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Conditions

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Assessment of the Treatment Performance of an Open-Air Green Wall Fed with Graywater under Winter Conditions

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ABSTRACT: Graywater (GW), i.e., the portion of household wastewater that excludes toilet flushes, is an interesting wastewater type because it requires only mild treatment. Green walls have been proposed as example of a nature-based solution for GW treatment due to low energy requirement and high ecological/societal benefits; however, indications about their treatment performances remain limited. This work presents experimental results of a laboratory modular green wall for GW treatment. Experiments have been performed outdoors during the winter season for three months. Each panel included four vertical columns of planted pots, and it was fed with 100 L of synthetic GW per day. Removal efficiencies were as follows (average values): 40% chemical oxygen demand, 97% biochemical oxygen demand, 61% total Kjeldhal nitrogen, 56% NO_3^--N, 57% total phosphorus, 99% *Escherichia coli*, and 63% anionic surfactants. This work proved the potential of an open-air green wall for treating GW, even under challenging conditions for biological treatment processes and with high hydraulic loading rates.

KEYWORDS: green wall, graywater, reuse, nature-based solution, treatment
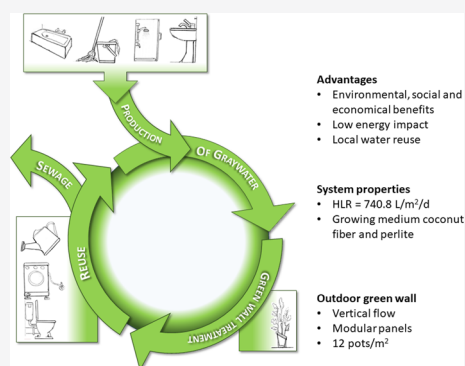
1. INTRODUCTION

United Nations Sustainable Development Goals (UN SDGs) 6.3 and 6.4 synergically promote novel strategies for overcoming the water stress scenario at the global level, giving safe water reuse, even at the household level, an essential role in reducing freshwater withdrawals and improving water use efficiency. This is especially relevant in areas where water scarcity is already a reality, such as Cape Town, in South Africa, or Viseu, in Portugal,[1] but these scenarios might become more frequent in the coming decades due to climate changes, with the Mediterranean being one of the most critical areas.[2] Alternative water sources should be actively sought, so that the urban water cycle will become a true cycle, and reclaimed wastewater reuse will proficiently contribute to the circular economy and to the achievement of UN SDGs.

Graywater (GW) recycling is a pillar in creating a new model of urban water supply, integrating this new source with rainwater-harvesting practices.[3,4] GW, discharged from sinks, showers, and washing machines (i.e., wastewater without the sanitary components, fecal matter, urine, and toilet paper), causes a low level of pollution, is relatively highly biodegradable, and is widely available in urban areas; indeed, GW represents approximately 70% of domestic wastewater in Europe, North America, and Asia, with a production range of 72−225 L/day per capita.[5]

Nature-based solutions (NBS) provide interesting preliminary results in GW treatment. Constructed wetlands (CWs) are low-cost NBS for wastewater treatment and have also been used to successfully treat GW.[6−9] However, the surface needed for CWs is scarce in urban areas; thus, green walls have been proposed for GW treatment to save horizontal space.[10] In this way, green walls simulate vertical flow CWs, in which GW infiltrates through the porous media that support the biofilm and plants, and the drained water can be further reused for nonpotable uses (toilet flushing, garden irrigation, street washing, etc.).[11−13] Moreover, green walls can also be designed to simulate horizontal flow CWs.[10] There are several types of green walls, with different water needs and support media for the plants, with systems simulating an assembly of pots, while others use fabric mats or similar materials to support the plants.[14] All of these configurations provide multiple benefits, such as high thermal/energy performance, air quality improvement, urban heat-island mitigation, and better quality of life.[10,15,16]

The use of green walls to produce reclaimed water has several advantages compared to wastewater treatment plants; the decentralized approach represents a sustainable strategy,

reducing the load on existing wastewater treatment plants, while also maintaining all of the mentioned benefits of traditional green walls.

