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Review

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

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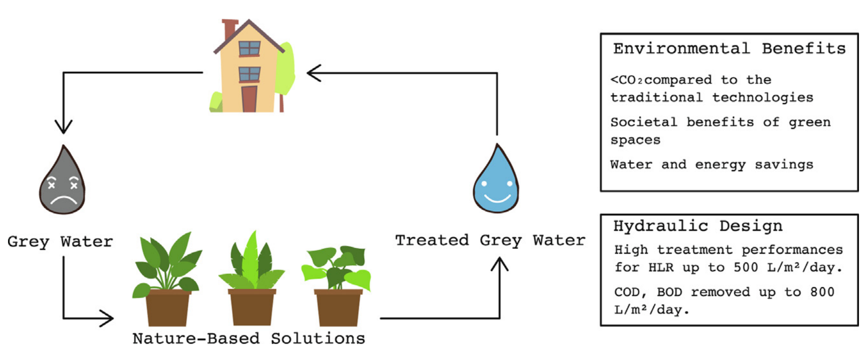
HIGHLIGHTS

- 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.

GRAPHICAL ABSTRACT



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ABSTRACT

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.

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Contents

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 and 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 and 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 (Ghaitidak and Yadav, 2013). The source separation of GW can reduce the volume sent to WW treatment 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, 2015). 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 and Jones, 2008; Castleton et al., 2010), reduce flooding risk (Rizzo et al., 2018), and promote a sustainable economic growth (Maes and 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 and Chiang, 2018). Finally, NBS support biodiversity by providing habitats for different species, significantly (Francis and 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.

NBS for GW treatment emerged around two decades ago. In a review of the technological approaches for 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 and 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 - green roofs", and "greywater - green walls" (and equivalent keywords). Limiting the research to the category "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.

Fig. 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 Fig. 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 mitigation and thermal insulation (Del Barrio, 1998). Similarly, green walls were firstly adopted as aesthetic elements

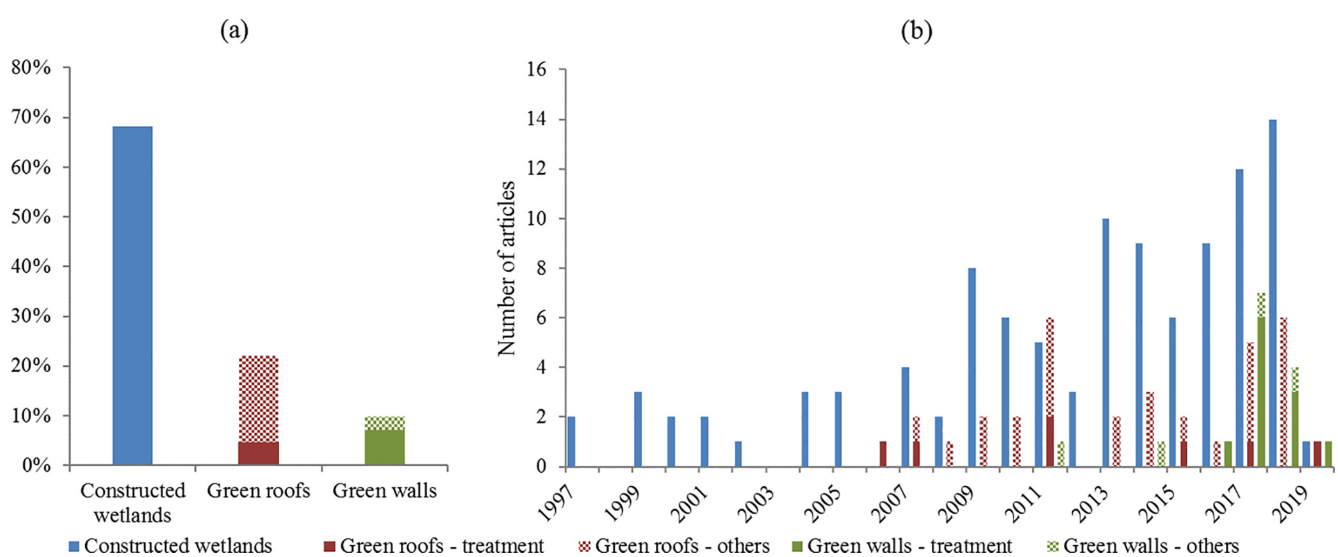

Fig. 1. (a) Relative contributions of the 154 studies regarding greywater use coupled with different nature-based solutions (105 on constructed wetlands, 34 on green roofs and 15 on green walls) and (b) number of articles about NBS based on greywater treatment published per year. Solid colours are related to NBS applications concerning greywater treatment and reuse, while hatched fillings indicate other types of applications. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

of buildings, providing architectural and environmental benefits (Köhler, 2008; Alexandri and Jones, 2008; Weinmaster, 2009). In the last decade, a growing interest in NBS designed for GW treatment can be observed (Fig. 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.

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 and 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 and Yadav, 2013; Leonard et al., 2016; Kaposztasova et al., 2014). These differences are attributed to the fact that toilet flushing represents a large fraction of WW in high-income countries, in contrast with the limited diffusion of household toilets in low-income countries.

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 and 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.

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

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 and Yadav, 2013); laundry GW contains high concentrations of chemical products (e.g. soaps and oils) and non-biodegradable fibres (Morel and Diener, 2006; Ghaitidak and Yadav, 2013); kitchen GW (from sinks or dishwashers) contains food, oils, fats, and detergents (Noah, 2002; Ghaitidak and 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 included into greywater, but high concentration of phosphorus and nitrogen are usually considered indicators 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 and Yadav, 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_5) 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 coun-

Table 2
Physico-chemical features of domestic greywater in different countries. All values are reported as minimum-maximum (average), depending on available data.

Country	pH	TSS (mg/L)	BOD_5 (mg/L)	COD (mg/L)	TN (mg/L)	TP (mg/L)	TC (MPN/100 mL)	FC (MPN/100 mL)	*E. coli* (MPN/100 mL)
	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)
Australia[a]	–	(74)	(104)	–	(5.3)	(3)	–	–	–
Canada[b]	6.7–7.6	–	–	278–435	–	0.24–1.02	–	4.7E+04–8.3E+05	–
Egypt[c]	6.05–7.96 (7)	70–202 (116)	220–375 (298.6)	301–557 (388)	–	8.4–12.1 (10.54)	–	–	–
France[d]	6.46–7.48 (7.28)	23–80 (59)	85–155 (110)	176–323 (253)	–	–	1.7E+08–1.4E+09 (4.9E+08)	4.0E+03–5.7E+06 (1.3E+06)	–
Germany[e]	(7.6)	–	(59)	(109)	(15.2)	(1.6)	–	(1.4E+05)	–
Ghana[f]	5.00–9.00 (6.89)	192–414 (296.8)	87–301 (204.1)	207–1299 (643.8)	–	1–3 (2.3)	2.5E+06–4.9E+06 (3.7E+06)	0–6.9E+06 (1.80E+06)	–
India[e,g,h,i,j]	5.90–8.34 (7.4)	53.80–788.00 (337.2)	17.10–290.00 (244.2)	43.90–733.00 (705.4)	17.00–28.82 (17.8)	0.01–3.84	–	5.0E+01–1.2E+02	–
Indonesia[k]	(6.85)	(18.00)	(8.50)	(15.00)	–	–	–	–	–
Israel[e]	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[e]	–	–	–	(675)	(25.6)	(1.1)	–	–	8.5E3-1.2E4
Jordan[e]	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)
Malaysia[m,n]	6.5–7.2 (6.85)	19–175 (114.4)	1.1–309 (188.85)	16–1103 (328.9)	–	(4.5)	–	0–1.9E+06 (2.9E+05)	0–6.7E+03 (1.1E+03)
Niger[e]	(6.9)	–	(106)	–	–	–	–	–	–
Norway[o,p]	(7.1)	(39)	(129)	(241)	(10.61)	(1.03)	(6.8E+06)	–	(4.9E+06)
Oman[e]	6.7–8.5 (7.5)	11–505	25–562	58–486	–	–	2.0E+02–3.5E+03	(2.0E+02)	(2.0E+02)
Pakistan[e]	(6.2)	(155)	(56)	(146)	–	–	–	–	–
Palestine[q]	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 Korea[e]	(7.4)	(2180)	(255)	–	–	–	–	–	–
Spain[e,r,s]	(7.39)	(336.09)	(130.32)	151–177 (409.11)	10.00–11.00 (16.17)	–	–	(1.0E+03)	(1.0E+03)
Sweden[e]	(7.8)	–	(425)	(890)	(75)	(4.2)	–	(1.7E+05)	–
Taiwan[e]	6.5–7.5	(29)	(23)	(55)	–	–	(5.1E+03)	–	–
Tunisia[e]	(7.5)	(33)	(97)	(102)	(8.1)	–	–	–	–
Turkey[e]	–	(54)	(91)	190–350	(7.6)	(7.2)	–	(1.1E+04)	–
UK[e,t]	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 Emirates[u]	–	–	–	(1020)	–	–	(1.0E+07)	–	–
USA[e]	(6.4)	(17)	(86)	–	(13.5)	(4)	–	–	(5.4E+02)
Western Europe [v]	6.1–9.6 (7.5)	20–361 (89)	20–756 (221)	25–1583 (362)	3–75 (14)	0–11 (4)	–	–	–
Yemen[e]	(6)	(511)	(518)	(2000)	–	–	–	(1.9E+07)	–

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

tries, 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 and Scholz (2016) provided a comparison between different recipes: some of them use commercial products, others chemical ones. There are also some studies that suggest different recipes (Rodríguez-Chueca et al., 2014; Charchalac et al., 2015; Chrispim and 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 and 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_5, 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.

