 the relationship, where available, between organic loading rate (OLR) and HLR. As expected, the three parameters are correlated. The plot in Figure 2a indicates a negative correlation between HRT and HLR with 65 66 longer retention times corresponding to lower HLRs, with a few exceptions for lightly loaded CWs. This 67 means that the two parameters provide similar information about the hydraulic behaviour of the system, and

68	Journal Pre-proofs
69	HLR or HRT. As to OLR, Figure 2b shows that it is directly proportional to HLR, as expected from the
70	relationship among these two parameters (OLR = HLR $\cdot C_{COD,i}$ , where $C_{COD,i}$ is the COD concentration in the
71	influent). The correlation among HLR, HRT, and OLR allows to investigate the treatment efficiency of
72	different systems primarily as a function of only one of these parameters, and the derived optimization
73	criteria can then be extended to the other parameters as well. Specifically, in our analysis we focus on HLR,
74	that is available or easily calculated for most of the considered case studies while HRT and OLR were
75	reported less frequently. Moreover, HLR is a very informative parameter for designers of systems treating
76	GW as it allows to determine the space required for the system.

79

#### Journal Pre-proofs

Figure 2: (a) Hydraulic retention time (HRT) and (b) organic loading rate (OLR) versus hydraulic loading rate (HLR). Pilot and full
scale constructed wetlands treating greywater (GW-CWs), lightly loaded constructed wetlands (LL-CWs), and green roofs and green
walls are distinguished. The regression lines do not include data from LL-CWs.

83

#### 84 4.1. Constructed wetlands: applications to GW

Both natural and CWs biodegrade or immobilize a range of emerging pollutants, including certain 85 pharmaceuticals, and perform better than conventional "grey" solutions (Kadlec & Wallace, 2009; Arden & 86 87 Ma, 2018). CWs for domestic wastewater treatment are widely recognized as a sustainable, low-cost, low 88 maintenance technology, given the reduced energy needs and simplicity of operation (Massoud, et al., 2009; 89 Kadlec & Wallace, 2009). The most common types of CWs include horizontal or vertical subsurface flow 90 (Figure 3), which restrict the contact with the wastewater, and provide good removal of total suspended 91 solids and organic matter, even with low spatial footprint (about 1 m<sup>2</sup>/PE in cold and temperate climates, less 92 than 0.5 m<sup>2</sup>/PE in warm climates (Masi, et al., 2010)). CWs can also be designed as free water surface 93 systems, which have the highest biodiversity level amongst all the possible wetland configurations (Hsu, et 94 al., 2011). Additionally, the abundance of organic surfaces (litter, plant stems, microalgae, etc.) supports the 95 adsorption of persistent organic pollutants, which are then slowly degraded (Matamoros, et al., 2016). 96 Several full-scale CWs for GW treatment are described in literature for single house applications, in many 97 cases with the goal of reuse, as well as other applications such as in condos or institutional facilities.

The use of CWs for the treatment of GW was identified as key option to promote a sustainable water 98 management in accordance with circular economy principles (Masi, et al., 2018). Within this framework, 99 100 Arden & Ma (2018) listed 11 different treatment schemes and the following ranges of removal efficiency: BOD<sub>5</sub> 63-98%; TSS 64-98% for subsurface flow systems and 25% for free water surface; turbidity 47–97%; 101 TN 44-59%; TP 24-63%; around 1-2 log removal for bacteria, protozoa and viruses. Despite high pathogen 102 removal, the authors highlighted the need of some additional disinfection steps (such as chlorine and UV) to 103 104 meet strict water reuse standards. Arden & Ma (2018) also evidenced a relatively low number of studies on 105 CWs for GW treatment and reuse application, with only 13 peer reviewed papers describing a total of 38 systems. Moreover, the majority of the literature deals with pilot systems and very small applications, with 106

107 Long a terr works (Faulti, et al., 2010, 1 allo, et al., 2010, 100, 101, 2010) retering to tar scale systems.
108 Therefore, the next section reviews the available literature, separately analysing the insights gained from
109 pilot studies and the diffusion of full-scale applications of CWs for GW treatment and reuse.

- 110
- 111

Figure 3: Schemes of (a) horizontal and (b) vertical subsurface flow constructed wetlands (Courtesy: IRIDRA srl).

112

## 113 4.1.1 Pilot-scale systems

The analysed pilot-scale studies included 13 peer-reviewed papers concerning constructed wetlands treating 114 greywater (GW-CWs) and included mostly subsurface flow systems, since free water surface systems could 115 be found only on Gerba, et al. (1999), in Arden & Ma (2018) and Jokerst et al. (2012). A total of 30 116 individual pilot systems were analysed, with more than half being horizontal-subsurface flow systems. Given 117 the small number of cases, a comparison was established with lightly loaded constructed wetlands (LL-118 CWs), namely studies performed with secondary or tertiary treated wastewater. A total of 24 individual 119 datasets were collected from 9 peer-reviewed studies with CWs treating secondary or tertiary wastewater. 120 121 From this set only 2 of the systems were surface flow.

Table 6 presents the range of inflow characteristics of GW and wastewater used in lightly loaded constructed wetlands (LL-CWs). Parameter values are in line with the ranges shown in Table 2. It can be observed that LL-CWs systems receive lower concentrations than GW systems for most parameters, except for TSS where the range of inflow concentrations is similar.

126

127 Table 6: Range of inflow characteristics of constructed wetland treating greywater (GW-CWs) and constructed wetlands treating
128 lightly loaded wastewater (LL-CWs).

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Figures 4a-b show the range distribution of COD and BOD<sub>5</sub> removal rates for GW-CWs and LL-CWs, as a function of HLR. Only systems for which HLR was available (or could be calculated) are displayed. The analysis of BOD<sub>5</sub> data (Figure 4a) shows removal rates mostly higher than 85%, for a HLR up to 450  $L/m^2/day$  with a global average of 90%. This behaviour indicates that GW has a higher degradability than secondary or tertiary wastewater, which is consistent with the lower COD removal rates in LL-CWs. Similarly, BOD<sub>5</sub> removal rates for LL-CWs are lower than for GW-CWs, and consistently close to a plateau

Journal Pre-proofs 136 137 removal rates higher than 60% for all systems except one, with a global average of 83%. Most LL-CW systems have a consistent removal rate around 50%, for HLR as high as 2100 L/m<sup>2</sup>/day with a global average 138 of 52%. This consistency seems to show an upper removal limit capacity for organic matter removal, most 139 140 likely revealing a more recalcitrant nature of COD in secondary or tertiary wastewater. The higher 141 biodegradability characteristics of GW had been previously reported in several studies in areas of water 142 availability (Karlgren, et al., 1967) as well as in water scarcity areas (Al-Jayyousi, 2003; Sall & Takahashi, 2006). 143

The removal efficiency for TSS of GW-CWs showed values higher than 85% for HLR between 80 and 450 L/m<sup>2</sup>/day (Figure 4c). For loading rates below 80 L/m<sup>2</sup>/day efficiencies were much lower, which might be due to biomass detachment since the corresponding HRT values are very high (>5 days). Most LL-CW showed TSS removal efficiencies higher than 60% for HLR up to 500 L/m<sup>2</sup>/day, dropping to values lower than 50% for higher loads.

Regarding nutrient removal, it was observed that inflow concentrations of TN and TP varied significantly, 149 between 5 and 34 mg/l and from near zero to 23 mg/l, respectively. In terms of removal efficiency, GW-CWs 150 and LL-CWs treatment systems showed a similar behaviour regarding TN, with a good removal between 151 152 46% and 78% for all systems except one (Figure 4d). On the other hand, TP removal in these systems (Figure 4e) showed very different results, including both high removal rates (close to 100%) as well as 153 extremely low levels (reaching negative values). This wide range of behaviours could be due to the filling 154 155 media in the CWs, which was mostly gravel or native stones. The only exception was a FWS system (Jokerst, et al., 2011), which had a 0.9 m deep base layer, consisting of approximately 50% native soil and 156 50% peat. In this system TP removal efficiency was 58%. 157

The number of studies considering pathogen concentrations in GW-CWs is low, and the analysis of different parameters between studies also hampers a consistent meta-analysis. Overall, GW data showed significant inflow concentration in several bacterial parameters, including Total Coliforms (TC), Faecal Coliforms (FC) and *E. coli*, stressing the importance of treating GW prior to direct contact use (see Table 2). GW-CWs exhibited significant removal levels for TC (Figure 4f), higher than 2.8 log units for all systems analysed. Nevertheless, outlet concentrations in most of the systems reached average concentrations in the order of 104 (Gerba, et al., 1999; Winward, et al., 2008), which is not enough to meet standards consistent with several

165	Journal Pre-proofs
166	typically one order of magnitude lower than GW-CWs. From a visual assessment of the possible correlation
167	between TC removal and HLR, the existence of a specific connection seems unlikely. This can suggest that
168	the main removal mechanism could be due to adsorption on media and filtration processes and, thus, strictly
169	related to the physico-chemical properties of the growing medium (Prodanovic, et al., 2017).
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173	
174	Figure 4: Removal efficiency data as a function of hydraulic loading rate (HLR) for (a) BOD <sub>5</sub> , (b) COD, (c) total suspended solids
175	(TSS), (d) total nitrogen (TN), (e) total phosphorus (TP), and (f) Total coliforms. Pilot and full scale constructed wetlands treating
176	greywater (GW-CW), lightly loaded constructed wetlands (LL-CW), and green roofs and green walls are distinguished. Values of
177	removal efficiencies in panel (f) refer to different bacterial indicators (Table 6 and Table 10).
178	

- 179 Table 7: Pilot scale CW for GW treatment and reuse from peer-reviewed literature (FWS = free water surface; VF = vertical flow; HSSF = horizontal subsurface flow; RVF = recycled vertical flow
- 180 *bioreactor;* SF = sand filter).
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The analysis of peer-reviewed manuscripts on full-scale systems evidenced the suitability of CWs for GW 183 treatment and reuse at real scale and conditions (Figure 5). Laaffat et al. (2015) reported satisfactorily high 184 removal efficiencies from a 100 days long monitoring of a horizontal flow constructed wetland (HF CW) 185 treating GW from a primary school in Marrakesh: BOD<sub>5</sub> >92%, COD >85%, TSS >94%, TN >45%, TP, TC 186 187 >99%. Paulo et al. (Paulo, et al., 2009) evidenced a high buffer capacity in terms of treatment performance of a hybrid HF/VF system treating GW from a single house with 9 people; the CW faced both daily and 188 monthly variable influent GW loads, showing high stability in overall treatment performances over a 120 189 190 days monitoring period (Table 8). Results from different CW systems in Mediterranean area monitored under the SWAMP (EC FP5) and the Zero-M (EC MEDA) projects (Regelsberger, et al., 2005; Scheumann, et al., 191 2009; Masi, et al., 2007; Masi, et al., 2010) demonstrated generally high removal efficiencies (see Table 8 192 193 and Figure 4). These full-scale studies also highlighted interesting information: (i) an overall sustainable water management approach can lead to high pollutant concentrations (relatively to the GW literature 194 ranges), since less water is used thanks to water saving devices; (ii) a polishing FWS stage can be adopted to 195 both accumulate treated GW for reuse and naturally disinfect it, since 4-6 logs of removal and concentrations 196 of 1-200 MPN/100ml were observed; (iii) a FWS tertiary unit can avoid the use of technological disinfection 197 198 units for unrestricted reuse; this choice is of particular interest for developing countries, even though the intense evapotranspiration can highly reduce the amount of produced "new water"; (iv) the use of a FWS 199 200 stage can lead to occasional increases in effluent COD in warm periods due to algae bloom or volume 201 reduction by evaporation, even if observed concentrations remain low (COD always below 100 mg/l). For particular cases with low values of ammonia and TN, the adoption of a HF system can bring advantages in 202 203 comparison to VF because of the different feeding needs (generally VF systems, operated in batch, are more 204 energy dependant in comparison to HF ones).