The possibility of exploiting green walls for GW treatment and reuse is still in the preliminary stage, with few studies available in the literature. Most studies focused on the treatment performance of different filter media and plant species, analyzing the removal of organic matter and nutrients. Considering the importance of filter media, preliminary and pilot studies were performed, suggesting that mixes with coconut fiber guarantee a favorable hydraulic retention time (HRT) for removing pollutants and avoiding clogging.[12,17,18] It was also observed that expanded clay mixes provide good removal results,[12,19] while Fowdar et al.[20] realized good performances with a sand-based medium. Plant selection is also an important design aspect, even if their influence on treatment performance is still being debated. Some studies[20−22] found a strong dependency of nitrogen and phosphorus removal on plant species, probably due to the development of roots and the rate of growth of plants, indicating the importance of the correct choice of plants. An extensive study[22] with 13 plants based on a perlite/coconut coir mix (1:2 ratio) reported 88% TN removal, with slight variation among species, while total phosphorus (TP) showed a stronger dependency on plant species, with removal efficiencies of 17−53%. However, a recent study[23] tested ornamental plants usually used in green walls and found no significant difference in TP, BOD$_5$, or COD removal efficiency, although plants showed different levels of well-being under high-moisture conditions. An even more limited number of studies addressed the removal of biological pollutants of green walls in GW treatment. Svete[11] reported *Escherichia coli* removal of 2 logs for an inflow concentration of >2 × 10^4 MPN/100 mL, while Prodanovic et al.[18] reported a maximum *E. coli* removal of 1 log from synthetic GW. These results are in line with those obtained in constructed wetlands (its parent technology) treating GW, also in full-scale systems.[24] The mentioned studies have demonstrated the potential of green walls for GW treatment. However, the current understanding of how GW treatment performance is influenced by the configuration (e.g., filling media and plants) and operating conditions (e.g., flow rate and climate) of a green wall is still far from complete. More studies are hence necessary to quantify these performances over a wide range of experimental conditions.

This work aims to investigate GW treatment performances of a pot-based green wall under conditions that are more challenging compared to more controlled laboratory and pilot studies,[10,11,18,20−22,25,26] i.e., considering open-air winter conditions and a high hydraulic loading rate (HLR). After preliminary tests on plants and filter media, experiments were performed at pilot scale on an outdoor green wall fed with synthetic GW, operating for three winter months (January to April in the northern hemisphere and in a continental climate area), when low temperatures may hinder biological removal of pollutants. The system was fed with high flow rates of GW to minimize the required space, reaching a considerable HLR compared to those of common practices. Treatment performances were quantified for a wide array of physicochemical and biological parameters. The presented pilot-scale green wall included a modular outline, thus providing easy scaling-up possibilities.

2. MATERIALS AND METHODS

2.1. Selection of the Filter Medium. A mix of coconut fiber (CF) and perlite (PL) was used as the filter medium, and preliminary tests were performed to choose the mix composition. According to previous studies,[17] CF allows better removal performances and longer HRTs, while PL limits clogging and reduces the weight of the medium. Different ratios of CF and PL were analyzed (volume percentages of 60% CF and 40% PL, 70% CF and 30% PL, 80% CF and 20% PL, 90% CF and 10% PL, and 100% CF and 0% PL) to identify a good compromise between hydraulic conductivity and overall weight.[17,18] Triplicate preliminary tests were performed on plexiglass columns (diameter, 0.06 m; height, 0.5 m; mix layer height, 0.3 m) to analyze the hydraulic behavior of the different filter medium mixtures without plants. For each mix, the porosity, particle density, and bulk density were measured, weighting each column before and after saturation with water. The saturated hydraulic conductivity (K_s) was evaluated using Darcy's falling head method.

2.2. Selection of Plants. After a literature overview,[10,20,27−30] five plant species (*Hedera helix, Carex morrowii, Iris germanica, Lonicera nitida,* and *Ranunculus asiaticus*) were selected in this study for their tolerance to climatic conditions and high soil moisture, limited space for root growth, plant size, aesthetic appearance, and local availability. These species were pretested under GW irrigation conditions (3−12 mm/day per pot, fed 5 days per week) for four months to choose the ones to be later installed in the green wall. A control group of the five plant species was irrigated with the same amount of tap water (TAP). Plant resistance was evaluated in terms of leaf health and appearance of new sprouts.

2.3. Synthetic Graywater. Domestic GW is highly variable in composition among and within countries in the world, due to the heterogeneity of the habits of the people and the use of commercial products.[24,31,32] In this study, synthetic light GW was prepared according to reference doses[32] based on easily available detergents and personal care products (see the Supporting Information). *E. coli* tablets (Ielab BAControl) were used, instead of the secondary effluent prescribed by reference doses, to guarantee the presence of microbiological pollutants.