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_5, 64% COD, 13% total nitrogen, >99% TOC and >99% *E. coli* (Ghaitidak and Yadav, 2013).

Table 3
Overview of regulations and guidelines for GW reuse.

Parameter	Regulations								Guidelines					
	Great Britain		USA		NSW-Australia		Italy	Japan	Germany	Slovenia	Jordan	China	India	Canada
	Sprinkler use	Non-sprinkler use	Residential	Commercial	Irrigation	Household								
pH	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	–	–	–	10,000	–	–	–
BOD_5 (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 (NTU)	<10	<10	<5	<2	–	–	–	Clear	Near clear	–	2–10	<10	–	<2
Anionic surfactants (mg/L)	–	–	–	–	–	–	0.5 (total)	30	–	1	30–100	0.5–1	<10	–
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)	–	–	–	–	–	–	1	–	–	–	1	–	–	–
FIB (MPN/100 mL)	Not detected	25 (*E.coli*)	~0	~0	<1	<4	100 (*E.coli*)	$<1\cdot10^5$	<10 (total) < 100 (faecal)	–	<1000 (faecal) 100 (*E.coli*)	–	–	<1000 (total) 2–200 (faecal)

Table 4
Proposed guidelines and reports for GW reuse (no standards or limits are available).

Country	Guideline	Main finding	Reference
Europe	Directive 91/271/EEC on urban waste water treatment (UWWTD)	→ Wastewater shall be reused whenever appropriate → Member States shall minimise any adverse effects on the environment from wastewater reuse → 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).	Directive 91/271/EEC concerning urban waste water treatment (UWWTD)
United Nations	Guidelines for the safe use of wastewater, excreta and greywater	→ Chapter 2: Technical perspectives → Chapter 8: Environmental aspects	(WHO, 2006)
Australia	National guidelines for water recycling: managing health and environmental risks (phase 1)	→ Chapter 2: Framework for management of recycled water quality and use → Chapter 3.7: Managing health risks in recycled water, GW, microbial and chemical risks	(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	→ Coarse filtration + metal strainer to retain suspended particlesMembrane filtration to clarify water	(Li, et al., 2010)
North America	Overview of greywater reuse: the potential of greywater systems to aid sustainable water management.	→ Challenges and opportunities for GW reuse → GW as percentage of total water use → GW and energy → GW and agriculture	(Allen, et al., 2010)
Israel	Greywater use in Israel and worldwide: standards and prospects	→ Separated collection of GW from wastewater → Biological treat is based on membrane + UV disinfectionReuse for 1 yard irrigation and/or toilet flushing	(Oron, et al., 2014)
Europe	Technical guide for greywater recycling systems	Treated GW shall only be used for the following applications: → Flushing of water closet (WC)/Urinal. → General washing (excluding high pressure jet washing and general washing at markets and food establishments) → Irrigation (excluding irrigation sprinklers)Cooling tower makeup water.	(PUB, 2014)
Australia	Code of practice – onsite wastewater management	→ Chapter 3: Onsite wastewater management in unsewered areas → Chapter 4.1: GW overview → overview of GW policies, regulations, and laws around the world	(EPA Victoria, 2016)
Europe	Guidelines on integrating water reuse into water planning and management in the context of the WFD (Water Framework Directive)	→ Water scarcity → Greywater reuse in agriculture → Greywater health risk	(EPA, 2016)
United Nations	United Nations: world water development report.	→ Chapter 2: wastewater and the sustainable development agenda → Chapter 4: technical aspects of wastewater → Chapter 5: municipal and urban wastewater → Chapter 7: agriculture as a user of wastewater	(United Nations, 2017)
United Kingdom	BS8525 and the Water Supply (Water Fittings) Regulations	GW can be reused on site for: → ornamental, garden and lawn irrigation, → toilet flushing	BS8525 and the Water Supply (Water Fittings) Regulations

Table 5
Conventional technologies for GW treatment and reuse.

Conventional/advanced process			GW origin	Treatment Efficiency (% removal)	Reference
Chemical	Coagulation	$FeSO_4$ (13%v/v) = 0.79 nM	Synthetic GW: Showers + sinks	BOD_5 = 85.37%, COD = 63.59% NO_3^--N = 8.96%, TN = 0.56%, TP = 96.39%	(Pidou, et al., 2008)
		$Al_2(SO_4)_3$ (48%v/v) = 0.89 nM		BOD_5 = 88.28%, COD = 63.72% NO_3^--N = 14.93%, TN = 12.78% TP = 94.58%	
	Electro-coagulation (EC)	Electrode combination = Al–Fe–Fe–Al, pH_i = 7.62, CD = 1 mA/cm^2	Real GW: showers, sinks and kitchen	COD = 90–95%	(Barışçı and Turkay, 2016)
	EC/O_3	pH = 7.0, Ozone = 47.4 mg/L, CD = 15 mA/cm^2	Real GW: showers, sinks	COD = 85%, TOC = 70%	(Barzegar, et al., 2019)
	EC/O_3/UV	pH = 7.0, Ozone = 47.4 mg/L, CD = 15 mA/cm^2, UV ray		COD = 95%, TOC = 87%, *E. coli* = 96%	
	Photocatalytic fuel cell (PFC)	ZnO/Zn photoanode, CuO/Cu photocathode, Illumination area = 6 $cm^{2.}$ Photoelectrodes distance = 4 cm. Light source = 365 nm mercury UV lamp. Air bubbling (2 L/min) supplied at the photocathode.	Real GW: laundry	COD = 55%, BOD_5 = 55%, Turbidity = 88%, NH_4^+-N = 75%	(Kee, et al., 2018)
Physical	Filtration	Recycled vertical flow bioreactor. Layer1: 2-cm thick crushed dolomite + limestone, average diameter 2.5 cm, Layer 2: 12 cm of plastic filter media with a high surface area (800 m^2/m^3) and large void volume	Synthetic GW: laundry + bath + kitchen	TSS = 93.48%, NH_4^+-N = 16.67%, Anionic surfactant = 98.37%, NO_2^--N = 96.92%, NO_3^--N = 48.57%, TP = 73.68%, *E.coli* = 97.5%	(Gross, et al., 2007b)
	Filtration: Filters: grain size distribution 0.8–6.3 mm; D_{10} = 1.4 mm, D_{60} = 3.1 mm; uniformity coefficient = 2.2.	Pine bark (bark)	Synthetic GW: Bath + laundry	pH from 7.8 to 6.1, BOD_5 = 98%, EC = 7.14%	(Ghaitidak and Yadav, 2013)
		Activated charcoal (charcoal)		BOD_5 = 97%, EC = 11.7%	
	Filtration: HRT = 24 h	Coarse	Real GW: shower + washing machine from household	TS = 14.21%, COD = 20%, NH_4^+-N = 17.74%, *E. coli* = 98.31%, SAR (sodium adsorption rate) = 4.9%	(Ghaitidak and Yadav, 2013)
	Filtration: HRT = 24 h	Layer1: coconut shell cover 20 cm, Layer2: saw dust (coarse) 15 cm, Layer3: charcoal 20 cm, bricks 10 cm and sand 15 cm.	Synthetic GW: kitchen	TSS = 82.61%, TDS = 69.98%, COD = 82.26%, Oil-grease = 96.97%, NH_4^+-N = 73.42%, NO_3^--N = 68.66%, PO_4^{3-}-P = 100%	(Parjane and Sane, 2011)
	HRT = 30–40 d; sand filter = 5 cm plexiglass column	Sedimentation tank + sand filter	Synthetic GW: kitchen	Turbidity = 20%, COD = 25%	(Kee, et al., 2018)
	HRT = 30–40 d; Flowrate = 2.8 L/h; Filtering velocity = 1.4 m/h.	Sedimentation tank + granular active carbon (GAC)		Turbidity = 25%, COD = 27%	
Physico-chemical	Electrochemical reactor (boron-doped diamond) anode = 80 × 80 × 1.3 mm, 3D reactor a packed bed of GAC was placed in between anode and cathode. Anodic CD = 15 A/m^2	Hybrid granular active carbon (GAC)-electrochemical (EC) system	Synthetic GW: shower	COD = 68%, TOC = 70%	(Andrés García, et al., 2018)
Biological	Rotating biological contactor (RBC)	RBC was made of plastic sheets with tank volume of 54 L	Real: laundry + bath + kitchen	pH value increased with HRT, TDS not changed, TSS = 8.98–11.08%, BOD_5 = 27.30–52.42%, COD = 21.48–60.36%	(Xiao, et al., 2018)

Table 5 (*continued*)