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Figure 5: Examples of CWs for greywater treatment and reuse: on the left, VF for treatment of GW from the College of Engineering
in Pune (India), reused for gardening and toilet flushing; on the right, beduin village in the West Bank (Palestine), GW treated by a
VF and reused for irrigation of olive plantation and fodder (Courtesy: IRIDRA srl).

209

Journal Pre-proofs 210 guidelines for design, construction, and operation of CWs for domestic and municipal wastewater (DWA), 211 which specifically discuss design indicators dedicated to the case of GW treatment (Nivala, et al., 2018). A 212 survey of technical literature was hence performed to investigate the diffusion of CWs for GW treatment and 213 214 reuse. To this aim, two sources were analysed: (i) Sustainable Sanitation Alliance (SuSanA -215 www.susana.org) case study database; (ii) personal information collected from Global Wetland Technology members (GWT – www.globalwettech.com), a consortium of 10 among the most relevant design companies 216 in the field of CWs. The results of this search are summarised in Table 9, which reports details from 15 cases 217 of HF and VF CW applications in real conditions for different GW types (from light GW of showers and 218 washbasins to dark GW of kitchens, as well as car washing GW). The scale of application and the volume of 219 220 treated GW are generally much higher than those reported in peer-review literature; CW systems up to hundreds of square meters were designed for real conditions, demonstrating the possibility to implement 221 NBS for GW treatment at large scales and not only for single houses. For those systems for which removal 222 efficiencies were available, the results are consistent with pilot-scale studies, in particular for organic matter 223 removal and TSS (Figure 4). The use of additional disinfection units seems to strongly vary among countries 224 and scales, probably driven by different legislative water quality standards for reuse, while the reuse of 225 226 treated GW for toilet flushing without additional disinfection also occurs. Various reuses of treated GW were proposed, ranging from irrigation to toilet flushing, as well as more productive reuses such as agro-forestry 227 and industrial reuse (i.e., concrete mixing and aquaculture). It is important to stress that the analysis of 228 technical literature presented here only represents a limited portion of real applications, and we reasonably 229 expect a much larger number of similar applications worldwide. Therefore, CWs should be considered a 230 231 mature technology for the treatment of GW and successive reuse.

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Table 9: A list of full-scale CW for GW treatment and reuse from Global Wetland and Technology Database.

Table 8: Full scale CW for GW treatment and reuse from peer-review literature (HF = horizontal flow; VF = vertical flow; FWS =

free water surface).

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## **4.2** Urban green infrastructures: Green walls and green roofs

Green roofs and green walls (Figure 6) represent important techniques for urban reconciliation ecology 240 (Francis & Lorimer, 2011), and are becoming integrated parts of modern building in many countries. Green 241 roofs are vegetated surfaces installed on a rooftop and generally made up of modular systems consisting of 242 an insulation layer, a waterproof membrane, and a vegetation layer planted in a growing substrate. The depth 243 of this substrate can range from 50 mm to over 1 meter, depending on the weight-bearing capacity of the roof 244 and the type of vegetation (Oberndorfer, et al., 2007). Green walls are vertical systems usually attached to 245 internal or external walls, and are made up of planted containers attached to the wall itself. The vegetation is 246 hence rooted across the entire vertical structure of the wall and not only at its base as for green facades 247 248 (Manso & Castro-Gomes, 2015). These green infrastructures were originally developed for aesthetic and 249 insulation purposes (Teotonio, et al., 2018), and they have only been recently proposed for wastewater 250 treatment systems.

251

- Figure 6: Examples of (a) green wall in Sydney (Courtesy: dr. Irene Soligno) and (b) aesthetic green roof at the Horniman Museum
  in London, UK (Reproduced without modifications under Creative Commons license CC BY-SA 2.0; author: secretlondon123).
- 254

Green roofs and green walls for GW treatment operate similarly to reed beds, in which GW percolates 255 256 through planted pots filled with a combination of granular media such as vermiculite, sand, growstone, 257 expanded clay, phytofoam, coco coir, and perlite (Prodanovic, et al., 2017). However, their low space 258 requirements represent a relevant advantage compared to the conventional reed beds (Masi, et al., 2016). If 259 GW treatment is sought, the choice of plants must fulfil multiple criteria: fitness to local climatic conditions and high survival capacity, aesthetic appearance (e.g., perennial species), low space required for root growth, 260 low weight, and good removal capacity of nutrients (Castellar da Cunha, et al., 2018). Additional features 261 262 such as potential use for agricultural or medical purposes are certainly added values (Eregno, et al., 2017). In this section, we provide a case study review of green roofs and green walls used as GW treatment systems, 263

with specific focus on removal efficiencies of the main contaminants. It is interesting to notice that the first studies on the irrigation of green infrastructures with GW focused on pilot applications on actual systems, while laboratory studies were only performed later to investigate the role of different treatment processes or

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with pilot studies, and concluding with real cases.

269 The first example is given by Fowdar et al. (2017), who employed a set of laboratory columns filled with 270 inert material (sand, gravel) and planted with ornamental species (climbers and flowers) to reproduce a living 271 wall system. The study investigated different operational and design aspects in terms of loading rates, 272 saturated zone design, vegetation species, filling materials, inflow concentrations, and dosing frequency. Compared to traditional stormwater biological filters, the authors tested if creating a saturated layer at the 273 filter bottom can improve nitrate removal through denitrification. However, it was observed that 274 275 denitrification was limited by the availability of organic matter in the saturated layer. The only carbon source 276 provided by the GW was quickly removed in the upper layers, limiting the presence of biodegradable matter 277 where denitrification could occur. The performance of the system in terms of BOD<sub>5</sub> removal proved to be 278 very high, while some improvements were still required for nitrogen and phosporous removal. A follow-up 279 experimental study (Fowdar, et al., 2018) investigated the main mechanisms governing removal and 280 transformation of nitrogen in living wall biofiltration systems, with particular attention to dissolved organic 281 nitrogen. The limitation observed in the previous experimental setup was addressed redesigning the saturation zone to enable the injection of an external source of organic carbon. However, the application of 282 283 an isotopic tracer (<sup>15</sup>N-urea) revealed a minor removal contribution provided by nitrification-denitrification pathway. Plant assimilation and media adsorption were likely predominant, highlighting the importance in 284 plant selection in the design phase. 285

Another laboratory study (Prodanovic, et al., 2017), examined different growing media for green walls. The 286 287 study aim was to identify the underlying pollutant removal processes and to maximize the treatment capacity 288 of the system. Experimental tests were performed in vertical columns filled with different porous media and 289 without vegetation. The tested materials were selected on the basis of physical and chemical characteristics 290 such as specific weight, water and nutrient retention capacity, porosity, capacity to distribute water and 291 support plant growth, sustainability, and local availability. Among the hydraulically fast and slow media 292 identified as suitable media, the analysis identified perlite and coconut coir as the two best performing 293 media, suggesting their combined use as best option. Mixing two materials with different hydraulic behavior 294 allows, on one hand, to limit clogging issues, and, on the other hand, to increase the treatment efficiency, providing enough time for biological processes. A following study (Prodanovic, et al., 2018) examined the 295

297 providing a complete picture about the physico-chemical removal processes. It was observed that biological 298 removal processes dominate nitrogen and COD removals, while physico-chemical processes dominate 299 removal in fast media.

300 Some pilot-scale implementations of green roofs irrigated with GW include adaptations of CWs located on 301 building rooftops, as in the Green Roof-top Water Recycling System (GROW), which was developed by Water Works UK Ltd. (London, UK; patent number GB 2375761) (Avery, et al., 2007). The system consists 302 of an inclined framework of interconnected horizontal troughs and weirs planted with several plant species. 303 304 The GROW system was compared with more established horizontal flow CW and vertical flow CW for the treatment of domestic GW (Frazer-Williams, et al., 2008). The study found a better efficiency of the GROW 305 system in reducing suspended solids and turbidity, with no significant difference in effluent BOD<sub>5</sub> and COD 306 concentrations. Ramprasad et al. (2017) evaluated the performance of the GROW system under various 307 climatic conditions, organic loading rates, and flow rates in order to understand the suitability of the system 308 for different geographic areas and climates. Moreover, they focused on the performance of the system in 309 removing surfactants from personal care products. The system was able to reach a removal efficiency higher 310 than 80% for all the parameters with values of HRT and HLR ranging from 0.8 to 1.3 days and from 7.75 to 311 312 15 L/m<sup>2</sup>/day, respectively. Overall, the removal efficiency depended on seasonality and hydraulic parameters. A lower environmental impact of GROW system compared to membrane based technologies 313 was demonstrated through a Life Cycle Assessment, mostly because of the electricity consumption during 314 315 the use phase (Memon, et al., 2007).

316 An experimental study concerning green roofs irrigated with GW was performed in the city of Al Ain 317 (United Arab Emirates), testing the difference in treatment capacity between intensive and extensive roof systems (Chowdhury & Abaya, 2018). Intensive and extensive prototypes were differentiated only for the 318 319 depth of the growing layer, while other characteristics were identical for the two tipologies. Generally, intensive green roof prototypes showed higher removal performances compared to extensive prototypes and 320 321 this can be attributed to the greater depth of the soil. However, the short retention time of the experiment – much less than 24 h – limited the removal efficiency for certain parameters such as turbidity and COD in 322 both systems. 323

Journal Pre-proofs 324 325 adapted biological filter intermittently dosed with domestic GW (Svete, 2012). In spite of the limited spatial 326 footprint (filter surface area around 0.54 m<sup>2</sup>), the system was able to operate with good removal performance 327 as enhanced aerobic conditions and residence times supported fast biogeochemical reaction rates. The system 328 included three different sections, among which the planted one showed higher removal rates, satisfying all 329 the expected treatment performances for biological filters in Norway, with the exception of E. coli. However, an Anova analysis demonstrated that the contribution of vegetation to treatment was not relevant, suggesting 330 331 that the higher perfomance were most probabily due to higher residence time and confirming the importance of this parameter. 332

A pilot installation of a green wall was conducted by Masi et al. (2016) for the treatment of GW produced by 333 334 an office building in Pune (India). The system involved a series of pots filled with porous media (LECA 335 mixed with either sand or coconut fibres) and with different plant species. The pots were arranged in 12 vertical columns (6 pots per column), and perforated pipes fed the upper row while the lower rows of pots 336 received water through vertical percolation. The effluent collected at the bottom of the wall was then reused 337 for garden irrigation. The main conclusion of the study was related to the importance of the substrate used in 338 the green wall. Even though TN removal was limited, the system was successful in minimizing the treatment 339 340 footprint while ensuring an effluent quality that respected the Indian regulations for reuse for irrigation 341 purposes and, for some samples, for toilet flushing.