2.4. Experimental Setup. A pilot-scale system made of four modular units (Figure 1) was built on the northeast facing wall of the Hydraulic Laboratory courtyard in Politecnico di Torino. The green wall was fed with a volume of 96 L/day per modular unit, close to the mean daily production of GW per capita in developed countries.[33] Each modular unit was a 1 m^2 metallic panel hosting 12 pots (three rows, four columns), with each column working as an independent vertical flow system. Each pot (18 cm × 18 cm × 22 cm, bulk volume of 6.5 L) was filled with a 0.2 m layer of filter medium and planted with one of the selected plant species. For each column, the plant type changed along the three rows to increase biodiversity, reduce the risk of phytodiseases, and improve the aesthetic appearance.[10] Three replicated columns were fed with GW, and three others with TAP. GW was prepared every 2 days in a 1.5 m^3 plastic tank mixed hourly by an automatic recirculation system. GW was pumped in a pressurized feeding system of plastic pipes and drippers (one per column) with a flow rate of 4 L/h. A separate piping system was fed with TAP. Each column was fed in batch mode (1 L flush for 15 min, followed by 45 min of resting time), like vertical flow constructed

Figure 1. Modular panel of the green wall. All columns (marked in yellow) were identical except for the type of irrigation, and each worked independently. Three columns of this panel were fed with graywater (GW), and one was fed with tap water (TAP).

wetlands to promote aerobic degradation.[34] In each column, water flowed vertically by gravity from the top pot to the ones below (middle and bottom) through 4 mm plastic tubes. At the bottom of the bottom pots, the plastic tubes, also used for sampling, were used to discharge water into 10 L tanks and then to the sewer system. The HLR calculated with the horizontal cross section area of the pots in the first row was 740.8 L m^{-2} day^{-1}. This value is much higher than the values of daily precipitation recorded during the sampling period (0−30.8 L m^{-2} day^{-1}), and it falls at the highest range of HLRs usually employed in both vertical CWs and green walls for GW treatment.[10,24,34]

All pots on the green wall panels were preliminarily washed with TAP (see the Supporting Information) before the experiments to verify the leaching potential of the filter medium and to remove the finest particles that could clog the system.

2.5. Sampling and Physicochemical and Microbiological Analyses. GW irrigation of the panels started January 8, 2019, and sampling operations happened weekly in the first two months (January 16, 23, and 30 and February 6, 13, and 27), which is consistent with previous studies,[9,12,23] to detect possible transient phenomena and twice per month in the last period (March 6 and 20 and April 2), nine samplings in total. The temperature, pH, electric conductivity (EC), total dissolved solids (TDS), and dissolved oxygen (DO) were analyzed through a WTW Multi 3320 portable two-channel probe equipped with specific sensors and/or electrodes. The total suspended solids (TSS) was analyzed by filtering 1 L through 0.45 μm cellulose membranes. The sulfate, chloride,

total Kjeldhal nitrogen (TKN), nitric nitrogen (NO$_3^-$-N), ammonia nitrogen (NH$_4^+$-N), total phosphorus (TP), chemical oxygen demand (COD), and methylene blue active substances (MBAS, i.e., anionic surfactants) were analyzed through Nanocolor reagents, a VELP COD ECO 16 thermoreactor (for total nitrogen, total phosphorus, and COD), and a model AL450 Multidirect photometer. BOD$_5$ was analyzed through a VELP FOC 215E cooled incubator equipped with 24 BOD sensor systems. *E. coli* was analyzed through Colitag water test reagents after 24 h according to EPA Standard Method 9221.

2.6. Statistical Analysis. A two-tailed nonparametric Mann−Kendall test was performed on time series of removal efficiencies to verify the presence of significant monotonic trends over time. The test was applied to analyze removal efficiencies for both TAP and GW columns. The interpretation of this test depends on the number of samples. In this study, the nine collected samples resulted in a threshold for the test statistic (|S|) of 17 (α = 0.05). When |S| was lower than this threshold, the measurements obtained over time were considered independent and no significant temporal trend was detected.