Conventional/advanced process		GW origin	Treatment Efficiency (% removal)	Reference
Moving bed biofilm reactor (MBBR)	Q_{max} = 31.2 L/h, $Q_{max\ air}$ = 110 L/min	Synthetic GW: laundry + bath	Turbidity = 66%, TSS = 87.07, TP = 12%, BOD_5 = 59%, COD = 70%	(Chrispim and Nolasco, 2017)
Membrane bioreactor (MBR)	Membrane area = 120 cm^{2}, Pore size membrane = 0.4 μm, High container = 1.8 L, Inoculum = activated sludge from wastewater treatment plant, HRT = 2.5–10 h	Real: laundry + bath + kitchen	Turbidity = 98%, BOD_5 = 93.37%	(Hu, et al., 2011)
Membrane bioreactor (MBR)	Reactor capacity = 3 L UF membrane	Real GW: Shower	Turbidity = 98.28%, NH_4^+-N = 72.03%, TN = 62.05%, TP = 18.75%, Anionic surfactant = 96.66%, COD = 86.24%, BOD_5 = 93.22%	(Merz, et al., 2007)
SBR	SBR volume tank = 2 L	Synthetic GW: Shower	COD = 90%	(Weitao, et al., 2006)
Up-flow anaerobic sludge blanket (UASB)	UASB = 5 L	Real GW: Shower (12 houses)	COD = 51%, N = 37%, NH_4^+-N = 47%, P = 50%	(Hernández Leal, et al., 2011)
Biological activated membrane bioreactor (BAMB)	HRT = 100 d, Q = 75 L/day, nutrient supplement 10 mg/L TOC to avoid inhibition from surfactant.	Real GW: Bathroom and washbasin	C = 95%	(Ziemba, et al., 2018)

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_5, 37–94% COD, 5–98% total N, 17–73% NH_4^+-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_5 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_5 and COD were 59% e 70% respectively (Chrispim and Nolasco, 2017). In the last decade, membrane-based techniques have been optimised to reach the GW reuse standards. Generally achieved removal efficiencies of membrane bioreactor (MBR) systems are: 98–99.9% turbidity, 100% TSS, 93–97% BOD_5, 86–99% COD, 52–63% TN, 6–72% NH_4^+-N, 19% TP and 10–40% PO_4^{3-}–P. MBR systems satisfy most standards for water reuse, as pH, turbidity, BOD_5, COD, TSS and NH_4^+-N (Chrispim and Nolasco, 2017). Sequencing batch reactor (SBR) systems are characterized by a great flexibility of operation and time-controlled sequence, and they are considered an effective nutrient removal technology (Rodda et al., 2011). As a whole, BOD_5 removal varied from 80 to 98% and a similar range of COD removal values is observed. Effluents from SBR treatment of GW from showers satisfied NH_4^+–N, BOD_5 and COD standards for wastewater reuse (Lamine et al., 2007).

4. Nature-based technologies for greywater treatment

Constructed wetlands (CW) were the first NBS applied to GW treatment (see Section 2). GW remediation 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 and Wallace, 2009). The efficiency of these systems in WW treatment is due to a strong interaction among plants, biofilms, substrate, atmosphere, and nutrients from wastewater. The contact among roots, substratum, and biofilm favours different fundamental mechanisms of pollutant and pathogen removal, such as sedimentation and filtration as physical processes, precipitation and adsorption as chemical processes, and microbiological degradation and plant uptake as biological processes. The large amount of involved mechanisms entails a huge variability in removal efficiency (Arden and Ma, 2018). Despite the growing interest on CW application to GW over the last decade, most literature refers to pilot systems with only a few full-scale applications.

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 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 lightly loaded constructed wetlands (LL-CWs), namely studies performed with secondary or tertiary treated wastewater. Among the papers related to GW treatment using NBS identified through the literature survey (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 studies as green roof and green wall applications. Compared to the studies shown in Section 2, we here analyse the subset of studies of NBS where treatment performances were quantified.

Fig. 2 shows the relationships among the main operational factors of the treatment systems considered in the meta-analysis, distinguishing among CWs treating GW (pilot and full scale), CWs treating lightly loaded WW, green roofs and green walls. Specifically, the upper panel represents the relationship, where available, between hydraulic retention time (HRT) and hydraulic loading rate (HLR), while the lower one represents the relationship, where available, between organic loading rate (OLR) and HLR. As expected, the three parameters are correlated. The plot in Fig. 2a indicates a negative correlation between HRT and HLR with longer retention times corresponding to lower HLRs, with a few exceptions for lightly loaded CWs. This means that the two parameters provide similar information about the hydraulic behaviour of the system, and a first-order analysis of the system performance in treating GW can be investigated as a function of either HLR or HRT. As to OLR, Fig. 2b shows that it is directly proportional to HLR, as expected from the relationship among these two parameters (OLR = HLR·$C_{COD,i}$, where $C_{COD,i}$ is the COD concentration in the influent). The correlation among HLR, HRT, and OLR allows to investigate the treatment efficiency of different systems primarily as a function of only one of these parameters, and the derived optimization criteria can then be extended to the other parameters as well. Specifically, in our analysis we focus on HLR, that is available or easily calculated for most of the considered case studies while HRT and OLR were reported less frequently. Moreover, HLR is a very informative parameter for designers of systems treating GW as it allows to determine the space required for the system.

4.1. Constructed wetlands: applications to GW

Both natural and CWs biodegrade or immobilize a range of emerging pollutants, including certain pharmaceuticals, and perform better than conventional "grey" solutions (Kadlec and Wallace, 2009; Arden and Ma, 2018). CWs for domestic wastewater treatment are widely recognized as a sustainable, low-cost, low maintenance technology, given the reduced energy needs and simplicity of operation (Massoud et al., 2009; Kadlec and Wallace, 2009). The most common types of CWs include horizontal

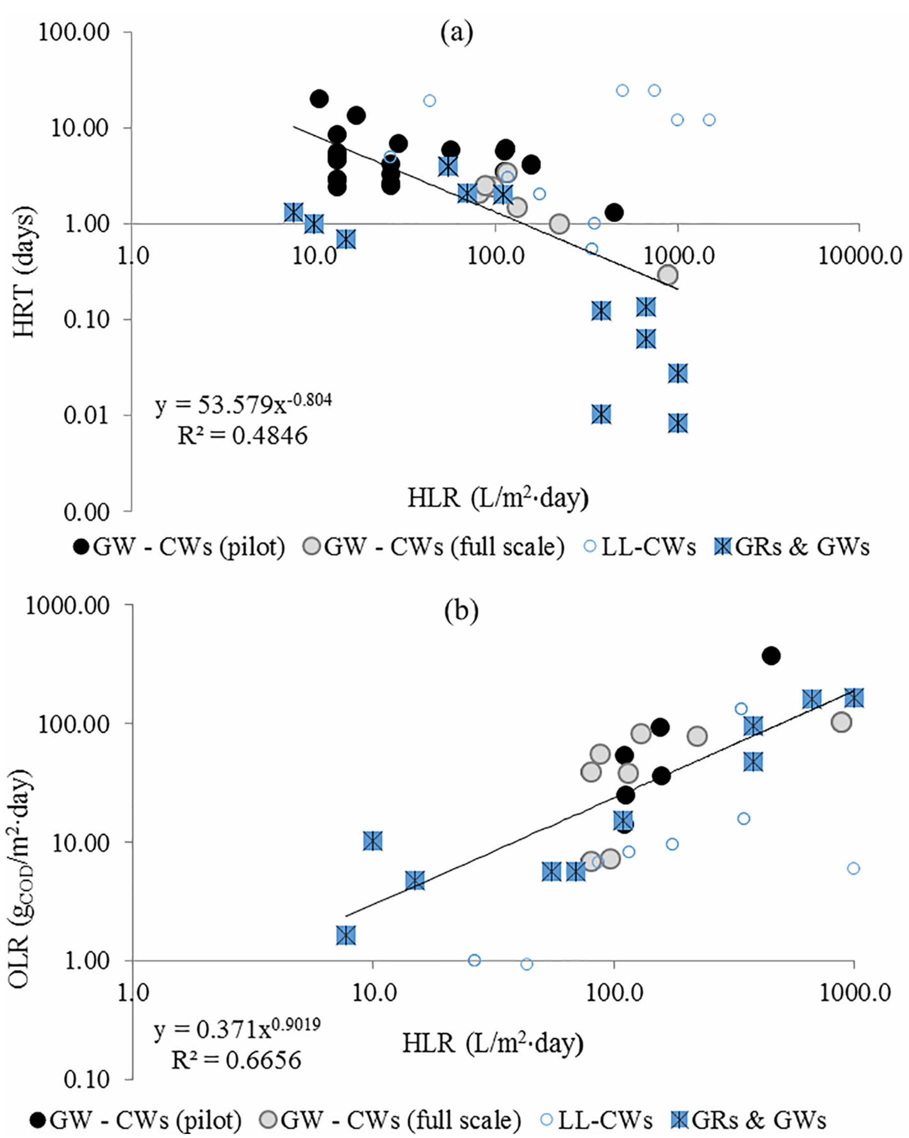

Fig. 2. (a) Hydraulic retention time (HRT) and (b) organic loading rate (OLR) versus hydraulic loading rate (HLR). Pilot and full scale constructed wetlands treating greywater (GW-CWs), lightly loaded constructed wetlands (LL-CWs), and green roofs and green walls are distinguished. The regression lines do not include data from LL-CWs. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