Another example of full-scale application (VertECO – Vertical Ecosystem) was installed in a hotel in Lloret 342 de Mar (Spain) (Gattringer, et al., 2016). It consisted of an indoor living wall aimed to treat 1 m<sup>3</sup> of GW per 343 344 day. An interesting outcome of the study deriving from the application of a Life Cycle Assessment was that 345 the environmental impact of such systems could be improved by using alternative materials, such as plastics 346 or wood, for the realization of the wall, and by employing sustainable energy sources, such as photovoltaic 347 energy. The effluent water quality was also analysed in terms of micropollutants (endocrine disrupting compounds and pharmaceutical active compounds), obtaining generally high removal efficiencies apart from 348 349 a few exceptions.

A different application was proposed by Eregno et al. (2017). The GW biofilter system was composed of a sequence of a primary settler, an unsaturated fixed-film biofilter, and a secondary clarifier, followed by an infiltration system (unvegetated filtration columns). The effluent from the filtration columns was then used Journal Pre-proofs plant growth. The focus of the study concerned the realibility of using treated greywater to cultivate food crop and the consecutive possibility of consuming lettuce irrigated with treated GW without health risks through a quantitative microbial risk assessment. The quality of the effluent from the infiltration columns was satisfactory in terms of nutrient and bacteria removal.

358 While the previous study provided a considerable amount of knowledge about the application of green walls and green roofs for GW treatment and reuse, a number of questions are still unsolved. The most relevant 359 issue that limits the application of these systems for GW treatment is the lack of sizing principles to predict 360 the amount of GW that can be fed and efficiently treated by a specific system. The indications can be 361 obtained from the existing studies will be discussed in section 5. Another important issue that will require 362 363 further research efforts is how the ensure durability of these systems. Problems that can mine durability of 364 these NBS span from clogging of porous medium to structural ageing, and long-term studies will be necessary to understand these issues and avoid loss of treatment efficiency over time. Finally, as stressed by 365 Pradhan et al. (2019), difficulties related to social acceptance of treated GW reuse should not be dismissed. 366

## 367 5 Hydraulic design criteria and greywater treatment performance

368 The optimization of the removal processes in NBS involves the selection of appropriate plant species and substrates as growing materials, the assessment of optimal hydraulic parameters, and the definition of 369 370 suitable operating conditions. While these issues have been widely addressed for constructed wetlands, much 371 less information are available for green walls and green roofs. Some works started to discuss and analyse the 372 selection criteria for defining the best combination of design parameters involved in the realization of 373 vegetated GW treatment systems (Castellar da Cunha, et al., 2018; Pradhan, et al., 2019). However, a 374 fundamental issue that has not been fully clarified yet is the influence of operational factors on removal 375 efficiencies. The identification of optimal design parameters for green walls and green roofs is essential to 376 ensure high pollutant removal and an efficient use of space. Hence, this section presents a critical summary of the previously discussed results about the relationship between the treatment capacity of green walls and 377 green roofs and HLR, with specific regard to the removal of organic matter, nutrients, and bacteria. From a 378 379 design perspective, high values of HLR are desirable because they result in NBS that can receive large GW amounts of with a low spatial footprint. However, high HLR values increase the velocity of filtration and 380

values of HLR are hence expected to reduce the removal of contaminants (and particularly those more proneto leaching).

Tables 10 and 11 summarize the design parameters, operational characteristics, and removal efficiencies of 384 385 the previously discussed case studies. Table 10 reports the main parameters of each study in terms of substrate, plants, and operational factors - flow, HLR, OLR, and HRT - distinguishing between pilot and 386 laboratory studies. All these parameters control the biogeochemical and physical processes occurring along 387 each pathway, affecting the treatment efficiency. For studies that did not report values of surface HLR in 388  $L/m^2/day$ , these values were calculated dividing the flow rate by the horizontal area of the system. Table 11 389 presents the treatment efficiency, indicating the effluent concentrations and the percentage of removal of the 390 considered contaminants. The ranges of removal efficiency reported in Table 11 refer to all the 391 configurations investigated in each study, also including configurations that showed a low performance and 392 were thus excluded as effective treatment systems. As shown in this table, the systems generally exhibited 393 good removal performances for organic matter (BOD<sub>5</sub> and COD), with removal rates that reach 90-99% and 394 outlet concentrations that fulfill the limitations for most non-potable water reuse (Ramprasad, et al., 2017; 395 Fowdar, et al., 2017; Svete, 2012). In some studies (Masi, et al., 2016; Prodanovic, et al., 2017; Prodanovic, 396 397 et al., 2018), a high variability in organic removal efficiency was observed.

The performance of the systems in removing microorganisms has been evaluted using different bacterial indicators, such as *E. coli*, total coliforms, and faecal coliforms, depending on the parameter analysed in each study (see Table 11). Removal efficiencies were quite variable, and sometimes they prevented achieving the limits for water reuse (Ramprasad, et al., 2017; Chowdhury & Abaya, 2018; Svete, 2012; Prodanovic, et al., 2017; Prodanovic, et al., 2018). A solution for this issue could be the addition of a disinfection unit in order to achieve the standard limits for reuse (Ramprasad, et al., 2017).

- 404
- 405 Table 10: Overview of main studies concerning greywater treatment through green walls and green roofs in terms of design 406 parameters
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410 *efficiency* 

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Figure 4 shows the removal efficiencies of BOD<sub>5</sub>, COD, TSS, TN, TP, and pathogens as a function of HLR. 412 413 It should be noticed that the values shown in Figure 4 differ from those reported in Table 11 because the latter represent the whole range of variation obtained in all experiments, while Figure 4 considers only the 414 best experimental configurations identified in each study in order to highlight the treatment capacity under 415 optimal conditions for each system. However, in those cases when various typologies of systems were tested, 416 the analysis included the results related to the different typologies in order to compare systems with different 417 418 characteristics and implemented according to different aims (Chowdhury & Abaya, 2018). In the study by Fowdar et al. (2017), the configurations with Carex appressa and Strelitzia Reginae showed high 419 performances for different water quality parameters and have been hence considered here. The configuration 420 with LECA and coir was chosen as reference in Masi et al. (2016). Regarding the studies conducted by 421 422 Prodanovic et al. (2017, 2018), we considered the results related to coconut coir, that has proved to have the highest potential to remove pollutants from greywater (Prodanovic, et al., 2017), and those related to the 1:3 423 424 to 2:1 mixes (perlite:coir 1:3, 1:2, 1:1, 2:1) (Prodanovic, et al., 2018). For all cases, transient operations 425 during the start-up phase or intermittent loadings were not considered because they are considered not stable 426 (Fowdar, et al., 2017; Ramprasad, et al., 2017).

For BOD<sub>5</sub> and COD, higher removal efficiencies were found when low HLR values were applied (Figures 427 428 4a-b). This behavior can be explained considering that organic matter is mostly removed by microbial 429 degradation, and an increase of HLR entails a reduction of HRT, resulting in less microbial removal of 430 organic pollutants. However, the relationship between removal efficiency and HLR in Figures 4a-b is 431 nonlinear, and increases in HLR are not necessarily detrimental for removal performances. For instance, the 432 high removal efficiency of organic matter observed for  $HLR = 670 \text{ L/m}^2/\text{day}$  (Svete, 2012) can be attributed 433 to high dosing frequency with low greywater volumes. The results in Figures 4a-b indicate that a high 434 treatment performance of organic matter can be obtained for values of HLR up to 600-800 L/m<sup>2</sup>/day.

The removal of TSS seems to be the least problematic since most of the systems showed very high efficiencies (Figure 4c). The relatively low removal efficiency (~50%) of TSS observed for HLR<100  $L/m^2/day$  (Fowdar, et al., 2017) with *Carex appressa* can be attributed to leaching of plant exudates as well

biofiltration studies in which high TSS removal was observed with *C. appressa* (Bratieres, et al., 2008) and further investigations are hence necessary. For HLR around 1000  $L/m^2/day$ , a drop in TSS removal efficiency was observed (Masi, et al., 2016).

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442 Concerning the removal of total nitrogen, Figure 4d apparently suggests that increasing HLR implies lower removal efficiency. However, this behavior is largely influenced by a couple datapoints (Svete, 2012; Masi, 443 et al., 2016) and deserves careful discussion; on one hand, it should be noticed that the value indicated for 444 HLR=1000 L/m<sup>2</sup>/day (Masi, et al., 2016) is likely underestimated because it is only related to removal of 445 446 Total Kjeldahl nitrogen (TN data were not collected), which implies that denitrification is neglected. Moreover, the low TN removal efficiency (~30%) at HLR=670 L/m<sup>2</sup>/day (Svete, 2012) can be attributed to 447 the absence of anaerobic conditions that hampered denitrification. On the other hand, Eregno et al. (2017) 448 found high treatment efficiency (~80%) with high HLR (600 L/m<sup>2</sup>/day). Despite the slight decrease in 449 efficiency with increasing HLR, it is concluded that high HLR values are not necessarily detrimental for TN 450 removal provided that the system configuration ensures proper conditions (i.e., anaerobic conditions and 451 carbon availability). As to total phosphorus, Figure 4e shows no clear trend between HLR and TP removal. 452 High removal efficiencies (>70%) were obtained up to 700 L/m<sup>2</sup>/day, suggesting that TP removal was 453 454 mainly governed by fast processes (e.g., sorption) and hence stressing the importance of the proper choice of 455 growing substrates.

456 Removal efficiency of microorganisms in Figure 4f does not exhibit any clear correlation between pathogen 457 removal and HLR. Efficiencies higher than 2 log units were found even at high HLR, suggesting that 458 filtration processes play an important role in removing pathogens.

The previous findings indicate that the best performances were found for low values of HLR, while efficiencies varied significantly for HLR larger than 500  $L/m^2/day$  according to the considered parameter. Specifically, BOD<sub>5</sub>, COD, and TSS showed more stable removal efficiencies, while TN and TP removal appear to be more dependent on the type of system. In fact, the removal of nitrogen is strictly dependent on the occurring of suitable conditions for denitrification, while the removal of phosphorus is strongly influenced by the specific type of growing medium. Analogously to what was established for HLR, it is possible to identify a minimum threshold value of retention time of the order of few hours in order to achieve

467 performances refer to values of organic loading rate up to  $100 \text{ g}_{\text{COD}}/\text{m}^2/\text{day}$ .

#### 468 6 Environmental assessment of greywater treatment processes

GW reuse has both positive and negative environmental impacts (Vuppaladadiyam, et al., 2018). The 469 470 acknowledged environmental positive issues are: facing water scarcity by recovering water resources, minimising sewage production, and reducing water supply costs. It was observed that GW reuse in urban 471 areas can decrease of about 30% water consumption in buildings (Memon, et al., 2007) and GW reuse in 472 473 multi-story buildings in Israel may save 150 Mm<sup>3</sup>/y of freshwater use (Oron, et al., 2014). Furthermore, the application of GW in soils and agriculture sector can enhance the availability of organic matter and nutrients 474 475 (Memon, et al., 2007). Drawbacks related to GW reuse concern microbial risks and the presence of metals and micropollutants, which represent a possible threat for human health (Turner, et al., 2016). Microbial 476 risks are due to the presence of pathogens from faecal contamination, skin, food preparation and mucus, such 477 478 as E. coli, Pseudomonas aeruginosa, Rotavirus, Legionella spp., Salmonella spp., Staphylococcus aureus 479 (Blanky, et al., 2015).