3. RESULTS AND DISCUSSION

3.1. Preliminary Tests. *3.1.1. Selection of the Filter Medium.* Table 1 compares coconut fiber (CF) and perlite

Table 1. Physical Characteristics of Coconut Fibers (CF) and Perlite (PL)[a]

material	porosity	particle density (g/cm^3)	bulk density (g/cm^3)
100% CF	0.652 (0.009)	1.023 (0.046)	0.355 (0.006)
100% PL	0.583 (0.013)	0.362 (0.103)	0.150 (0.013)

[a]Average and standard deviation values were obtained from three replicates, with the standard deviation in parentheses.

(PL) in terms of porosity and density. The particle density of CF is almost 3 times higher than that of PL, and the bulk density is more than double. Perlite is less dense than water; thus, it floats. CF adsorbs water and reaches a particle density around that of water (0.997 g/cm^3).

Figure 2 shows the comparison among different CF/PL mixes in terms of hydraulic conductivity (K_s) and bulk density (ρ_b). As expected, K_s strongly increased with PL content, because CF are hydraulically slower than PL.[17] The average

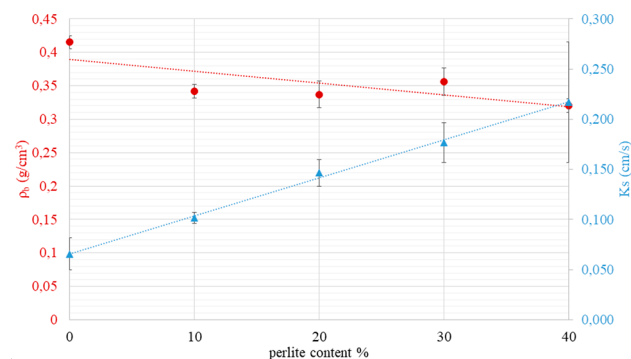**Figure 2.** Average values and standard deviations of hydraulic conductivity [K_s (blue triangles)] and bulk density [ρ_b (red circles)] for mixes with different percentages of coconut fibers and perlite.

value of K_s varied between 0.066 ± 0.016 cm/s for pure CF and 0.217 ± 0.060 cm/s for the 60% CF/40% PL mix, showing a strong linear correlation between K_s and PL content ($R^2 = 0.998$). The average ρ_b decreased with PL content ($R^2 = 0.577$), varying between 0.415 ± 0.01 (pure CF) and 0.320 ± 0.01 (60% CF/ 40 % PL mix). The slopes of the trend lines in Figure 2 indicate that the hydraulic conductivity was more sensitive to PL content than ρ_b.

While an increase in PL content is positive in terms of reducing the weight of the filter medium, the increase in K_s may not provide adequate contact time for the removal of contaminants. On the basis of the results presented here, the 80% CF/20% PL mix was chosen for the experiments as it allowed us to reduce ρ_b while matching the typical values of K_s of porous media employed in constructed wetlands with good removal performances.[35] According to the authors' experience, this choice was regarded as a good compromise between weight (0.337 ± 0.02 g/cm^3) and hydraulic conductivity (0.146 ± 0.013 cm/s).

3.1.2. Selection of Plants. During the preliminary tests, *Ranunculus* and *Iris* showed low resistance to moisture and were discarded. *Lonicera*, *Carex*, and *Hedera* resisted high moisture, temperature oscillations, and sun exposure (Figure 3). For these three species, qualitative observations on leaf

Figure 3. Tested plant species after one month: (a) *L. nitida*, (b) *I. germanica*, (c) *C. morrowii*, (d) *R. asiaticus*, and (e) *H. helix*. Red (solid) and blue (dashed) lines indicate the graywater (GW) and tap water (TAP) groups, respectively.

health and the appearance of new sprouts showed no particular difference in health conditions or growth between groups fed with GW or TAP. The results of the preliminary tests hence indicated that GW had no negative impact on *Lonicera*, *Carex*, or *Hedera*, each of which was selected for the green wall.