or vertical subsurface flow (Fig. 3), which restrict the contact with the wastewater, and provide good removal of total suspended solids and organic matter, even with low spatial footprint (about 1 m^2/PE in cold and temperate climates, less than 0.5 m^2/PE in warm climates (Masi et al., 2010)). CWs can also be designed as free water surface systems, which have the highest biodiversity level amongst all the possible wetland configurations (Hsu et al., 2011). Additionally, the abundance of organic surfaces (litter, plant stems, microalgae, etc.) supports the adsorption of persistent organic pollutants, which are then slowly degraded (Matamoros et al., 2016). Several full-scale CWs for GW treatment are described in literature for single house applications, in many 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 management in accordance with circular economy principles (Masi et al., 2018). Within this framework, Arden and Ma (2018) listed 11 different treatment schemes and the following ranges of removal efficiency: BOD_5 63–98%; TSS 64–98% for subsurface flow systems and 25% for free water surface; turbidity 47–97%; TN 44–59%; TP 24–63%; around 1–2 log removal for bacteria, protozoa and viruses. Despite high pathogen removal, the authors highlighted the need of some additional disinfection steps (such as chlorine and UV) to meet strict water reuse standards. Arden and Ma (2018) also evidenced a relatively low number of studies on 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 only a few works (Laaffat et al., 2015; Paulo et al., 2009; Masi et al., 2010) referring to full-scale systems. Therefore, the next section reviews the available literature, separately analysing the insights gained from pilot studies and the diffusion of full-scale applications of CWs for GW treatment and reuse.

4.1.1. Pilot-scale systems

The analysed pilot-scale studies included 13 peer-reviewed papers concerning constructed wetlands treating greywater (GW-CWs) and included mostly subsurface flow systems, since free water surface systems could be found only on Gerba et al. (1999), in Arden and Ma (2018) and Jokerst et al. (2012). A total of 30 individual pilot systems were analysed, with more than half being horizontal-subsurface flow systems. Given the small number of cases, a comparison was established with lightly loaded constructed wetlands (LL-CWs), namely studies performed with secondary or tertiary treated wastewater. A total of 24 individual datasets were collected from 9 peer-reviewed studies with CWs treating secondary or tertiary wastewater. 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 Table 7.

Fig. 4a and b show the range distribution of COD and BOD_5 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_5 data (Fig. 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_5 removal rates for LL-CWs are lower than for GW-CWs, and consistently close to a plateau around 60% (global average of 62%) when HLR are higher than 500 L/m^2/day. In fact, GW-CWs show COD 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^2/day with a global average of 52%. This consistency seems to show an upper removal limit capacity for organic matter removal, most likely revealing a more recalcitrant nature of COD in secondary or tertiary wastewater. The higher biodegradability characteristics of GW had been previously reported in several studies in areas of water availability (Karlgren et al., 1967) as well as in water scarcity areas (Al-Jayyousi, 2003; Sall and Takahashi, 2006).

The removal efficiency for TSS of GW-CWs showed values higher than 85% for HLR between 80 and 450 L/m^2/day (Fig. 4c). For loading rates below 80 L/m^2/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^2/day, dropping to values lower than 50% for higher loads.

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

Parameter	Inflow (mg/l)	
	GW-CWs	LL-CWs
TSS	12.3–158.0	2.1–189.0
BOD_5	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
TP	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

* MPN/100 mL.
** m^3/d.

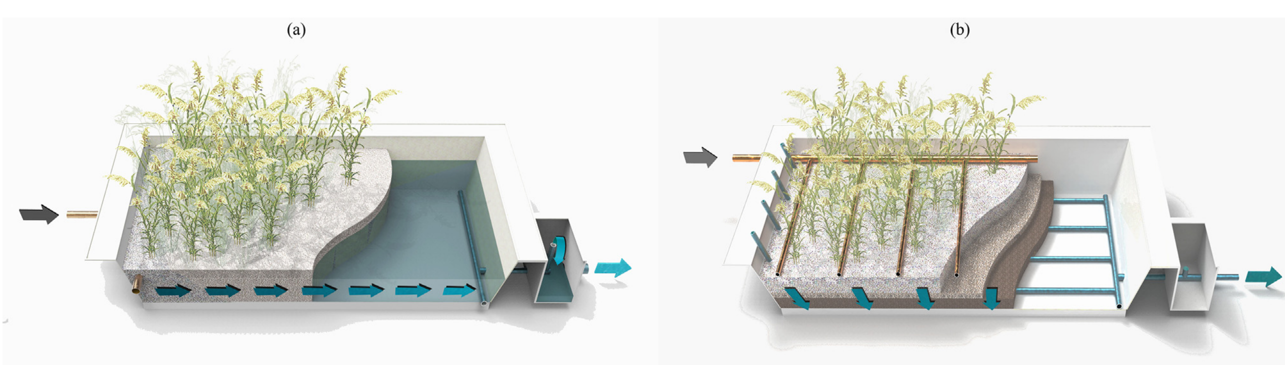

Fig. 3. Schemes of (a) horizontal and (b) vertical subsurface flow constructed wetlands (Courtesy: IRIDRA srl).

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).

Reference	CW tech	Plants	Filling media	GW source	Areal footprint (m^2)	Flow (m^3/day)	HLR (L/m^2/day)	HRT (days)	OLR (g_{COD}/m^2/day)
(Jokerst, et al., 2011)	FWS, HSSF	*Typha latifolia* (FWS) *Scirpus acutus* (HSSF)	Gravel (HSSF)	Bathroom sink, shower	10–17	0.29	17–29	9.3–12	–
(Dallas and Ho, 2005)	HSSF	*Coix lacryma-jobi*	PET (100–150 mm) or crushed rock (20 mm)	Greywater, nonspecific	0.38	0.005–0.01	13–27	2.5–7.2	–
(Winward, et al., 2008)	VF, HSSF	*Phragmites australis*	Sand/soil/compost	Bathroom sink, shower	6	0.48	80	2.1	7–40
(Alfiya, et al., 2013)	RVF	No information	Wood- chips, tuff gravel, limestone pebbles.	Greywater, nonspecific	1	0.11–0.16	112–157	4.1–5.8	251–933
(Blanky, et al., 2015)	RVF	No information	Wood- chips, tuff gravel, limestone pebbles.	Bathroom sink, bath, shower, washing machine	1	0.11	110	6.1	–
(Gross, et al., 2007a)	RVF	No information	Organic soil, tuff or plastic media, limestone pebbles.	Laundry, showers and sink	1	0.45	45	1.3	3775
(Yu, et al., 2015)	RVF	*Carex spissa, Phyla nodiflora*, household plants (*Aeonium purpureum* and *Crassula ovate, Equisetum hyemale, Nasturtium, Narcissus impatiens, Anigozanthos, Colocasia*)	Coconut coir, cross-flow media	Bathroom sink, shower, laundry	0.77	0.086	111	3.5	142–545
(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 and Vråle, 2003)	HSSF	*Phragmites australis*	Light weight aggregates	Secondary GW from aerobic biofilter	–	–	–	6–7	–

Regarding nutrient removal, it was observed that inflow concentrations of TN and TP varied significantly, between 5 and 34 mg/l and from near zero to 23 mg/l, respectively. In terms of removal efficiency, GW-CWs and LL-CWs treatment systems showed a similar behaviour regarding TN, with a good removal between 46% and 78% for all systems except one (Fig. 4d). On the other hand, TP removal in these systems (Fig. 4e) showed very different results, including both high removal rates (close to 100%) as well as extremely low levels (reaching negative values). This wide range of behaviours could be due to the filling 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 50% peat. In this system TP removal efficiency was 58%.

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 (Fig. 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 10^4 (Gerba et al., 1999; Winward et al., 2008), which is not enough to meet standards consistent with several reuse applications. LL-CWs had removal rates lower than 2 log units, but inflow concentrations where typically one order of magnitude lower than GW-CWs. From a visual assessment of the possible correlation between TC removal and HLR, the existence of a specific connection seems unlikely. This can suggest that the main removal mechanism could be due to adsorption on media and filtration processes and, thus, strictly related to the physico-chemical properties of the growing medium (Prodanovic et al., 2017).