Table 12 summarises 30 Life Cycle Analyses and environmental assessment studies performed in the last 480 decades to evaluate pros and cons of GW reuse according to GW origins, treatment technology and reuse 481 482 purpose. Vertical subsurface flow constructed wetlands represent a valid mitigation tool for greenhouse gas 483 emissions, entailing a reduction of about 50% in emissions compared to traditional wastewater treatment plants (WWTP) (Pan, et al., 2011). This improvement for vertical flow CW is distributed among 484 transportation stage (up to ~19 kg CO<sub>2</sub>-eq/PE/y less), treatment stage (up to ~40 kg CO<sub>2</sub>-eq/PE/y less), and 485 486 sludge handling. A considerable reduction in energy consumption can be obtained by switching from 487 centralised WW reuse systems (1.224-1.914 kWh/m<sup>3</sup>) to decentralised WW and GW reuse systems (0.246-488  $0.970 \text{ kWh/m}^3$ ), with a reduction of up to ~80% (Chang et al. 2017). Another remarkable advantage of WW 489 and GW reuse systems compared to conventional WWTP is the reduction of the amount of treated WW 490 released into natural water bodies, with a consequent decrease of contaminant loads and concentrations 491 (Chang et al. 2017).

492 Several advantages can be obtained from GW reuse of different origin. For example, do Couto et al., (2013)
493 demonstrated that the quality of airport-produced GW is similar to household GW and can be easily treated

Journal Pre-proofs 494 other environmental benefits. Sgroi et al. (2018) discussed how different political, decisional, social, 495 economic, technological, and environmental factors should be considered for a sustainable water reuse 496 497 implementation, stating that a holistic approach is needed to evaluate GW reuse sustainability in a circular 498 economy prospective. From a general overview, what emerges is that even though results are sometimes sitespecific - particularly in terms of net economic benefits - most studies showed that GW treatment with 499 decentralised approaches are more beneficial than conventional treatments in terms of net energy 500 consumption and economic costs. 501

Finally, it is important to highlight that irrigation with raw GW can negatively affect soil hydraulic properties. In fact, GW-induced hydrophobicity resulted to be controlled by factors such as the nature of organic matter (i.e., biodegradability) and surface-active components that clog soil pores. GW-induced hydrophobicity of sand can be limited by using treated GW rather than raw GW (Maimon, et al., 2017). Therefore, treated GW should be preferred to direct reuse of untreated GW for irrigation purposes to maintain plant health.

## 7 Conclusions

Results from the reviewed GW treatment applications (constructed wetlands, green roofs, and green walls) suggest high removal performances, indicating the suitability of these systems in treating domestic GW. In particular, the reviewed data about green walls and green roofs showed that a high removal efficiency (~80%) in terms of organic matter can be obtained in systems with hydraulic loading rates up to 800  $L/m^2/day$ . Removal efficiency of total nitrogen was around 60-80% for HLR up to 500  $L/m^2/day$ , while removal of total phosphorus did not exhibit a correlation with HLR. Very high performances (often >90%) were obtained for removal of total suspended solids for HLR up to 700  $L/m^2/day$ . The mentioned data provide a broad indication that values of HLR up to 500  $L/m^2/day$  can be employed in green walls and green roofs irrigated with greywater without reducing the removal efficiency for the parameters considered in this analysis. Further experimental studies to better constrain these sizing criteria are definitely warranted.

The analysis of several Life Cycle Analysis studies performed in the last decades showed good results in terms of environmental and energetic benefits when integrated treatment systems using green structures are compared with traditional systems. Future efforts should be devolved to optimizing the treatment system and editing guidelines for the development of these systems. Finally, the implementation of trial systems has mostly been conducted for limited time periods (e.g., some months to 1-2 years) and the application of NBS for GW treatment needs hence to be further investigated in order to thoroughly understand the feasibility of these approaches over more realistic operating time (i.e., 15-20 years).

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Table 6. Conventional parameters to characterize domestic greywater

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

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Country	рН	TSS (mg/L)	BOD <sub>5</sub> (mg/L)	COD (mg/L)	TN (mg/L)	TP (mg/L)	TC (MPN/100mL)	FC (MPN/100mL)	E. coli (MPN/100mL)
Country	Min-Max (Avg)	Min-Max (Avg)	Min-Max (Avg)	Min-Max (Avg)	Min-Max (Avg)	Min-Max (Avg)	Min-Max (Avg)	Min-Max (Avg)	Min-Max (Avg)
Australiaª	-	(74)	(104)	-	(5.3)	(3)	-	-	-
Canada <sup>b</sup>	6.7-7.6	-	-	278-435	-	0.24-1.02	-	4.7E+04-8.3E+05	-
Egypt <sup>c</sup>	6.05-7.96 (7)	70-202 (116)	220-375 (298.6)	301-557 (388)	-	8.4-12.1 (10.54)	-	-	-
- 1							1.7E+08-1.4E+09	4.0E+03-5.7E+06	
France <sup>d</sup>	6.46-7.48 (7.28)	23-80 (59)	85-155 (110)	176-323 (253)	-	-	(4.9E+08)	(1.3E+06)	-
Germany <sup>e</sup>	(7.6)	-	(59)	(109)	(15.2)	(1.6)	-	(1.4E+05)	-
	5 00 0 00 (( 00)	100 414 (006 0)	07.001 (004.1)	207-1299		1.2 (2.2)	2.5E+06-4.9E+06	0-6.9E+06	
Ghana <sup>f</sup>	5.00-9.00 (6.89)	192-414 (296.8)	87-301 (204.1)	(643.8)		1-3 (2.3)	(3.7E+06)	(1.80E+06)	-
L. J. eghii	5 00 9 24 (7 4)	53.80-788.00	17.10-290.00	43.90-733.00	17.00-28.82	0.01.2.84		5.00+01-1.20+02	
India <sup>e,g,h,i,j</sup>	5.90-8.34 (7.4)	(337.2)	(244.2)	(705.4)	(17.8)	0.01-3.84	-	5.0E+01-1.2E+02	-
Indonesiak	(6.85)	(18.00)	(8.50)	(15.00)	-	-	-	-	-
Israel <sup>e</sup>	6.3-8.2	30-298	74-890	840-1340	10-34.3	1.9-48	-	3.5E+04-4.0E+06	(5.0E+04)
Japan <sup>e</sup>	-	-	-	(675)	(25.6)	(1.1)	-	-	8.5E3-1.2E4
Jordan <sup>e</sup>	6.4-9	23-845	36-1240	58-2263	6.44-61	0.69-51.58	250-1.0E+07	1.3E+01-3.0E+05	(2.0E+05)
						<i></i>		0-1.9E+06	
Malaysia <sup>m,n</sup>	6.5-7.2 (6.85)	19-175 (114.4)	1.1-309 (188.85)	16-1103 (328.9)	-	(4.5)	-	(2.9E+05)	0-6.7E+03 (1.1E+03)
Niger <sup>e</sup>	(6.9)	-	(106)	-	-	-	-	-	-
Norway <sup>o,p</sup>	(7.1)	(39)	(129)	(241)	(10.61)	(1.03)	(6.8E+06)	-	(4.9E+06)
Oman <sup>e</sup>	6.7-8.5 (7.5)	11-505	25-562	58-486	-	-	2.0E+02-3.5E+03	(2.0E+02)	(2.0E+02)
Pakistane	(6.2)	(155)	(56)	(146)	-	-	-	-	-
Palestineq	5.8-8.26 (7.8)	304-4952 (1290)	407-512 (470.6)	863-1240 (995)	111-322 (199)	5.8-15.16 (10.45)	-	-	-
Republic of	(7.4)	(2180)	(255)	-	-	-	-	-	-

Table 7. Physico-chemical features of domestic greywater in different countries. All values are reported as minimum-maximum (average), depending on available data.

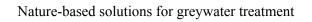
Korea<sup>e</sup>

Spain <sup>e,r,s</sup>	(7.39)	(336.09)	(130.32)	151-177	10.00-11.00			(1.0E+03)	(1.0E+03)
Spanity	(7.59)	(330.09)	(150.52)	(409.11)	(16.17)	-		(1.0±+05)	(1.02+03)
Sweden <sup>e</sup>	(7.8)	-	(425)	(890)	(75)	(4.2)	-	(1.7E+05)	-
Taiwan <sup>e</sup>	6.5-7.5	(29)	(23)	(55)	-	-	(5.1E+03)	-	-
Tunisia <sup>e</sup>	(7.5)	(33)	(97)	(102)	(8.1)	-		-	-
Turkey <sup>e</sup>	-	(54)	(91)	190-350	(7.6)	(7.2)	-	(1.1E+04)	-
UK <sup>e,t</sup>	6.6-7.8	37-153	8.7-155	33-587	4.6-10.4	0.4-0.9	1.8E+03-2.2E+07	1.0E+01-2.2E+05	1.0E+01-3.9E+05
United Arab				(1020)			(1.05+07)		
Emirates <sup>u</sup>	-	-	-	(1020)	-		(1.0E+07)	-	-
USA <sup>e</sup>	(6.4)	(17)	(86)	-	(13.5)	(4)	-	-	(5.4E+02)
Western Europe <sup>v</sup>	6.1-9.6 (7.5)	20-361 (89)	20-756 (221)	25-1583 (362)	3-75 (14)	0-11 (4)	-	-	-
Yemen <sup>e</sup>	(6)	(511)	(518)	(2000)		-	-	(1.9E+07)	-

<sup>a</sup> (Fowdar, et al., 2017) <sup>b</sup> (Finley, et al., 2009); <sup>c</sup> (Abdel-Shafy & Al-Sulaiman, 2014); <sup>d</sup> (Hourlier, et al., 2010); <sup>e</sup> (Ghaitidak & Yadav, 2013); <sup>f</sup> (Oteng-Peprah, et al., 2018); (Masi, et al., 2016); (Ramprasad, et al., 2017)<sup>b</sup>; <sup>i</sup> (Tilve, 2014); <sup>j</sup> (Vakil, et al., 2014); <sup>k</sup> (Wijaya & Soedjono, 2018); <sup>m</sup> (Leong, et al., 2018); <sup>n</sup> (Wurochekke, et al., 2014); <sup>o</sup> (Eregno, et al., 2017); <sup>p</sup> (Svete, 2012); <sup>q</sup> (Al-Atawneh & Mahmoud, 2014); <sup>r</sup> (Gattringer, et al., 2016); <sup>s</sup> (Rodríguez-Chueca, et al., 2014); <sup>t</sup> (Frazer-Williams, et al., 2008); <sup>u</sup> (Chowdhury & Abaya, 2018); <sup>v</sup> (Boutin & Eme, 2016).