3.2. Green Wall Performances. Figure 4a shows average temperature values of TAP and GW at the inlet and outlet of the green wall. The recorded temperature range is comparable with winter experiments in greenhouses (minimum value of 4 °C)[25] and obviously lower than summer temperatures (maximum values of 44 °C[25] and 45 °C[20]). Inlet average pH values (Figure 4b) were neutral for TAP (7.01) and slightly alkaline for GW (7.35); the outlet pH was 7.24 ± 0.252 for TAP and 7.40 ± 0.107 for GW, both values within the recommended range of 6–7.5 for plant growth[26] and within the optimal range of 6.5–8.5 for nitrogen removal.[36] EC showed the same increase trend from inlet to outlet (Figure

4c), even if inlet and outlet values were similar in the last samples for both TAP and GW. DO values (Figure 4d) were similar for output TAP and output GW due to the aerobic conditions of vertical flow systems that resulted in oxygen concentrations close to saturation (10–13 mg/L for the observed temperatures) and promoted aerobic reactions in the columns. Inlet GW showed a strong DO decrease over time due to an increase in the outdoor temperature, possibly because of aerobic reactions occurring in the storage tank. Due to insufficient mixing in the storage tank, the average TSS concentration in inlet GW was low (8.56 mg/L). The average outlet TSS concentration from GW columns (9.37 ± 0.96 mg/L) was in line with common outflow values from vertical constructed wetlands with similar TSS loads (6–60 mg/L),[37] suggesting that lower outflow concentrations cannot be attained for this TSS load. Chloride and sulfate (see the Supporting Information) exhibited a slight release in TAP columns but exhibited rather conservative behavior in GW columns.

Figure 5a shows average COD concentrations at the inlets and outlets of the green wall columns along the sampling period. Despite the prewashing (see the Supporting Information), the filter medium released an average of 47.63 mg of COD/h, as shown by the comparison between inlet (67.67 mg/L) and outlet (122.00 ± 4.93 mg/L) concentrations of TAP samples. In contrast, COD was always removed in GW-fed columns with removal rates in the range of 12.33–184.00 mg/h. The removal efficiency significantly increased over time from 4.7% to 82.3% ($|S| = 32$), possibly due to a decreasing rate of release from the filter medium, an increase in temperature, and/or progressive biofilm growth.[12,17] This trend agrees with the literature findings where after an establishing phase COD removal performances increased up to 86%.[12] On average, this study found 40.4% COD removal efficiency in GW columns (102.67 mg/h average removal rate), similar to the range of 46.0–49.3% in ref 38 but lower than the values of 92.4% in ref 26, in which sampling started after 16 weeks, and 95% in ref 19. These results indicate a good efficiency for the removal of COD from GW, within the range observed in laboratory studies of green walls.

Figure 5b reveals a low rate of release of BOD$_5$ from the filter medium in TAP columns (inlet, 0.43 mg/L; outlet, 1.18 ± 0.33 mg/L). Despite this release from the organic filter medium, the release of BOD$_5$ was offset by removal processes in GW columns. Indeed, the BOD$_5$ removal efficiency in columns fed with GW ranged between 96.1% and 99.3%, exhibiting a performance at the higher range of previous studies showing 86–98%.[12,19,20] As opposed to COD, the BOD$_5$ average removal efficiency (97.65 ± 1.34%) did not change over time, suggesting a fast removal by physicochemical processes (e.g., sorption on the filter medium) followed by biological degradation under favorable aerobic conditions.[34]

The COD:BOD$_5$ ratio in inlet GW was 5.25 ± 1.61, higher than those of typical real GW that are between 1.05 and 3.86[24] but within the upper boundary of 6.98 for synthetic GW.[39] The average value for outlet GW was much higher (182.82 ± 177.18) and 2 orders of magnitude higher than values (2.3–3.6) reported in other studies.[12,19] This difference probably derives from the limited removal of COD during the establishment period.