4.1.2. Full scale systems

The analysis of peer-reviewed manuscripts on full-scale systems evidenced the suitability of CWs for GW treatment and reuse at real scale and conditions (Fig. 5). Laaffat et al. (2015) reported satisfactorily high removal efficiencies from a 100 days long monitoring of a horizontal flow constructed wetland (HF CW) treating GW from a primary school in Marrakesh: BOD_5 > 92%, COD > 85%, TSS > 94%, TN > 45%, TP, TC > 99%. 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 monthly variable influent GW loads, showing high stability in overall treatment performances over a 120 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., 2009; Masi et al., 2007; Masi et al., 2010) demonstrated generally high removal efficiencies (see Table 8 and Fig. 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 ranges), since less water is used thanks to water saving devices; (ii) a polishing FWS stage can be adopted to both accumulate treated GW for reuse and naturally disinfect it, since 4–6 logs of removal and concentrations of 1–200 MPN/100 mL were observed; (iii) a FWS tertiary unit can avoid the use of technological disinfection 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 stage can lead to occasional increases in effluent COD in warm periods due to algae bloom or volume reduction by evaporation, even if

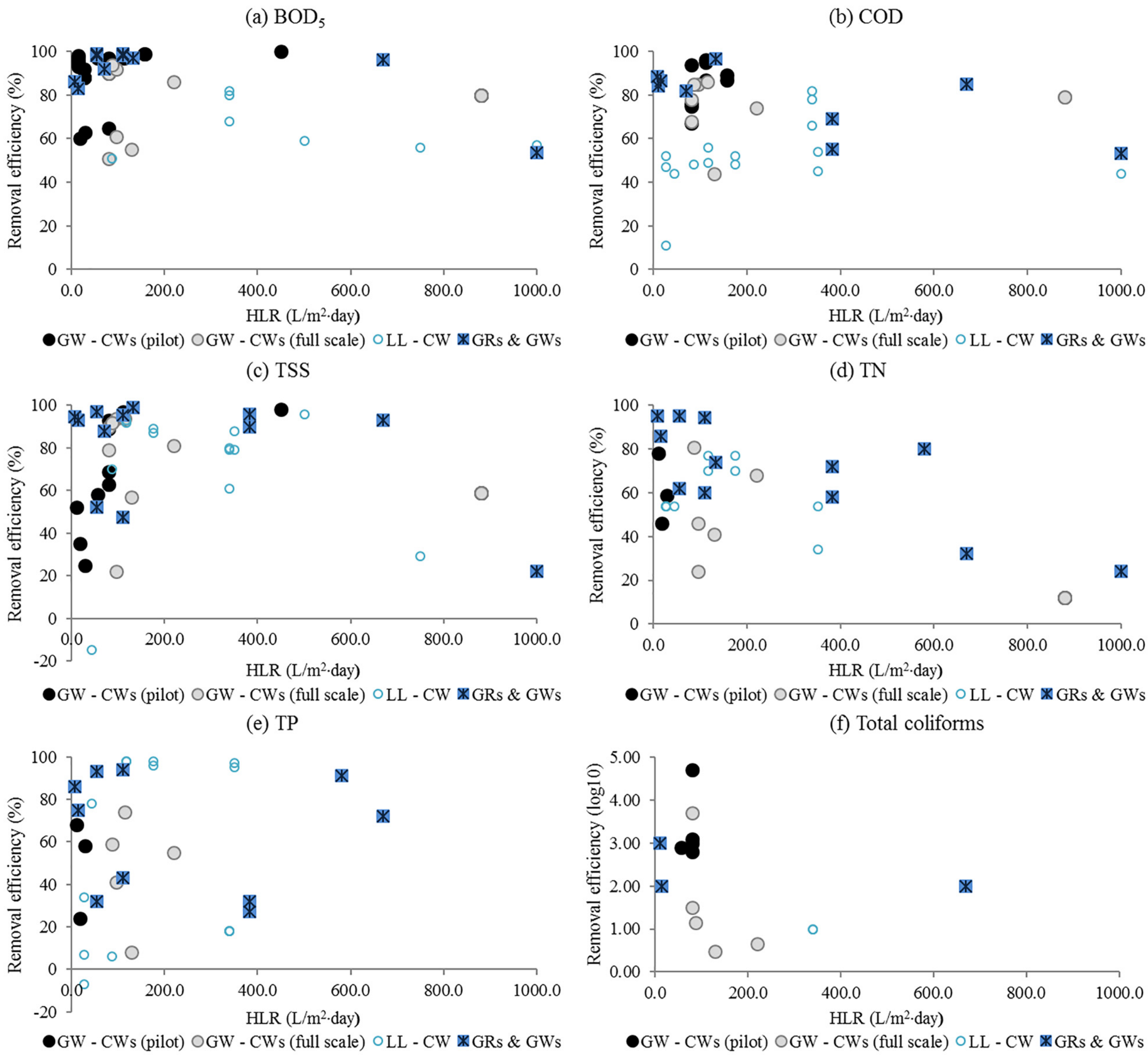

Fig. 4. Removal efficiency data as a function of hydraulic loading rate (HLR) for (a) BOD_5, (b) COD, (c) total suspended solids (TSS), (d) total nitrogen (TN), (e) total phosphorus (TP), and (f) Total coliforms. Pilot and full scale constructed wetlands treating greywater (GW-CW), lightly loaded constructed wetlands (LL-CW), and green roofs and green walls are distinguished. Values of removal efficiencies in panel (f) refer to different bacterial indicators (Table 6 and Table 10). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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 comparison to VF because of the different feeding needs (generally VF systems, operated in batch, are more energy dependant in comparison to HF ones).

The growing demand for employing NBS to treat GW is exemplified by the recently revised German guidelines for design, construction, and operation of CWs for domestic and municipal wastewater (DWA), which specifically discuss design indicators dedicated to the case of GW treatment (Nivala et al., 2018). A survey of technical literature was hence performed to investigate the diffusion of CWs for GW treatment and reuse. To this aim, two sources were analysed: (i) Sustainable Sanitation Alliance (SuSanA – 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 in the field of CWs. The results of this search are summarised in Table 9, which reports details from 15 cases of HF and VF CW applications in real conditions for different GW types (from light GW of showers and washbasins to dark GW of kitchens, as well as car washing GW). The scale of application and the volume of 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 NBS for GW treatment at large scales and not only for single houses. For those systems for which removal efficiencies were available, the results are consistent with pilot-scale studies, in particular for organic matter removal and TSS (Fig. 4). The use of additional disinfection units seems to strongly vary among countries and scales, probably driven by different legislative water quality standards for reuse, while the reuse of 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 and industrial reuse (i.e., concrete

(a) (b)

Fig. 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).

mixing and aquaculture). It is important to stress that the analysis of technical literature presented here only represents a limited portion of real applications, and we reasonably expect a much larger number of similar applications worldwide. Therefore, CWs should be considered a mature technology for the treatment of GW and successive reuse.

4.2. Urban green infrastructures: Green walls and green roofs

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

Green roofs and green walls for GW treatment operate similarly to reed beds, in which GW percolates through planted pots filled with a combination of granular media such as vermiculite, sand, growstone, expanded clay, phytofoam, coco coir, and perlite (Prodanovic et al., 2017). However, their low space requirements represent a relevant advantage compared to the conventional reed beds (Masi et al., 2016). If 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, low weight, and good removal capacity of nutrients (Castellar da Cunha et al., 2018). Additional features 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, 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 to compare different growing media. In the present section we first analyse laboratory studies, continuing with pilot studies, and concluding with real cases.

The first example is given by Fowdar et al. (2017), who employed a set of laboratory columns filled with inert material (sand, gravel) and planted with ornamental species (climbers and flowers) to reproduce a living wall system. The study investigated different operational and design aspects in terms of loading rates, 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 filter bottom can improve nitrate removal through denitrification. However, it was observed that denitrification was limited by the availability of organic matter in the saturated layer. The only carbon source provided by the GW was quickly removed in the upper layers, limiting the presence of biodegradable matter where denitrification could occur. The performance of the system in terms of BOD_5 removal proved to be very high, while some improvements were still required for nitrogen and phosporous removal. A follow-up experimental study (Fowdar et al., 2018) investigated the main mechanisms governing removal and transformation of nitrogen in living wall biofiltration systems, with particular attention to dissolved organic 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 an isotopic tracer (^{15}N-urea) revealed a minor removal contribution provided by nitrification-denitrification pathway. Plant assimilation and media adsorption were likely predominant, highlighting the importance in plant selection in the design phase.