				Regula	ations						Guidelines			
Parameter	Gre	at Britain	U	SA	NSW-	Australia	Italy	Japan	Germany	Slovenia	Jordan	China	India	Canada
	Sprinkler	Non-sprinkler	Residential	Commercial	Irrigation	Household								
	use	use												
pН	5–9.5	5–9.5	6–9	6–9	6	9	6–9.5	6–9	6-9	7-9	6-9	6-9	6-9	7-9
EC (µS/cm)	-	-	-	_	_	-	3000	-	-	_	10000	_	_	_
BOD <sub>5</sub> (mg/L)	_	-	<10	<10	<20	<20	20	<20	20		30-300	10-20	<30	200
COD (mg/L)	_	-	-	-	-	-	100	/-	-	200	100-500	<15	<250	280
TSS (mg/L)	_	_	_	_	30	10	10	-	Near free	80	50-150	10-50	<200	<100
Turbidity	<10	<10	<5	<2	_		_	Clear	Near clear	_	2-10	<10	_	<2
(NTU)														
Anionic														
surfactants	_	_	_	_	-	-	0.5	30	_	1	30-100	0.5-1	<10	_
(mg/L)							(total)							
Sodium (mg/L)	_	_	_	-		-	_	-	-	_	230	_	_	_
TN (mg/L)	_	_	_	-	_	_	15	20-30	_	10	50-70	15-20	_	_
TP (mg/L)	-	-	-		-	_	2	1-4	_	1	30	1-5	_	_
Boron (mg/L)	-	_	-	2	_	_	1	-	_	_	1	_	_	_
FIB	Not	25 (E.coli)	~0	~0	<1	<4	100	<1.105	<10 (total)	_	<1000	_	_	<1000
(MPN/100 mL)	detected						(E.coli)		<100 (faecal)		(faecal)			(total)
											100 (E.coli)			2-200

### Table 8. Overview of regulations and guidelines for GW reuse.



Country	Guideline	Main finding	Reference
Europe	Directive 91/271/EEC on urban waste water treatment (UWWTD)	<ul> <li>Wastewater shall be reused whenever appropriate</li> <li>Member States shall minimise any adverse effects on the environment from wastewater reuse</li> <li>The nutrient removal requirements concern Sensitive Areas (i.e. eutrophic/in danger fresh-water bodies, sources of drinking water, bathing waters, natural habitats, fish waters).</li> </ul>	Directive 91/271/EEC concerning urban waste water treatment (UWWTD)
United Nations	Guidelines for the safe use of wastewater, excreta and greywater	<ul><li>Chapter 2: Technical perspectives</li><li>Chapter 8: Environmental aspects</li></ul>	(WHO, 2006)
Australia	National guidelines for water recycling: managing health and environmental risks (phase 1)	<ul> <li>Chapter 2: Framework for management of recycled water quality and use</li> <li>Chapter 3.7: Managing health risks in recycled water, GW, microbial and chemical risks</li> </ul>	(EPHC, NRMMC, AHMC , 2006)
Australia	NSW guidelines for greywater reuse in sewered single household residential premises	GW reuse in urban area	(NSW-DEUS, 2007)
United Kingdom	Guidelines for greywater reuse: health issues	GW health risks	(Dixon, 2007)
Ireland	Rainwater harvesting and greywater treatment systems for domestic application in Ireland	<ul> <li>Coarse filtration + metal strainer to retain suspended particles</li> <li>Membrane filtration to clarify water</li> </ul>	(Li, et al., 2010)
North America	Overview of greywater reuse: the potential of greywater systems to aid sustainable water management.	<ul> <li>challenges and opportunities for GW reuse</li> <li>GW as percentage of total water use</li> <li>GW and energy</li> <li>GW and agriculture</li> </ul>	(Allen, et al., 2010)

Table 9. Proposed guidelines and reports for GW reuse (no standards or limits are available).

# Journal Pre-proofs

Israel	Greywater use in Israel and worldwide: standards and prospects	<ul> <li>Separated collection of GW from wastewater</li> <li>Biological treat is based on membrane + UV disinfection Reuse for 1 yard irrigation and/or toilet flushing</li> </ul>	(Oron, et al., 2014)
Europe	Technical guide for greywater recycling systems	<ul> <li>Treated GW shall only be used for the following applications:</li> <li>Flushing of water closet (WC)/Urinal.</li> <li>General washing (excluding high pressure jet washing and general washing at markets and food establishments)</li> <li>Irrigation (excluding irrigation sprinklers)</li> <li>Cooling tower makeup water.</li> </ul>	(PUB, 2014)
Australia	Code of practice – onsite wastewater management	<ul> <li>Chapter 3: Onsite wastewater management in unsewered areas</li> <li>Chapter 4.1: GW overview overview of GW policies, regulations, and laws around the world</li> </ul>	( EPA Victoria, 2016)
Europe	Guidelines on integrating water reuse into water planning and management in the context of the WFD (Water Framework Directive)	<ul> <li>Water scarcity</li> <li>Greywater reuse in agriculture</li> <li>Greywater health risk</li> </ul>	(EPA, 2016)
United Nations United Kingdom	United Nations: world water development report. BS8525 and the Water Supply (Water Fittings) Regulations	<ul> <li>Chapter 2: wastewater and the sustainable development agenda</li> <li>Chapter 4: technical aspects of wastewater</li> <li>Chapter 5: municipal and urban wastewater</li> <li>Chapter 7: agriculture as a user of wastewater</li> <li>GW can be reused on site for: <ul> <li>ornamental, garden and lawn irrigation,</li> <li>toilet flushing</li> </ul> </li> </ul>	(United Nations, 2017) BS8525 and the Water Supply (Water Fittings)

	Conventional/advar	nced process	GW origin	Treatment Efficiency (% removal)	Reference
				BOD <sub>5</sub> =85.37%, COD=63.59%	
	Coagulation	FeSO <sub>4</sub> (13%v/v)= 0.79 nM		NO <sub>3</sub> <sup>-</sup> N =8.96%, TN=0.56%, TP=96.39%	
			Synthetic GW		(Pidou, et al.,
			Showers + sinks	BOD <sub>5</sub> =88.28%, COD=63.72%	2008)
		$Al_2(SO_4)_3 (48\% v/v) = 0.89 \text{ nM}$		NO3 <sup>-</sup> -N =14.93%, TN=12.78%	
				TP=94.58%	
Chemical		Electrode combination =Al-Fe-Fe-Al,	Real GW		
Chennical	Electro-coagulation (EC)	pH <sub>i</sub> = 7.62,	showers, sinks and	COD= 90-95%	(Barışçı & Turkay, 2016)
		$CD = 1 \text{ mA/cm}^2$	kitchen		·· · · <b>, ·</b> · · · <b>,</b>
		pH = 7.0, Ozone = 47.4 mg/L			
	EC/O <sub>3</sub>	CD=15 mA/cm <sup>2</sup>		COD= 85%, TOC=70%	
			Real GW		(Barzegar, et al.,
			showers, sinks		2019)
	EC/O <sub>3</sub> /UV	pH = 7.0, Ozone = 47.4 mg/L		COD= 95%, TOC=87%, <i>E. coli</i> =96%	
		CD=15 mA/cm <sup>2</sup> , UV ray		COD 7570, 10C 0770, E. COU-7070	

 Table 10. Conventional technologies for GW treatment and reuse.

		ZnO/Zn photoanode			
		CuO/Cu photocathode		COD= 55%,	
		Illumination area = $6 \text{ cm}^2$	Real GW:	BOD <sub>5</sub> = 55%	(Kee, et al.,
	Photocatalytic fuel cell (PFC)	Photoelectrodes distance= 4 cm.	laundry	Turbidity=88%	2018)
		Light source =365 nm mercury UV lamp.		NH <sub>4</sub> <sup>+</sup> -N=75%	
		Air bubbling (2 L/min) supplied at the			
		photocathode.			
		Recycled vertical flow bioreactor.			
	Filtration	Layer1: 2-cm thick crushed dolomite+		TSS = 93.48%, NH <sub>4</sub> <sup>+</sup> -N= 16.67%	
		limestone, average diameter 2.5 cm	Synthetic GW:	Anionic surfactant= 98.37%,	(Gross, et al.,
		Layer 2: 12 cm of plastic filter media	laundry + bath +	NO <sub>2</sub> -N =96.92%, NO <sub>3</sub> -N = 48.57%	2007b)
		with a high surface area ( $800 \text{ m}^2/\text{m}^3$ ) and	kitchen	TP = 73.68%, <i>E.coli</i> = 97.5%	
		large void volume			
	Filtration:			pH from 7.8 to 6.1, BOD <sub>5</sub> = 98%	
Physical	Filters: grain size distribution 0.8 -	Pine bark (bark)	Synthetic GW	EC= 7.14%	(Ghaitidak &
	6.3 mm; $D_{10}$ = 1.4 mm, $D_{60}$ = 3.1		Bath + laundry		Yadav, 2013)
	mm; uniformity coefficient = $2.2$ .	Activated charcoal (charcoal)		BOD <sub>5</sub> =97%, EC= 11.7%	
	Filtration:		Real GW shower +	TS= 14.21%, COD=20%	(Ghaitidak &
		Coarse	washing machine	,	
	HRT = 24h		from household	NH <sub>4</sub> <sup>+</sup> -N=17.74%, <i>E. coli</i> =98.31%	Yadav, 2013)

TSS= 82.61%, TDS=69.98% Layer1: coconut shell cover 20 cm, Synthetic GW COD=82.26%, Oil-grease=96.97% Filtration: Layer2: saw dust (coarse) 15 cm, (Parjane & Sane, 2011)  $NH_4^+-N=73.42\%$ ,  $NO_3^--N=68.66\%$ , HRT = 24hkitchen Layer3: charcoal 20 cm, bricks 10 cm and sand 15 cm.  $PO_4^{3}-P=100\%$ HRT=30-40 d Turbidity=20% Sedimentation tank+ sand filter COD=25% sand filter = 5 cm plexiglass column Synthetic GW (Kee, et al., HRT=30-40 d 2018) kitchen Turbidity=25% Sedimentation tank+ granular active Flowrate =2.8 L/hcarbon (GAC) COD=27% Filtering velocity= 1.4 m/h. Electrochemical reactor B boron-doped diamond) Synthetic GW: COD=68% Hybrid granular active carbon (GAC)-Physico-(Andrés García, anode=80×80×1,3 mm, 3D reactor chemical electrochemical (EC) system et al., 2018) a packed bed of GAC was placed in shower TOC=70% between anode and cathode. Anodic  $CD = 15 \text{ A/m}^2$ 

SAR (sodium adsorption rate) = 4.9%

				pH value increased with HRT	
	Rotating biological contactor (RBC)	RBC was made of plastic sheets with tank volume of 54 L	Real: laundry + bath + kitchen	TDS not changed TSS= 8.98-11.08% BOD <sub>5</sub> = 27.30-52.42% COD= 21.48-60.36%	(Xiao, et al., 2018)
	Moving bed biofilm reactor (MBBR)	Q <sub>max</sub> = 31.2 L/h	Synthetic GW:	Turbidity= 66%, TSS=87.07	(Chrispim &
Biological		Q <sub>max</sub> air= 110 L/min	laundry + bath	TP=12%, BOD <sub>5</sub> =59% COD=70%	Nolasco, 2017
		Membrane area = $120 \text{ cm}^2$			
		Pore size membrane=0.4 um			
	Membrane bioreactor (MBR)	High container= 1.8 L	Real: laundry +	Turbidity= 98%	(Hu, et al.,
		Inoculum= activated sludge from	bath + kitchen	BOD <sub>5</sub> =93.37%	2011)
		wastewater treatment plant HRT=2.5-10 h			

			Turbidity= 98.28%, NH <sub>4</sub> +-N=72.03%,	
	Reactor capacity= 3 L	Real GW	TN =62.05%, TP=18.75%, Anionic	(Merz, et al.,
Membrane bioreactor (MBR)	UF membrane	Shower	surfactant= 96.66%	2007)
			COD= 86.24%, BOD <sub>5</sub> =93.22%	
		<u> </u>		(TTT 1 1
SBR	SBR volume tank $=$ 2 L	Synthetic GW	COD =90%	(Weitao, et al.
		Shower		2006)
		Real GW	COD=51%, N= 37%,	(Hernández
Up-flow anaerobic sludge blanket	UASB=5 L	Sharray (12	000 5170,10 5770,	Leal, et al.,
(UASB)		Shower (12 houses)	NH <sub>4</sub> <sup>+</sup> -N=47%, P=50%	2011)
	HRT=100 d			
<b></b>		Real GW:		
Biological activated membrane	Q=75 L/day nutrient supplement	Detherson on d	C =95%	(Ziemba, et al
bioreactor (BAMB)	10 mgC/L TOC to avoid inhibition from	Bathroom and washbasin		2018)
	surfactant.			