Figure 6 shows the concentration of nitrogen species (TKN and NO$_3^-$-N) and TP. A release of TKN from TAP columns
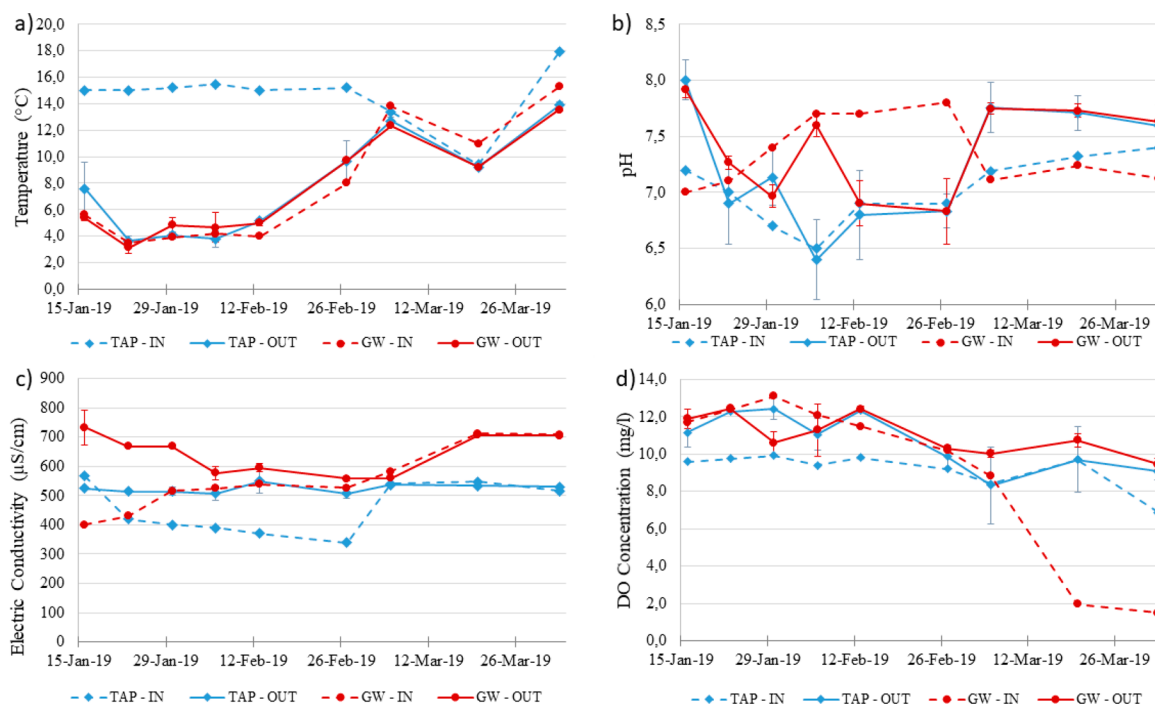
Figure 4. (a) Temperature, (b) pH, (c) electric conductivity (EC), and (d) dissolved oxygen (DO) at the inlet and outlet of TAP and GW columns.
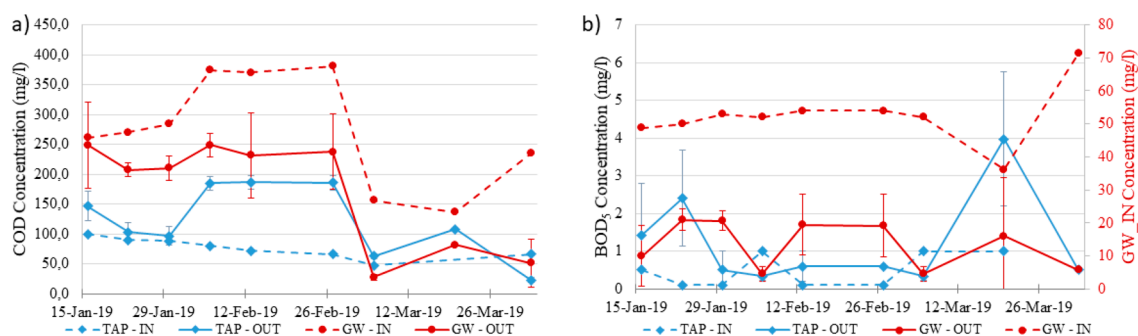

Figure 5. Concentrations of (a) COD and (b) BOD$_5$ at the inlet and outlet of TAP and GW columns.

(inlet, 2.42 mg/L; outlet, 4.34 ± 0.19 mg/L) occurred (Figure 6a), possibly due to the organic components of CF, but a removal in GW columns (inlet, 5.01 mg/L; outlet, 2.66 ± 0.14 mg/L) was observed. TKN was mainly composed of organic nitrogen due to the very low NH$_4^+$-N concentrations (mostly <1 mg/L) in TAP and GW inlets. The TKN removal efficiency (Figure 6a) in GW samples strongly increased over time (|S| = 28), to 92.7 ± 2.4% (in agreement with the value of 94% in ref 19), possibly due to the progressive development of biological processes (e.g., microbial and plant uptake[21]) favored by an increase in temperature and system growth, with an average removal equal to 34.63 ± 29.67%. NO$_3^-$-N was released from the filter medium in TAP columns (inlet, 1.94 mg/L; outlet, 2.19 ± 0.15 mg/L) (Figure 6b); however, GW columns removed NO$_3^-$-N up to 55.6 ± 13.7% (average rate of 25.65 ± 28.80%). Literature results vary significantly, with removal efficiencies as high as 98.6%;[26] however, a strong (440.3%) release was also reported in a study[19] with a NO$_3^-$-N outlet concentration (3.61 mg/L) close to that of the study presented here. The observed NO$_3^-$-N removal could be caused by denitrification in anaerobic microzones inside the pots and by plants and microbial uptake. As inlet concentrations were