Another laboratory study (Prodanovic et al., 2017)~~,~~ examined different growing media for green walls. The study aim was to identify the underlying pollutant removal processes and to maximize the treatment capacity of the system. Experimental tests were performed in vertical columns filled with different porous media and without vegetation. The tested materials were selected on the basis of physical and chemical characteristics such as specific weight, water and nutrient retention capacity, porosity, capacity to distribute water and support plant growth, sustainability, and

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

CW technology	Location & Year of construction	GW source	Areal footprint (m^2)	Treated flow rate (m^3/d)	HLR ($L/m^2/d$)	Designed effluent water quality or measured removal efficiencies	Operating time	Additional disinfection units	Type of GW reuse	Reference
HF	“La cava” camping, Arezzo (Italy) 2003		241 m^2	80 PE	110–230	Measured removal efficiencies: COD 81% BOD_5 92% NH_4^+ 62% FC 97%	~1.5 years		Irrigation	(Masi, et al., 2007; Scheumann, et al., 2009)
HF + VF	Campo Grande (Brazil) 2007	Shower, kitchen sink, laundry	4.6 + 2.3	0.5–2.5	150	Measured removal efficiencies: TSS 92% ± 12, COD 88% ± 11, BOD_5 95% ± 4, TP 58% ± 35, TN 82% ± 20, *E. coli* 93 ± 12	~5 months	None	Not reported	(Paulo, et al., 2009)
HF	Rabat (Morocco) 2005		4.5	8	880	Measured removal efficiencies: COD 75% BOD_5 80% TP 50% Anionic surfactants 97% FC < 1log	~4 years	Sand filter (4.5 m^2) + UV	Toilet flushing	(Masi, et al., 2010)
HF	Cairo (Egypt) 2007		12	1	83	COD 82–91% TSS 85% NH_4^+ 30–57%	~2 years	None	Toilet flushing, irrigation	(Masi, et al., 2010)
HF + FWS	Istanbul (Turkey) 2007		28 + 35	1	110	TSS < 10 mg/l COD < 50 mg/l BOD_5 < 10 mg/l NO_3^- < 1 mg/l NH_4^+ < 0.5 mg/l	~2 years	UV lamp	Toilet flushing, landscaping irrigation	(Masi, et al., 2010)
HF	Marrakech (Morocco) 2012	Hand wash sinks	12.5	1.2	96	BOD_5 3.45 ± 3.2 COD 11.43 ± 4.5 SS 0.29 ± 0.2 TN 3.89 ± 1.5 TP 0.47 ± 0.3 *E. coli* $5x10^1$	~3.5 months	None	Irrigation of landscape green areas	(Laaffat, et al., 2015)

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

CW technology	Location	Year of construction	GW source	Areal footprint (m^2)	Treated flow rate (m^3/d)	Additional disinfection units	Type of GW reuse	Reference
VF single stage (sand filled)	Polderdrift, Arnhem (Netherlands)	1996	All	230 m^2	6.4 m^3/d	None	Toilets flushing	www.globalwettech.com
Forced Bed Aeration™ (Aerated CW)	National Great Rivers Education and Research Centre, Alton, Illinois (US)		Washbasins, showers, kitchens	47 m^2	5.3 m^3/d	UV	Toilet flushing, landscape irrigation	www.globalwettech.com
VF	West Bank bedouin villages (Palestine)	2012	All	30–60 m^2	70–120 PE	None	Irrigation of olive camps or folder production	www.globalwettech.com
HF	Group of houses in Preganziol (Italy)	2009	All	230 m^2	14.5 m^3/d	None	Toilets flushing	www.globalwettech.com
HF	Group of houses in Vicchio (Italy)	2008	Showers, Hand wash sinks, laundry	50 m^2	30 PE	UV	Toilet flushing, landscape irrigation	www.globalwettech.com
VF	Hostel campus of College of Engineering, Pune (India)	2015	All	130 m^2	40 m^3/d	Chlorine	Toilet flushing, landscape irrigation	(Patil, et al., 2016)
roof HF	Resort in Grumenti community, Serengeti (Tanzania)	2015	All	23 m^2	4 m^3/d	UV	Irrigation	www.globalwettech.com
VF	Private school, Lima (Peru)	2008	Kitchen	16 m^2	1.5 m^3/d	None	Landscape irrigation	(Platzer, et al., 2016a, 2016b) (Hoffman, et al., 2009)
VF	Guest house of 160 apartments, Lima (Peru)	2011	Restaurants and showers	100 m^2	12 m^3/d	Chlorine	Toilet flushing, landscape irrigation	(Platzer, et al., 2016a, 2016b)
VF	Housing project for 288 families, Lima (Peru)	2011	Showers	60 m^2	9 m^3/d	None	Landscape irrigation	(Platzer, et al., 2016a, 2016b)
SuSaNa Database								
VF	Waagner head office (Dubai)	2006	Showers, Washbasins, car washing	500 m^2	270 staff members	None	Concrete mixing, soil watering, car washing, fish ponds	(Sievert and Schilick, 2009)
VF	SAMA Dubai site office (Dubai)	2006	All	40 m^2	200 staff members	None	Manual garden irrigation and car washing	(Sievert and Schilick, 2009)
VF	Dubai Municipality (Dubai)	2006	All	170 m^2	60 staff members	UV (optional)	Drip irrigation and boat washing	(Sievert and Schilick, 2009)
HF	Secondary School, Nakuru (Kenya)	2008	Kitchen	2 m^2	1 m^3/d	none	Agro-forestry	(Machiri, et al., 2009)

Fig. 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).

local availability. Among the hydraulically fast and slow media identified as suitable media, the analysis identified perlite and coconut coir as the two best performing media, suggesting their combined use as best option. Mixing two materials with different hydraulic behavior 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 removal performance of different mixes of coir and perlite in terms of TSS, TN, TP, COD and *E. coli*, providing a complete picture about the physico-chemical removal processes. It was observed that biological removal processes dominate nitrogen and COD removals, while physico-chemical processes dominate removal in fast media.

Some pilot-scale implementations of green roofs irrigated with GW include adaptations of CWs located on 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 of an inclined framework of interconnected horizontal troughs and weirs planted with several plant species. 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 system in reducing suspended solids and turbidity, with no significant difference in effluent BOD_5 and COD concentrations. Ramprasad et al. (2017) evaluated the performance of the GROW system under various climatic conditions, organic loading rates, and flow rates in order to understand the suitability of the system for different geographic areas and climates. Moreover, they focused on the performance of the system in removing surfactants from personal care products. The system was able to reach a removal efficiency higher 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 15 L/m^2/day, respectively. Overall, the removal efficiency depended on seasonality and hydraulic parameters. A lower environmental impact of GROW system compared to membrane based technologies was demonstrated through a Life Cycle Assessment, mostly because of the electricity consumption during the use phase (Memon et al., 2007).

An experimental study concerning green roofs irrigated with GW was performed in the city of Al Ain (United Arab Emirates), testing the difference in treatment capacity between intensive and extensive roof systems (Chowdhury and Abaya, 2018). Intensive and extensive prototypes were differentiated only for the 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 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 both systems.

The first application of vegetated walls for GW treatment was developed in Norway and consisted in a wall-adapted biological filter intermittently dosed with domestic GW (Svete, 2012). In spite of the limited spatial footprint (filter surface area around 0.54 m^2), the system was able to operate with good removal performance as enhanced aerobic conditions and residence times supported fast biogeochemical reaction rates. The system included three different sections, among which the planted one showed higher removal rates, satisfying all the expected treatment perfomances 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 that the higher perfomance were most probably due to higher residence time and confirming the importance of this parameter.

A pilot installation of a green wall was conducted by Masi et al. (2016) for the treatment of GW produced by an office building in Pune (India). The system involved a series of pots filled with porous media (LECA 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 received water through vertical percolation. The effluent collected at the bottom of the wall was then reused for garden irrigation. The main conclusion of the study was related to the importance of the substrate used in the green wall. Even though TN removal was limited, the system was successful in minimizing the treatment footprint while ensuring an effluent quality that respected the Indian regulations for reuse for irrigation purposes and, for some samples, for toilet flushing.

Another example of full-scale application (VertECO – Vertical Ecosystem) was installed in a hotel in Lloret de Mar (Spain) (Gattringer et al., 2016). It consisted of an indoor living wall aimed to treat 1 m^3 of GW per day. An interesting outcome of the study deriving from the application of a Life Cycle Assessment was that the environmental impact of such systems could be improved by using alternative materials, such as plastics or wood, for the realization of the wall, and by employing sustainable energy sources, such as photovoltaic 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 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 for the hydroponic cultivation of lettuce in a green wall, with the addition of urine as a nutrient source for 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.

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 issue that limits the application of these systems for GW treatment is the lack of sizing principles to predict the amount of GW that can be fed and efficiently treated by a specific system. The indications can be obtained from the existing studies will be discussed in Section 5. Another important issue that will require further research efforts is how the ensure durability of these systems. Problems that can mine durability of 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 Pradhan et al. (2019), difficulties related to social acceptance of treated GW reuse should not be dismissed.

5. Hydraulic design criteria and greywater treatment performance

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 suitable operating conditions. While these issues have been widely addressed for constructed wetlands, much less information are available for green walls and green roofs. Some works started to discuss and analyse the selection criteria for defining the best combination of design parameters involved in the realization of vegetated GW treatment systems (Castellar da Cunha et al., 2018; Pradhan et al., 2019). However, a fundamental issue that has not been fully clarified yet is the influence of operational factors on removal efficiencies. The identification of optimal design parameters for green walls and green roofs is essential to 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 green roofs and HLR, with specific regard to the removal of organic matter, nutrients, and bacteria. From a 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 reduce HRT, liming the contact time between GW and microbial biofilms and plant roots. Excessively high values of HLR are hence expected to reduce the removal of contaminants (and particularly those more prone to leaching).