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Table 6. Range of inflow characteristics of constructed wetland treating greywater (GW-CWs) and constructed wetlands treating lightly

Parameter	Inflow (mg/l)						
_	GW-	·CW	S	LI	L-CV	Ws	
TSS	12.3	-	158.0	2.1	-	189.0	
BOD <sub>5</sub>	20.0	-	466.0	1.4	-	220.0	
COD	87.0	-	839.0	5.9	-	385.0	
TN	5	-	34.3	8.9	-	17.9	
ТР	0.83	-	22.8	0.64	-	11.0	
TC*	2.5E+05	-	4.0E+07	2.3E+04	-	1.0E+06	
Flow**	0.005	-	7.9	0.001	-	172.8	

loaded wastewater (LL-CWs).

\*MPN/100 ml; \*\* m<sup>3</sup>/d

Reference	CW tech	Plants	Filling media	GW source	Areal	Flow	HLR	HRT	OLR	
					footprint (m <sup>2</sup> )	(m <sup>3</sup> /day)	(L/m <sup>2</sup> /day)	(days)	$(g_{COD}/m^2/day)$	
(Jokerst, et al.,		Typha latifolia								
	FWS, HSSF	(FWS) Scirpus	Gravel (HSSF)	Bathroom sink, shower	10-17	0.29	17-29	9.3-12	-	
2011)		acutus (HSSF)								
			PET (100-150 mm)			0.005				
(Dallas & Ho,	HSSF	Coix lacryma-jobi	or crushed rock (20	Greywater, nonspecific	0.38	0.005-	13-27	2.5-7.2	-	
2005)		mm)			0.01					
(Winward, et al.,										
2008)	VF, HSSF	Phragmites australis	Sand/soil/compost	Bathroom sink, shower	6	0.48	80	2.1	7-40	
			Wood- chips, tuff							
(Alfiya, et al.,	RVF	No information	gravel, limestone	Greywater, nonspecific	1	0.11-	112-157	4.1-5.8	251-933	
2013)			pebbles.			0.16				
			Wood- chips, tuff							
(Blanky, et al.,	RVF	No information	gravel, limestone	Bathroom sink, bath,	1	0.11	110	6.1	-	
2015)			pebbles.	shower, washing machine			110			
			Organic soil, tuff or							
(Gross, et al.,	RVF	No information	plastic media,	Laundry, showers and	1	0.45	45	1.3	3775	
(007a)	1. 7 1		•	sink	1	0.75	υ	1.5	5775	
			limestone pebbles.							

Table 7. Pilot scale CW for GW treatment and reuse from peer-review literature (FWS = free water surface; VF = vertical flow; HSSF = horizontal subsurface flow; RVF = recycled

vertical flow bioreactor; SF = sand filter).

(Yu, et al., 2015 )	RVF	Carex spissa, Phyla nodiflora, household plants (Aeonium purpureum and Crassula ovate, Equisetum hyemale, Nasturtium,	Coconut coir, cross- flow media	Bathroom sink, shower, laundry	0.77	0.086	111	3.5	142-545
		Narcissus impatiens, Anigozanthos, Colocasia)							
(Gerba, et al., 1995)	FWS+SF	Water hyacinth (FWS), tomatoes, peppers (SF)	Sand (SF)	Washing machine, clean half of kitchen sink, bathroom sink, tub, shower	7.2	0.41	56	6	-
(Abdel-Shafy, et al., 2009)	HSSF	Phragmites australis	Sand (0.5–1.0 mm)	Secondary treated greywater (UASB)	1.1	-	-	5	-
(Jenssen & Vråle, 2003)	HSSF	Phragmites australis	Light weight aggregates	Secondary GW from aerobic biofilter	-	-	-	6-7	-
		300							

CW	Location	GW source	Areal	Treated	HLR	Designed effluent water quality	Operating	Additional	Type of GW	Reference
technology	& Year of		footprint	flow rate	$(L/m^2/d)$	or measured removal	time	disinfection	reuse	
	construction		(m <sup>2</sup> )	(m <sup>3</sup> /d)		efficiencies		units		
HF	"La cava"		241 m <sup>2</sup>	80 PE	110-230	Measured removal efficiencies:	~ 1.5 years		Irrigation	(Masi, et al., 2007;
	camping, Arezzo					COD 81%				Scheumann, et al.,
	(Italy)					BOD <sub>5</sub> 92%				2009)
	2003					NH4 <sup>+</sup> 62%				
						FC 97%				
HF+VF	Campo Grande	Shower,	4.6+2.3	0.5-2.5	150	Measured removal efficiencies:	$\sim 5$ months	None	Not reported	(Paulo, et al., 2009)
	(Brazil)	kitchen sink,				TSS 92%±12, COD 88%±11,				
	2007	laundry				BOD <sub>5</sub> 95%±4, TP 58%±35, TN				
						82%±20, <i>E. coli</i> 93±12				
HF	Rabat (Morocco)		4.5	8	880	Measured removal efficiencies:	~ 4 years	Sand filter (4.5	Toilet flushing	(Masi, et al., 2010)
	2005					COD 75%		$m^2$ ) + UV		
						BOD <sub>5</sub> 80%				
						TP 50%				
						Anionic surfactants 97%				
						FC < 1log				
HF	Cairo (Egypt)		12	1	83	COD 82-91%	~ 2 years	None	Toilet flushing,	(Masi, et al., 2010)
	2007					TSS 85%	-		irrigation	

### UV lamp Istanbul (Turkey) 110 TSS < 10 mg/l $\sim 2$ years Toilet flushing, (Masi, et al., 2010) HF+FWS 28+35 1 2007 COD < 50 mg/llandscaping irrigation $BOD_5 < 10 \text{ mg/l}$ $NO_{3} < 1 mg/l$ $NH_4{}^+\,{<}\,0.5~mg/l$ HF 1.2 96 $BOD_{5} \ 3.45 \pm 3.2$ Marrakech Hand wash 12.5 $\sim 3.5 \text{ months}$ None Irrigation of (Laaffat, et al., (Morocco) COD 11.43 ± 4.5 landscape green 2015) sinks SS 0.29±0.2 2012 areas TN 3.89±1.5 TP 0.47±0.3 E. coli 5x10<sup>1</sup>

NH4<sup>+</sup> 30-57%

CW technology	Location	Year of	GW source	Areal footprint	Treated flow rate	Additional	Type of GW reuse	Reference
		construction		(m <sup>2</sup> )	(m <sup>3</sup> /d)	disinfection units	6	
VF single stage	Polderdrift, Arnhem	1996	All	230 m <sup>2</sup>	6.4 m <sup>3</sup> /d	None	Toilets flushing	www.globalwettech.com
(sand filled)	(Netherlands)							-
Forced Bed	National Great Rivers		Washbasins,	47 m <sup>2</sup>	5.3 m <sup>3</sup> /d	UV	Toilet flushing,	www.globalwettech.com
Aeration <sup>™</sup>	Education and		showers, kitchens				landscape irrigation	
(Aerated CW)	Research Centre, Alton,							
	Illinois (US)							
	W . D . I . I					N.	* ·	
VF	West Bank bedouin	2012	All	30-60 m <sup>2</sup>	70-120 PE	None	Irrigation of olive	www.globalwettech.com
	villages (Palestine)						camps or folder	
							production	
HF	Group of houses in	2009	All	230 m <sup>2</sup>	14.5 m <sup>3</sup> /d	None	Toilets flushing	www.globalwettech.com
	Preganziol (Italy)							
HF	Group of houses in	2008	Showers, Hand	50 m <sup>2</sup>	30 PE	UV	Toilet flushing,	www.globalwettech.com
	Vicchio (Italy)		wash sinks,				landscape irrigation	
			laundry					
VF	Hostel campus of	2015	All	130 m <sup>2</sup>	40 m <sup>3</sup> /d	Chlorine	Toilet flushing,	(Patil, et al., 2016)
	College of Engineering,						landscape irrigation	
	Pune (India)							
roof HF	Resort in Grumenti		All	23 m <sup>2</sup>	4 m <sup>3</sup> /d	UV	Irrigation	www.globalwettech.com

### Table 9. A list of full-scale CW for GW treatment and reuse from Global Wetland and Technology Database.

Nature-based solutions for greywater treatment

	community, Serengeti	2015						
	(Tanzania)							
VF	Private school, Lima	2008	Kitchen	16 m <sup>2</sup>	1.5 m <sup>3</sup> /d	None	Landscape irrigation	(Platzer, et al., 2016)
	(Peru)							(Hoffman, et al., 2009)
VF	Guest house of 160	2011	Restaurants and	100 m <sup>2</sup>	12 m <sup>3</sup> /d	Chlorine	Toilet flushing,	(Platzer, et al., 2016)
	apartments, Lima		showers				landscape irrigation	
	(Peru)							
VF	Housing project for 288	2011	Showers	60 m <sup>2</sup>	9 m³/d	None	Landscape irrigation	(Platzer, et al., 2016)
	families, Lima (Peru)							
SuSaNa Database								
VF	Waagner head office	2006	Showers,	500 m <sup>2</sup>	270 staff	None	Concrete mixing,	(Sievert & Schilick, 2009)
	(Dubai)		Washbasins, car		members		soil watering, car	
			washing				washing, fish ponds	
VF	SAMA Dubai site	2006	All	40 m <sup>2</sup>	200 staff	None	Manual garden	(Sievert & Schilick, 2009)
	office (Dubai)				members		irrigation and car	
							washing	
VF	Dubai Municipality	2006	All	170 m <sup>2</sup>	60 staff members	UV (optional)	Drip irrigation and	(Sievert & Schilick, 2009)
	(Dubai)						boat washing	
HF	Secondary School,	2008	Kitchen	2 m <sup>2</sup>	1 m <sup>3</sup> /d	none	Agro-forestry	(Machiri, et al., 2009)
	Nakuru (Kenya)							

	T C					Operation	al factors	
Authors	Type of study	Plants	Size	Substrate	Flow (L/day)	HLR (L/m²/day)	OLR (g <sub>COD</sub> /m <sup>2</sup> /day)	HRT (hours)
(Frazer- Williams, et al., 2008)	pilot	Iris pseudocorus, Saururus cernuus, Glyceria variegates, Juncus effusus, Iris versicolor, Caltha palustris, Lobelia cardinalis, Menta aquatica	6.86 m <sup>2</sup>	38% light-weight expanded clay and 62% gravel chippings	480 (continuous flow)	70	5.7	50.4
(Svete, 2012)	pilot	Lettuce, marigolds	2.34 m <sup>2</sup> vertically	Lightweight expanded clay aggregates	360	670-980	161.5	1.5-3.3
(Gattringer, et al., 2016)	pilot	Different species of marshplants (e.g. <i>Typha</i> , <i>Iris</i> ), graminoids ( <i>Carex</i> , <i>Cyperus</i> ) tropical and subtropical plants (e.g. <i>Ficus, Spathiphyllum</i> , <i>Epiprenum</i> )	2 m <sup>3</sup> substrate volume, 5 m in length, 1.50 m in width and 2.50 m in height	Expanded clay	500-1500 L/day (semi- continuous batch)	250	100	-
(Masi, et al., 2016)	pilot	Abelia, Wedelia, Portulaca, Alternanthera, Duranta, Hemigraphis	0.72 m <sup>2</sup>	50% light expanded clay mixed with 50% coco coir or sand	240 (an hourly flush of 10 L)	1000	60	0.2-0.67
(Eregno, et al., 2017)	pilot	No plants	0.078 m <sup>2</sup>	88% filtralite and 12% granular activated carbon	45	580	-	-
(Fowdar, et al., 2017)	laboratory	Strelitzia nicolai, Phormium spp. Canna lilies, Strelitzia reginae, Lonicera japonica, Carex appressa, Phragmites	70 columns, 0.05 m <sup>2</sup> each	68% sand mixed with 5% of cedar mulch, 16% coarse sand,	2.50 -5	55-110	5.7-15.4	96-48

Table 10. Overview of main studies concerning greywater treatment through green walls and green roofs in terms of design parameters.