limited and vertical flow systems are not specifically designed for NO$_3^-$-N removal, the observed removal performances for NO$_3^-$-N obtained in this work were considered satisfactory.

TP (Figure 6c) was characterized by low inlet concentrations (<1.70 mg/L), compared to values in the literature for real and synthetic GW (0.01−51.58 mg/L).[24,26,40] TP concentrations varied negligibly in TAP columns, while GW columns showed a positive removal efficiency with a slightly increasing trend (|S| = 18), as also observed by Prodanovic et al., possibly due to plant uptake, microbial activity, and sorption,[22,41] and favored by the increasing temperature.[9] The average output concentration (2.74 ± 0.25 mg/L) and removal efficiencies (−22% to 56.7%) are consistent with previous studies (1−2.6 mg/L and 35−72.6%, respectively, on average).[20,22,26,38]

The *E. coli* average removal efficiency (Figure 7a) was excellent (98.9 ± 1.8% and 2.74 ± 1.55 log units) and significantly improved over time (|S| = 20), ensuring a very low average concentration in the output (25 MPN/100 mL) compared to that of another vegetated pot system with values of 2.72 × 10^3 MPN/100 mL[26] or 0.3−0.6 log unit.[23] Hence, a single modular panel reduced the *E. coli* concentration by ≤4
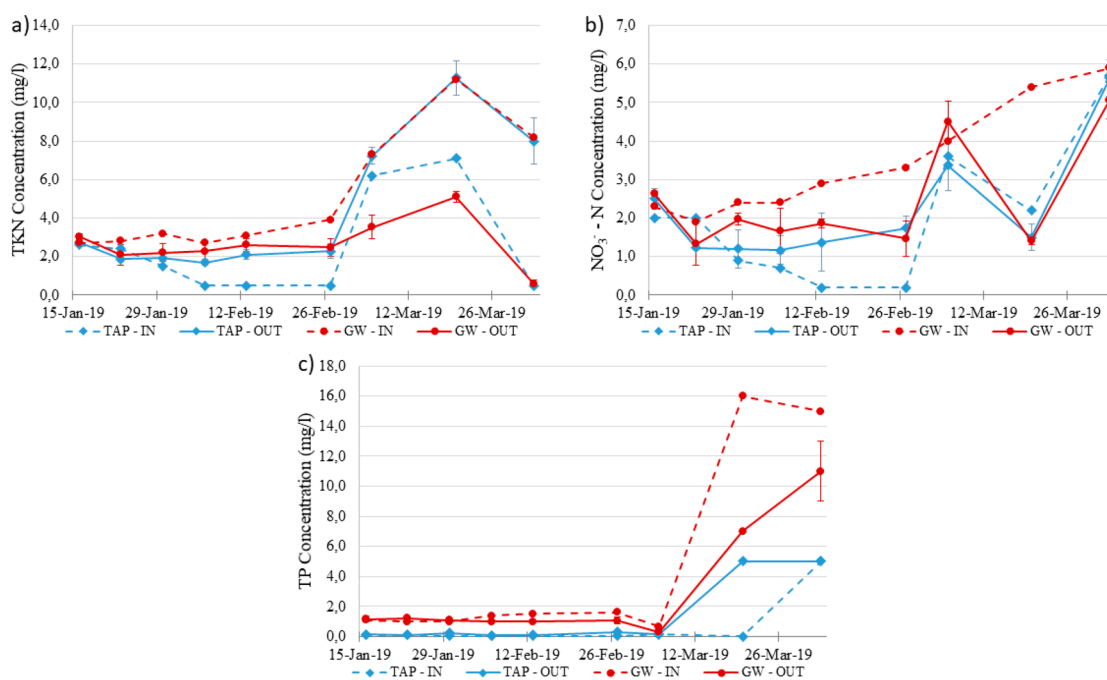
Figure 6. Concentrations of (a) TKN, (b) NO_3^--N, and (c) TP at the inlet and outlet of TAP and GW columns.
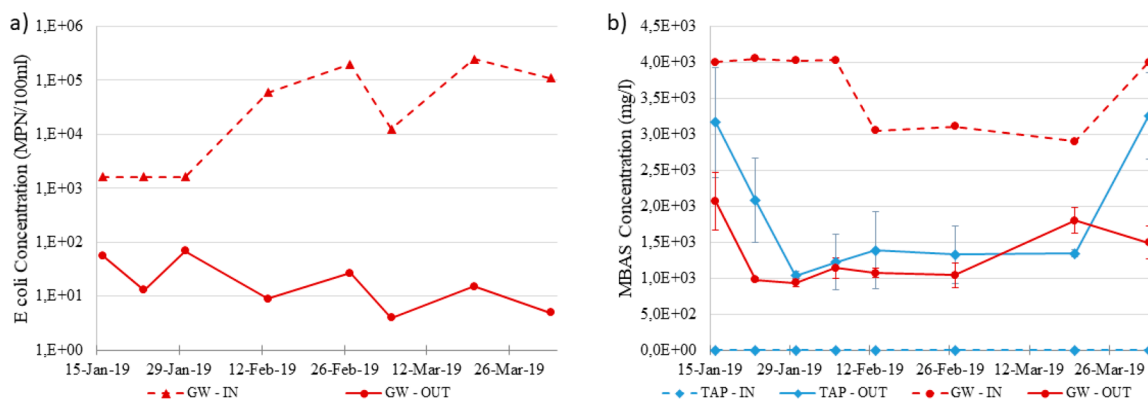