Tables 10 and 11 summarize the design parameters, operational characteristics, and removal efficiencies of 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 laboratory studies. All these parameters control the biogeochemical and physical processes occurring along each pathway, affecting the treatment efficiency. For studies that did not report values of surface HLR in L/m^2/day, these values were calculated dividing the flow rate by the horizontal area of the system. Table 11 presents the

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

Authors	Type of study	Plants	Size	Substrate	Operational factors			
					Flow (L/day)	HLR (L/m^2/day)	OLR (g_{COD}/m^2/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^2	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^2 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. *Typha, Iris*), graminoids (*Carex, Cyperus*) tropical and subtropical plants (e.g. *Ficus, Spathiphyllum, Epiprenum*)	2 m^3 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^2	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^2	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 australis, Vitis vinifera, Parthenocissus tricuspidata, Pandorea jasminoides, Billardiera scandens*	70 columns, 0.05 m^2 each column	68% sand mixed with 5% of cedar mulch, 16% coarse sand, 16% gravel	2.50–5	55–110	5.7–15.4	96–48
(Prodanovic, et al., 2017)	laboratory	No plants	0.00785 m^2	100% coir or rockwool, or 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^3, 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 and Abaya, 2018)	pilot	*Phalaris arundinacea L.*	2.64 m^2 (4 modules, 0.66 m^2 each)	Sand	6.0–7.0	–	10.2	$\ll$24
(Prodanovic, et al., 2018)	laboratory	No plants	0.00785 m^2	Coconut fibre and perlite (C: P 1:4, 1:3, 1:2, 1:1, 2:1, 3:1)	3	382	48.1–122.2	–

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

Authors	Type of GW	Organic removal		N removal		P removal		Bacteria removal	
		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_5: 0–7.5, COD: 3–120	BOD_5: 92, COD: 81.7	–	–	–	–	*E. coli*: <10^4, FC: <10^4 TC: <10^4	*E. coli*: 2.9 log, FC: 2.2 log,TC: 3.8 log (>99%)
(Svete, 2012)	real	BOD_5: 2–6, COD: 29–43	BOD_5: 95–98, COD: 82–88	TN: 8.4–8.8	TN: 31–34	TP: 0.26–0.36	TP: 69–77	*E. coli*: >2·10^4	*E. coli*: 98–99%, 2log
(Gattringer, et al., 2016)	real	BOD_5: 3.81, COD: 13.89	BOD_5: 97.1, COD: 96.6	TN: 4.21, NO_3^--N: 3.61, NH_4^+-N: 0.17	TN: 74, NO_3^--N: −440.3, NH_4^+-N: 96.6	PO_4^{3-}-P: 0.32	PO_4^{3-}-P: 74.6	–	–
(Masi, et al., 2016)	real	BOD_5: 6.7–29.9, COD: 21.4–69.4	BOD_5: 8–86, COD: 7–86	–	–	–	–	–	–
(Eregno, et al., 2017)	real	–	–	TN:1.53	TN: 82	TP: 0.08	TP: 91	–	*E. coli*: 5.1 log, TC: 3.7 log
(Fowdar, et al., 2017)	synthetic	BOD_5: <3.5	BOD_5: >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	–	*E. coli*: 60–100%
(Ramprasad, et al., 2017)	real	BOD_5: <10, COD: <16–24	BOD_5: 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
(Chowdhury and Abaya, 2018)	real	COD: 41–405	COD: 79–90	–	–	–	–	TC: 10^4-10^7	TC: 99% – 3 log
(Prodanovic, et al., 2018)	synthetic	COD: 50–170	COD: 20–85	TN: 1.1–4.8	TN: 15–80	TP: 1.5–4	TP: −156–42	*E. coli*: 0.2·10^3 – 2.4·10^3	*E. coli*: 32–90%

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

treatment efficiency, indicating the effluent concentrations and the percentage of removal of the considered contaminants. The ranges of removal efficiency reported in Table 11 refer to all the configurations investigated in each study, also including configurations that showed a low performance and were thus excluded as effective treatment systems. As shown in this table, the systems generally exhibited good removal performances for organic matter (BOD_5 and COD), with removal rates that reach 90–99% and outlet concentrations that fulfill the limitations for most non-potable water reuse (Ramprasad et al., 2017; Fowdar et al., 2017; Svete, 2012). In some studies (Masi et al., 2016; Prodanovic et al., 2017; Prodanovic 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 and 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).

Fig. 4 shows the removal efficiencies of BOD_5, COD, TSS, TN, TP, and pathogens as a function of HLR. It should be noticed that the values shown in Fig. 4 differ from those reported in Table 11 because the latter represent the whole range of variation obtained in all experiments, while Fig. 4 considers only the best experimental configurations identified in each study in order to highlight the treatment capacity under optimal conditions for each system. However, in those cases when various typologies of systems were tested, the analysis included the results related to the different typologies in order to compare systems with different characteristics and implemented according to different aims (Chowdhury and Abaya, 2018). In the study by Fowdar et al. (2017), the configurations with *Carex appressa* and *Strelitzia Reginae* showed high performances for different water quality parameters and have been hence considered here. The configuration with LECA and coir was chosen as reference in Masi et al. (2016). Regarding the studies conducted by 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 to 2:1 mixes (perlite:coir 1:3, 1:2, 1:1, 2:1) (Prodanovic et al., 2018). For all cases, transient operations during the start-up phase or intermittent loadings were not considered because they are considered not stable (Fowdar et al., 2017; Ramprasad et al., 2017).

For BOD_5 and COD, higher removal efficiencies were found when low HLR values were applied (Fig. 4a and b). This behavior can be explained considering that organic matter is mostly removed by microbial degradation, and an increase of HLR entails a reduction of HRT, resulting in less microbial removal of organic pollutants. However, the relationship between removal efficiency and HLR in Fig. 4a-b is nonlinear, and increases in HLR are not necessarily detrimental for removal performances. For instance, the high removal efficiency of organic matter observed for HLR = 670 L/m^2/day (Svete, 2012) can be attributed to high dosing frequency with low greywater volumes. The results in Fig. 4a and b indicate that a high treatment performance of organic matter can be obtained for values of HLR up to 600–800 L/m^2/day.

The removal of TSS seems to be the least problematic since most of the systems showed very high efficiencies (Fig. 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 as decomposition of plant roots. However, this result is in contrast with the results obtained in stormwater 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).

Concerning the removal of total nitrogen, Fig. 4d apparently suggests that increasing HLR implies lower removal efficiency. However, this behavior is largely influenced by a couple datapoints (Svete, 2012; Masi et al., 2016) and deserves careful discussion; on one hand, it should be noticed that the value indicated for

Table 12
Evaluation of GW reuse by means of LCA and environmental-social-economic assessment.

Objective	Method	Results	Reference
Changzhou, China, evaluation of CO_2, CH_4, N_2O emissions from a vertical subsurface flow **constructed wetland** (VF CW, 1000 m^2) and conventional wastewater treatment plants (WWTPs)	LCA cradle to grave	- WWTP: 7.3 kg CO_2-eq to remove 1 kg BOD_5 - VF CW: 3.18 kg CO_2-eq to remove 1 kg BOD_5 - VF CW may reduce GHG emissions by 8–17 million tons CO_2-eq per year compared to WWTP.	(Pan, et al., 2011)
Brazil: evaluation of **GW reuse** 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	• GW impact is the GW treatment on soil phosphatase activity • GW benefit is the worm avoidance.	(Reichman and Wightwick, 2013)
Quantitative Microbial Risk Assessment (QMRA) was performed for Legionella in (light) **LGW.**	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 toilet flushing using LGW.	- QMRA for treated and chlorinated GW was not significantly higher than the for potable water - Health risk stemming from treated GW is acceptable regarding Legionella infection.	(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 and Tier 2, 2) drinking water standards	- Risk quotient <0.2, which means not appreciable danger for human health over a lifetime exposure to potable water - 14 compounds have risk quotient >risk quotients above 0.2 which may warrant further investigation if GW is used for potable reuse.	(Etchepare and Van der Hoek, 2015)
Morelia in Mexico evaluation pros-cons water reuse in urban area	LCA cradle to grave; Multi-objective optimization: total annualized cost, fresh water consumption, and environmental impact	Best Scenario: simultaneous GW recycling and reusing and rainwater harvesting optimal solution for: Total Cost = 585.57 10^3 €/y, freshwater consumption = −13% compared to conventional treatments, complex environmental impact benefit = +32% compared to conventional treatments	(García-Montoya, et al., 2016)
Sakharale, District Sangli, India: GW reuse coming out from hotel	Evaluation of integrated on-site GW Treatment system (IOGTS)	IOGTS satisfy GW standards for reuse in land application in India.	(Patil, et al., 2016)
Evaluation of irrigation impacts terrestrial and aquatic environments using GW	- Comparison of 4 GW irrigated residential lots with 4 adjacent non-irrigated lots (controls) - Evaluation of metals accumulation in soil, groundwater and surface water comparing measured concentrations to national and international guidelines	- GW increased concentrations of As, B, Cr and Cu exceeding guidelines after only 4 y of irrigation. - 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	(Turner, et al., 2016)
South Korea: evaluation of energy consumption and GHG emissions of conventional water treatments and **water reuse**	- Scenario 1: Conventional water treatment - Scenario 2: Centralised wastewater reuse - Scenario 3: Decentralised wastewater and GW reuses	- Scenario 1: energy consumption = 0.511 kWh/m^3 and GHG emissions = 0.43 kg CO_2-eq/m^3 - Scenario 2 energy consumption = 1.224–1.914 kWh/m^3 and GHG emissions = 0.72–0.83 kg CO_2-eq/m^3 - Scenario 3: energy consumptions 0.246–0.970 kWh/m^3 and GHG emissions = 0–0.33 kg CO_2-eq/m^3	(Chang, et al., 2017)
Evaluation of **GW reuse** for irrigation	• Experiments in Lab on quartz sand	- GW reuse reduces soil wettability. - Wettability of sand wetted with raw GW was reduced. about 15.7% - Washing and biodegradation can reduce GW-induced hydrophobicity of sand. - GW has to be treated before reuse for irrigation.	(Maimon, et al., 2017)
Falmouth, MA town, USA: environmental and economic evaluation of water sanitation and **water reuse**	LCA cradle to grave • Scenario 1: Centralized water and sewage • Scenario 2: Centralized water and composing toilet + on-site GW reuse	Scenario 4 offered the best configuration: - Local Human Health impact = $1*10^{-1}$ daily - Equivalent annual cost = 888 €/y/household	(Schoen, et al., 2017)