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		australis, Vitis vinifera, Parthenocissus tricuspidata, Pandorea jasminoides, Billardiera scandens	column	16% gravel 100% coir or rockwool, or				
(Prodanovic, et al., 2017)	laboratory	No plants	0.00785 m²	phyto-foam, or perlite, or vermiculite, or growstone, or expanded clay, or river sand	3	382	95.5	0.25-2.5
(Ramprasad, et al., 2017)	pilot	Canna indica, Canna flaccida, Canna lily-hybrid, Cardamine pratensis, Plectranthus amboinicus, Crossandrain fundibu- liformis, Phragmites australis, Solanum trilobatum	1.84 m <sup>3</sup> , 15 cm depth	Mixture of sand, brick bats and gravel (1:1:1)	62, 70, 82, 100, 120	7.75 -15	1.62-4.8	16.8- 31.2
(Chowdhury & Abaya, 2018)	pilot	Phalaris arundinacea L.	2.64 m <sup>2</sup> (4module, 0.66 m <sup>2</sup> each)	Sand	6.0-7.0	-	10.2	<<24
(Prodanovic, et al., 2018)	laboratory	No plants	0.00785 m <sup>2</sup>	Coconut fibre and perlite (C:P 1:4, 1:3, 1:2, 1:1, 2:1, 3:1)	3	382	48.1-122.2	-

	Type of	Organic re	moval	N rem	oval	P re	emoval	Bacteria r	emoval
Authors	GW	Out (mg/L)	Rem.%	Out (mg/L)	Rem.%	Out (mg/L)	Rem.%	Out (MPN/100 mL)	Rem.%-log units
(Frazer- Williams, et al., 2008)	real	BOD <sub>5</sub> : 0-7.5, COD: 3-120	BOD <sub>5</sub> : 92, COD: 81.7	-	-	-	-	<i>E. coli</i> : < 10 <sup>4</sup> , FC: <10 <sup>4</sup> TC: <10 <sup>4</sup>	<i>E. coli</i> : 2.9 log, FC: 2.2 log, TC: 3.8 log (>99%)
(Svete, 2012)	real	BOD <sub>5</sub> : 2–6, COD: 29-43	BOD <sub>5</sub> : 95- 98, COD: 82-88	TN: 8.4- 8.8	TN: 31- 34	TP:0.26- 0.36	TP: 69-77	<i>E. coli</i> : >2·10 <sup>4</sup>	<i>E. coli</i> : 98- 99%, 2log
(Gattringer, et al., 2016)	real	BOD <sub>5</sub> : 3.81, COD: 13.89	BOD <sub>5</sub> : 97.1, COD: 96.6	TN: 4.21, NO <sub>3</sub> -N: 3.61, NH <sub>4</sub> - N: 0.17	TN: 74, NO <sub>3</sub> -N: - 440.3, NH <sub>4</sub> -N: 96.6	PO <sub>4</sub> -P: 0.32	PO <sub>4</sub> -P: 74.6	-	-
(Masi, et al., 2016)	real	BOD <sub>5</sub> : 6.7–29.9, COD: 21.4-69.4	BOD5: 8– 86, COD: 7–86		-	-	-	-	-
(Eregno, et al., 2017)	real		-	TN:1.53	TN: 82	TP: 0.08	TP: 91	-	<i>E. coli</i> : 5.1 log, TC: 3.7 log
(Fowdar, et al., 2017)	synthetic	BOD <sub>5</sub> : <3.5	BOD <sub>5</sub> : >97	TN: 0.40- 4.90	TN: 7– 92	TP: 0.46- 2.80, FRP: 0.28-2-28	TP: -13-99, FRP: -10.87	-	-
(Prodanovic, et al., 2017)	synthetic	-	COD: 22- 70	-	TN: 30- 75	-	TP: 10-60	-	<i>E. coli</i> : 60- 100%
(Ramprasad, et al., 2017)	real	BOD <sub>5</sub> : <10, COD: <16-24	BOD <sub>5</sub> : 84- 90, COD: 84-92	TN: 1.5- 3.8	TN: 82- 99	TP: 0.8- 1.4	TP: 65-92	FC: 4-12	FC:92-96%, 2-3 log

### Table 11. Overview of main studies concerning greywater treatment through green walls and green roofs in terms of removal efficiency.

## Journal Pre-proofs

(Chowdhury			COD: 79-						TC: 99% - 3
& Abaya,	real	COD: 41-405		-	-	-	-	TC: 10 <sup>4</sup> -10 <sup>7</sup>	_
2018)			90						log
(Prodanovic,	aunthatia	COD: 50-170	COD: 20-	TN: 1.1-	TN: 15-	TD: 1.5.4	TP: -156-42	<i>E. coli</i> : $0.2 \cdot 10^3$	E. coli: 32-
et al., 2018)	synthetic	COD: 50-170	85	4.8	80	TP: 1.5-4	+ TP: -156-42	- 2.4·10 <sup>3</sup>	90%

TN: Total Nitrogen, TP: Total Phosphorus, FRP: filterable reactive phosphorus, FC: Faecal Coliforms, TC: Total Coliforms

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Objective	Method	Results	Reference
Changzhou, China, evaluation of CO <sub>2</sub> , CH <sub>4</sub> , N <sub>2</sub> O emissions from a vertical subsurface flow <b>constructed</b> <b>wetland</b> (VF CW, 1000 m <sup>2</sup> ) and conventional wastewater treatment plants (WWTPs)	LCA cradle to grave	<ul> <li>WWTP: 7.3 kg CO<sub>2</sub>-eq to remove 1 kg BOD<sub>5</sub></li> <li>VF CW: 3.18 kg CO<sub>2</sub>-eq to remove 1 kg BOD<sub>5</sub></li> <li>VF CW may reduce GHG emissions by 8– 17 million tons CO<sub>2</sub>-eq per year compared to WWTP.</li> </ul>	(Pan, et al., 2011)
Brazil: evaluation of <b>GW</b> <b>reuse</b> in airport complexes	Descriptive and multivariate statistics	Quality of GW produced in the airport is similar to GW produced in residences GW produced in the airport meet the non- potable reuse + water savings (40%) + minimisation of financial resources (-25%).	(do Couto, et al., 2013)
Evaluation of low environmental impact GW	Experimental evaluation of GW on lettuce and radish	<ul> <li>- GW impact is the GW treatment on soil</li> <li>phosphatase activity</li> <li>- GW benefit is the worm avoidance.</li> </ul>	(Reichman & Wightwick, 2013)
Quantitative Microbial Risk Assessment (QMRA) was performed for Legionella in (light) <b>LGW</b> .	Risk analysis with two         approaches:         –       Inhalation of         contaminated aerosols         generated by sprinkler         irrigation with LGW         during gardening and         recreational activity.         –       Inhalation of         contaminated aerosols         generated by sprinkler         irrigation with LGW         during gardening and         recreational activity.         –         Inhalation of         contaminated aerosols         generated by toilet         flushing using LGW.	<ul> <li>QMRA for treated and chlorinated GW was not significantly higher than the for potable water</li> <li>Health risk stemming from treated GW is acceptable regarding Legionella infection.</li> </ul>	(Blanky, et al., 2015)
Evaluation of presence and health risks of organic micro-pollutants in GW	Literature survey of 280 organic micro-pollutants detected in GW grouped on the basis of: 1) toxicology Tier 1	<ul> <li>Risk quotient &lt; 0.2, which means not</li> <li>appreciable danger for human health over a</li> <li>lifetime exposure to potable water</li> <li>14 compounds have risk quotient &gt;risk</li> </ul>	(Etchepare & Van der Hoek, 2015)

	Journal P	re-proofs	
	standards	further investigation if GW is used for potable reuse.	
Morelia in Mexico evaluation pros-cons water reuse in urban area Sakharale, District Sangli, India: GW reuse coming out from hotel	LCA cradle to grave Multi-objective optimization: total annualized cost, fresh water consumption, and environmental impact Evaluation of integrated on-site GW Treatment system (IOGTS) – Comparison of 4 GW irrigated residential lots with	Best Scenario: simultaneous GW recycling and reusing and rainwater harvesting optimal solution for: Total Cost =585.57 10 <sup>3</sup> €/y, freshwater consumption = -13% compared to conventional treatments, complex environmental impact benefit = +32% compared to conventional treatments IOGTS satisfy GW standards for reuse in land application in India. - GW increased concentrations of As, B, Cr and Cu exceeding guidelines after only 4 y	(García- Montoya, et al., 2016) (Patil, et al., 2016)
Evaluation of irrigation impacts terrestrial and aquatic environments using GW	<ul> <li>4 adjacent non-irrigated lots (controls)</li> <li>– Evaluation of metals accumulation in soil, groundwater and surface water comparing measured concentrations to national and international guidelines</li> </ul>	<ul> <li>of irrigation.</li> <li>Movement of metals from the irrigation areas resulted in: Al, As, Cr, Cu, Fe, Mn, Ni and Zn concentrations in groundwater and Cu, Fe and Zn surface water exceeding environmental quality guidelines after 4 y of irrigation</li> </ul>	(Turner, et al., 2016)
South Korea: evaluation of energy consumption and GHG emissions of conventional water treatments and <b>water reuse</b>	<ul> <li>Scenario1: Conventional water treatment</li> <li>Scenario2: Centralised wastewater reuse</li> <li>Scenario3: Decentralised wastewater and GW reuses</li> </ul>	<ul> <li>Scenario1: energy consumption=0.511 kWh/m<sup>3</sup> and GHG emissions=0.43 kg CO<sub>2</sub>-eq/m<sup>3</sup></li> <li>Scenario2 energy consumption=1.224– 1.914 kWh/m<sup>3</sup> and GHG emissions=0.72-0.83 kg CO<sub>2</sub>-eq/m<sup>3</sup></li> <li>Scenario3: energy consumptions 0.246– 0.970 kWh/m<sup>3</sup> and GHG emissions=0– 0.33 kg CO<sub>2</sub>-eq/m<sup>3</sup></li> </ul>	(Chang, et al., 2017)
Evaluation of <b>GW reuse</b> for irrigation	• Experiments in Lab on quartz sand	<ul><li>GW reuse reduces soil wettability.</li><li>Wettability of sand wetted with raw GW</li></ul>	(Maimon, et al., 2017)