Figure 7. Concentrations of (a) *E. coli* in the input and output of GW columns and (b) MBAS at the inlet and outlet of TAP and GW columns.

orders of magnitude. Figure 7b indicates that MBAS were released from TAP columns for all samples (inlet, 0.15 mg/L; outlet, 1852.38 ± 171.46 mg/L), probably because of the leaching of natural organic substances from the filter medium that react during MBAS analysis. However, along GW columns, MBAS concentrations decreased remarkably, and output GW concentrations reached the values of output TAP. Thus, the system achieved very good performances (63.0 ± 13.6% average removal efficiency) in removing anionic surfactants from GW.

4. CONCLUSIONS

This study evaluated the treatment performance of an outdoor green wall with ornamental plants, treating synthetic domestic graywater for nonpotable reuse. Preliminary tests led us to choose a mixture of coconut fiber (80%) and perlite (20%) for filter media and to identify ornamental plants resistant to graywater. The system showed good tolerance to high HLR and continental winter conditions for the three months of monitoring. BOD_5 and *E. coli* removal were excellent (97.65 ± 1.34% and 2.74 ± 1.55 log units, respectively, on average).

COD and TKN increased over time to 82.3% and 92.7%, respectively, possibly due to favorable oxidizing conditions, the development of roots and biofilm, and an increase in temperature. NO_3^--N and TP showed good removal efficiencies, even at low concentrations as per TP. These performances are similar to those obtained in previous laboratory studies, confirming the potential of green walls to treat graywater even in outdoor winter conditions with high HLR. It should be noted that the work presented here investigates a specific set of design conditions, and GW treatment performances are expected to change if different green wall types and configurations are considered. Hence, further studies are needed to improve our understanding of how GW treatment performance is influenced by the type of filter medium and the operating conditions of green walls.

■ ASSOCIATED CONTENT

ⓢ Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsestwater.0c00117.

Synthetic graywater recipe and characteristics (section S1 and Table S1), preliminary washing procedure (section S2), tap water volumes and sampling times of the preliminary washing procedure (Table S2), compounds that were significantly released from the columns during the preliminary washing procedure with tap water assessed by (a) pH and electrical conductivity and (b) NO_3^--N, SO_4^{2-}, and Cl^- concentrations (Figure S1), sulfate and chloride results (section S3), and concentrations of (a) sulfate and (b) chloride at the inlet and outlet of TAP and GW columns (Figure S2) (PDF)

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Author Contributions

Conceptualization, methodology, and supervision: F.B. and S.F. Writing of the original draft: E.C. and F.B. Experimental activity: E.C., A.C., and F.D. All of the authors reviewed the manuscript.

Notes

The authors declare no competing financial interest.

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