(*continued on next page*)

Table 12 (*continued*)

Objective	Method	Results	Reference
	• Scenario 3: Centralized water and urine- diversion toilet with septicScenario 4: Centralized water and digester with GW non-potable reuse	- Eutrophication potential = 6.1×10^{-2} kg N/day/household - GWP = 1 kg CO_2-eq/day/household - Energy use = 0.88–1. 00 MJ/day/household	
Comparison of three **GW treatments:** • photocatalysis • photovoltaic solar-driven photocatalysis • membrane biological reactor	LCA cradle to gateImpact 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 **GW** photocatalytic fuel cell (PFC) with ZnO/Zn photoanode and CuO/Cu photocathode	- X-ray diffraction (XRD), - field-emission scanning electron microscopy (FESEM) - energy dispersive X-ray (EDX) - Fourier transform infrared (FTIR) spectroscopy	PFC with ZnO/Zn photoanode and CuO/Cu photocathode: effective GW as well energy recovery	(Kee, et al., 2018)
Qatar: environmental comparison of conventional water treatment, desalination and **GW reuse**	Existing academic and grey literatures on greywater in Qatar	GW can replace more expensive conventional water treatments and desalinated resources	(Lambert and Lee, 2018)
Evaluation of on-site separation of black water from **GW and onsite reuse**	Critical analysis of risks assessment and scientific papers	- GW use for toilet flushing and irrigation may benefit users for nutrients and the water saving - GW might pose health and environmental risks, which have to be quantified and standardised - Reuse of GW should consider local conditions and intended usage. - Technology and regulations should be routinely audited to mitigate potential risks	(Maimon and Gross, 2018)
Evaluation of mitigate water scarcity in urban areas by means of decentralised rainwater harvesting, **GW recycling**, and **hybrid rainwater-GW systems**	Rain-TANK model	- Domestic and commercial rainwater systems supply >90% and <43% of non-potable demand (NPD). - Domestic and commercial GW supply >92.1% and >36.2% of NPD. - Hybrid systems produce higher water savings than rainwater or GW alone >95% of NPD	(Oh, et al., 2018)
Economic and environmental evaluation of **GW** according to boundary system and functional unit	- Critical analysis of papers from 1990s to 2016 - LCA cradle to grave/cradle-to-gate/ - LCA m^3 influent/m^3 effluent, tons of dry solids	- LCA was used to determine the impact of wastewater treatment and the technologies used. - GW reclamation and reuse positive contributes to environmental benefits, while from economic perspectives depends on the adopted technology of treatment	(Sabeen, et al., 2018)
Political + Economic + Social + Environmental GW reuse in Circular economy perspectives	- Energy recovery evaluation - Economic plan - Water supply	- Economic perspective is the highest barrier to actual development of GW reuse. - Holistic Approach: LCA + LCC + LCSC is needed to evaluate GW reuse sustainability. - Water recovery is the pivoting parameter for on-site resource recovery	(Sgroi, et al., 2018)
Evaluation of annual probability of infection for non-potable exposures to distributed **GW** and domestic wastewater treated with aerobic membrane bioreactor (MBR) + chlorination	Monte Carlo approach captured variation; Reference pathogens: *Norovirus*, *Rotavirus*, *Campylobacter jejuni*, and *Cryptosporidium* spp.	The predicted 95th percentile annual risks for non-potable indoor reuse of distributed GW and domestic wastewater at district and building scales were lower than the selected health benchmark of 10^{-4} infections per person per year for all pathogens except *Cryptosporidium* spp.	(Schoen, et al., 2018)
Comparatively evaluation of life cycle costs and expected monetary benefits of decentralized GW reuse	- LCA cradle to grave - LCC - Sensitivity analysis	- GW reuse LCC: 44.4% capital costs + 46.4% operational energy costs - Conventional wastewater treatment LCC: 39.9%, operational energy costs + 25.5% land use costs + 24.3% piping capital costs	(Yerri and Piratla, 2019)

HLR = 1000 L/m^2/day (Masi et al., 2016) is likely underestimated because it is only related to removal of 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^2/day (Svete, 2012) can be attributed to the absence of anaerobic conditions that hampered denitrification. On the other hand, Eregno et al. (2017) found high treatment efficiency (~80%) with high HLR (600 L/m^2/day). Despite the slight decrease in efficiency with increasing HLR, it is concluded that high HLR values are not necessarily detrimental for TN removal provided that the system configuration ensures proper conditions (i.e., anaerobic conditions and carbon availability). As to total phosphorus, Fig. 4e shows no clear trend between HLR and TP removal. High removal efficiencies (>70%) were obtained up to 700 L/m^2/day, suggesting that TP removal was mainly governed by fast processes (e.g., sorption) and hence stressing the importance of the proper choice of growing substrates.

Removal efficiency of microorganisms in Fig. 4f does not exhibit any clear correlation between pathogen removal and HLR. Efficiencies higher than 2 log units were found even at high HLR, suggesting that 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_5, 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 acceptable removal efficiencies. In terms of OLR, it is possible to notice that applications with high removal performances refer to values of organic loading rate up to 100 g_{COD}/m^2/day.

6. Environmental assessment of greywater treatment processes

GW reuse has both positive and negative environmental impacts (Vuppaladadiyam et al., 2018). The 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 areas can decrease of about 30% water consumption in buildings (Memon et al., 2007) and GW reuse in multi-story buildings in Israel may save 150 Mm^3/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 (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 risks are due to the presence of pathogens from faecal contamination, skin, food preparation and mucus, such as E. coli, Pseudomonas aeruginosa, Rotavirus, Legionella spp., Salmonella spp., Staphylococcus aureus (Blanky et al., 2015).

Table 12 summarises 30 Life Cycle Analyses and environmental assessment studies performed in the last decades to evaluate pros and cons of GW reuse according to GW origins, treatment technology and reuse purpose. Vertical subsurface flow constructed wetlands represent a valid mitigation tool for greenhouse gas 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 transportation stage (up to ~19 kg CO_2-eq/PE/y less), treatment stage (up to ~40 kg CO_2-eq/PE/y less), and sludge handling. A considerable reduction in energy consumption can be obtained by switching from centralised WW reuse systems (1.224–1.914 kWh/m^3) to decentralised WW and GW reuse systems (0.246–0.970 kWh/m^3), with a reduction of up to ~80% (Chang et al. 2017). Another remarkable advantage of WW and GW reuse systems compared to conventional WWTP is the reduction of the amount of treated WW released into natural water bodies, with a consequent decrease of contaminant loads and concentrations (Chang et al. 2017).

Several advantages can be obtained from GW reuse of different origin. For example, do Couto et al. (2013) demonstrated that the quality of airport-produced GW is similar to household GW and can be easily treated for reuse purposes. This led to large water savings (-40%) and cost reductions (−25%), in addition to the other environmental benefits. Sgroi et al. (2018) discussed how different political, decisional, social, economic, technological, and environmental factors should be considered for a sustainable water reuse implementation, stating that a holistic approach is needed to evaluate GW reuse sustainability in a circular economy prospective. From a general overview, what emerges is that even though results are sometimes site-specific – particularly in terms of net economic benefits – most studies showed that GW treatment with decentralised approaches are more beneficial than conventional treatments in terms of net energy consumption and economic costs.

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).

CRediT authorship contribution statement

Fulvio Boano: Conceptualization, Methodology, Supervision, Writing - original draft, Writing - review & editing. **Alice Caruso:** Writing - original draft, Writing - review & editing. **Elisa Costamagna:** Writing - original draft, Writing - review & editing. **Luca Ridolfi:** Writing - original draft, Writing - review & editing. **Silvia Fiore:** Conceptualization, Methodology, Supervision, Writing - original draft, Writing - review & editing. **Francesca Demichelis:** Writing - original draft, Writing - review & editing. **Ana Galvão:** Writing - original draft, Writing - review & editing. **Joana Pisoeiro:** Writing - original draft, Writing - review & editing. **Anacleto Rizzo:** Writing - original draft, Writing - review & editing. **Fabio Masi:** Writing - original draft, Writing - review & editing.

Declaration of Competing Interest

The authors declare that there is no conflict of interest regarding the publication of this article.

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Authors' contributions

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