Falmouth, MA town, USA: environmental and economic evaluation of water sanitation and <b>water reuse</b>	<ul> <li>LCA cradle to grave</li> <li>Scenario 1: Centralized water and sewage</li> <li>Scenario 2: Centralized water and composing toilet + on- site GW reuse</li> <li>Scenario 3: Centralized water and urine- diversion toilet with septic</li> <li>Scenario 4: Centralized water and digester with GW non-</li> </ul>	<ul> <li>Washing and biodegradation can reduce GW-induced hydrophobicity of sand.</li> <li>GW has to be treated before reuse for irrigation.</li> <li>Scenario 4 offered the best configuration:         <ul> <li>Local Human Health impact= 1*10<sup>-1</sup> DAILY</li> <li>Equivalent annual cost = 888 €/y/household</li> <li>Eutrophication potential= 6.1 × 10<sup>-2</sup> kg N/day/household</li> <li>GWP= 1 kg CO<sub>2</sub>-eq/day/household</li> <li>Energy use= 0.88-1.00 MJ/day/household</li> </ul> </li> </ul>	(Schoen, et al., 2017)
	potable reuse		
Comparison of three <b>GW</b> <b>treatments:</b> • photocatalysis • photovoltaic solar-driven photocatalysis • membrane biological reactor	LCA cradle to gate Impact categories: - atmospheric acidification - global warming - human health - effects photochemical ozone formation stratospheric ozone depletion)	Highest energy saving and treatment performances (+50%) achieved with photovoltaic solar-driven photocatalysis	(Dominguez, et al., 2018)
Evaluation of energy consumption of <b>GW</b> photocatalytic fuel cell (PFC) with ZnO/Zn photoanode CuO/Cu photocathode	<ul> <li>X-ray diffraction (XRD),</li> <li>field-emission scanning electron microscopy (FESEM)</li> <li>energy dispersive X-ray (EDX)</li> <li>Fourier transform infrared (FTIR) spectroscopy</li> </ul>	PFC with ZnO/Zn photoanode and CuO/Cu photocathode: effective GW as well energy recovery	(Kee, et al., 2018)

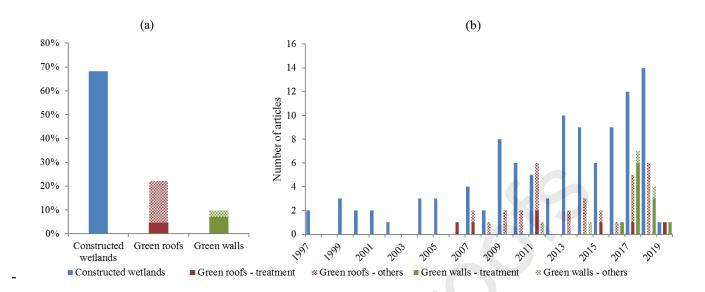
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comparison of conventional water treatment, desalination and <b>GW reuse</b>	Existing academic and grey literatures on greywater in Qatar	GW can replace more expensive conventional water treatments and desalinated resources	(Lambert & Lee, 2018)	
Evaluation of on-site separation of black water from <b>GW and onsite reuse</b>	Critical analysis of risks assessment and scientific papers	<ul> <li>GW use for toilet flushing and irrigation may benefit users for nutrients and the water saving</li> <li>GW might pose health and environmental risks, which have to be quantified and standardised</li> <li>Reuse of GW should consider local conditions and intended usage.</li> <li>Technology and regulations should be routinely audited to mitigate potential risks</li> </ul>	(Maimon & Gross, 2018)	
Evaluation of mitigate water scarcity in urban areas by means of decentralised rainwater harvesting, <b>GW</b> recycling, and hybrid rainwater-GW systems	Rain-TANK model	<ul> <li>Domestic and commercial rainwater systems supply &gt;90% and &lt;43% of non- potable demand (NPD).</li> <li>Domestic and commercial GW supply &gt;92.1% and &gt;36.2% of NPD.</li> <li>Hybrid systems produce higher water savings than rainwater or GW alone &gt;95% of NPD</li> </ul>	(Oh, et al., 2018)	
Economic and environmental evaluation of <b>GW</b> according to boundary system and functional unit	<ul> <li>Critical analysis of papers from 1990s-2016</li> <li>LCA cradle to grave/cradle- to-gate/</li> <li>LCA m<sup>3</sup> influent/m<sup>3</sup> effluent, tons of dry solids</li> </ul>	<ul> <li>LCA was used to determine the impact of wastewater treatment and the technologies used.</li> <li>GW reclamation and reuse positive contributes to environmental benefits, while from economic perspectives depends on the adopted technology of treatment</li> </ul>	(Sabeen, et al., 2018)	
Political + Economic+ Social+ Environmental GW reuse in Circular economy perspectives	<ul> <li>Energy recovery evaluation</li> <li>Economic plan</li> <li>Water supply</li> </ul>	<ul> <li>Economic perspective is the highest barrier to actual development of GW reuse.</li> <li>Holistic Approach: LCA+LCC+LCSC is needed to evaluate GW reuse sustainability.</li> </ul>	(Sgroi, et al., 2018)	

### Journal Pre-proof

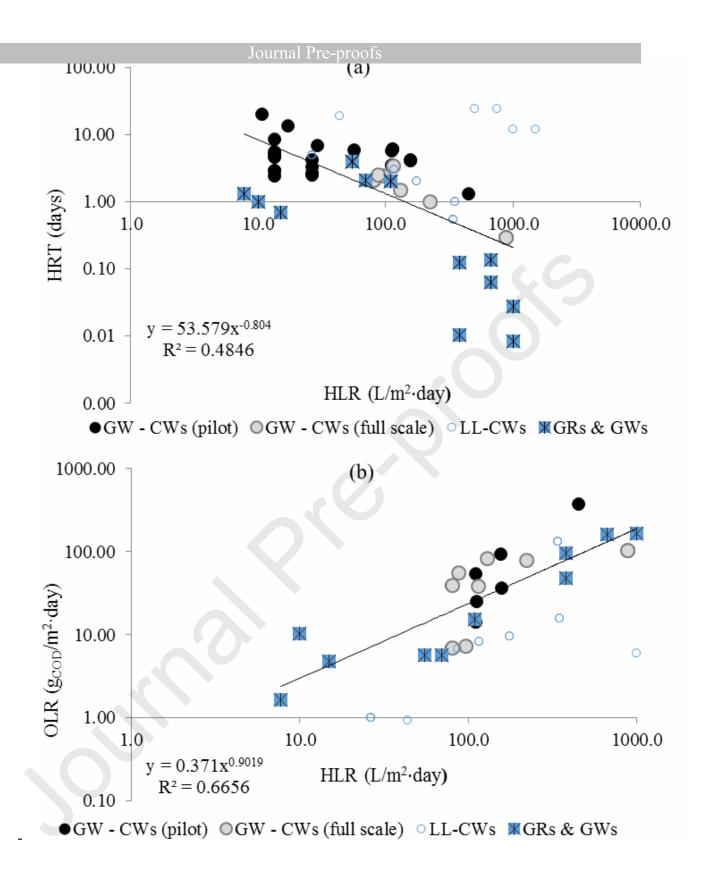
		for on-site resource recovery	
Evaluation of annual		The predicted 95th percentile annual risks for	
	Monte Carlo approach captured	non-potable indoor reuse of distributed GW	
probability of infection for	variation	and domestic wastewater at district and	
non-potable exposures to distributed <b>GW</b> and domestic	Reference pathogens:	building scales were lower than the selected	(Schoen, et al.,
	Norovirus, Rotavirus,	health benchmark of 10 <sup>-4</sup> infections per	2018)
wastewater treated with	Campylobacter jejuni, and	person per year for all pathogens except	
aerobic membrane bioreactor	Cryptosporidium spp.	Cryptosporidium spp	
(MBR) + chlorination			
		CW man LCC 44.40/ sprits sector	
Comparatively evaluation of		- GW reuse LCC: 44.4% capital costs +	
life cycle costs and expected	<ul> <li>LCA cradle to grave</li> </ul>	46.4% operational energy costs	(Yerri &
monetary benefits of	– LCC	- Conventional wastewater treatment LCC:	Piratla, 2019)
	- Sensitivity analysis	39.9%, operational energy costs + 25.5%	1 iiaiia, 2019)
decentralized GW reuse		land use costs+ 24.3% piping capital costs	

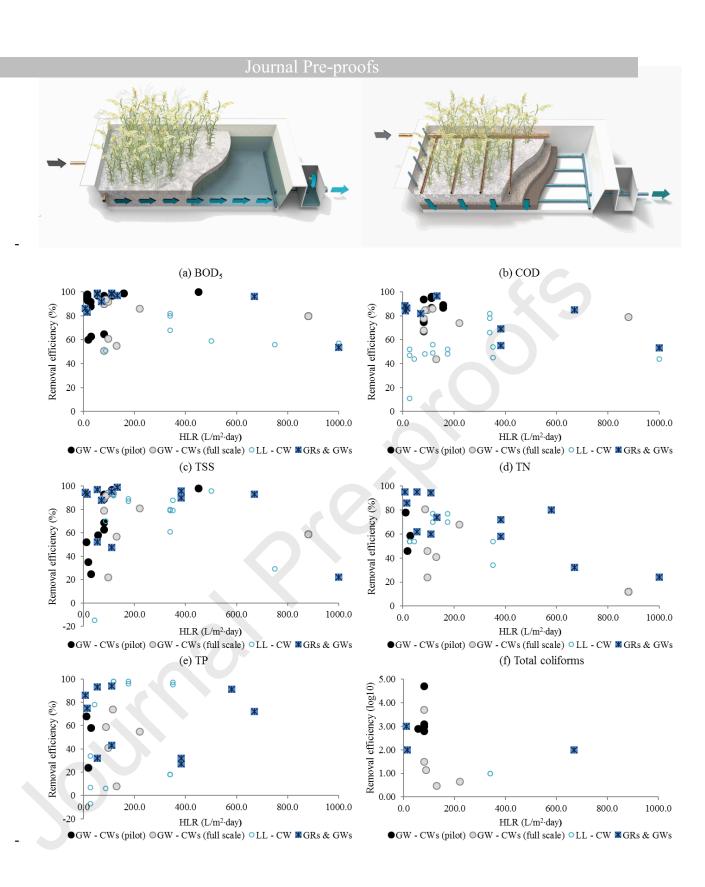
- Nature-based solutions (NBS) are a viable option for greywater (GW) treatment
- Review of case studies provided numerical thresholds for hydraulic design of NBS
- Life cycle assessment studies demonstrated the benefits of NBS for GW treatment

Authors' contributions: conceptualization, methodology and supervision: Fulvio Boano and Silvia Fiore; writing of original draft: all authors; review and editing of the manuscript: Fulvio Boano, Silvia Fiore, Luca Ridolfi, Ana Galvão, Anacleto Rizzo and Fabio Masi



Nature-based solutions for greywater treatment





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Nature-based solutions for greywater treatment

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