Water Science & Technology

© 2023 The Authors

Water Science & Technology Vol 88 No 11, 2751 doi: 10.2166/wst.2023.389

Assessing the efficacy and mechanisms of glycol-contaminated water treatment through floating treatment wetlands

Tao Lyu *, Randy Williams, Benjamin Exton and Robert C. Grabowski

School of Water, Energy and Environment, Cranfield University, College Road, Cranfield, Bedfordshire MK43 0AL, United Kingdom
*Corresponding author. E-mail: t.lyu@cranfield.ac.uk

TL, 0000-0001-5162-8103; BE, 0000-0001-8393-8667; RCG, 0000-0002-0926-1202

ABSTRACT

The growing concerns surrounding water pollution and the degradation of ecosystems worldwide have led to an increased use of nature-based solutions (NbSs). This study assessed the feasibility of using floating treatment wetlands (FTWs) as an NbS to treat propylene glycol-contaminated water and quantitatively investigated different removal pathways. With an environmentally relevant concentration of propylene glycol (1,250 mg/L), FTWs containing *Acorus calamus* and mixed species demonstrated the highest average glycol mass removal efficacy (99%), followed by *Carex acutiformis* (98%), *Juncus effusus* (93%), and the control group without plants (10%) after 1 week. Additional mesocosm-scale experiments with varying FTW configurations, including surface coverage to reduce evaporation and photodegradation processes, and the addition of antibiotics to inhibit microbial activity, were conducted to quantify glycol removal pathways. Mass balance analysis results revealed that microbial biodegradation (33.3–39.7%) and plant uptake (37.9–45.2%) were the primary pathways for glycol removal. Only 15.5–19.5% of the glycol removal via evaporation and photodegradation was accounted in this study, which may be attributed to the mesocosm experimental setup (static water and no wind). Aligned with the broader discussion regarding biodiversity improvements and carbon storage capacity, this study demonstrated that FTWs are an environmentally friendly and effective NbS for addressing glycol-contaminated water.

Key words: biodiversity, constructed wetlands, green technology, multi-benefits, propylene glycol, volatile organic compound (VOCs)

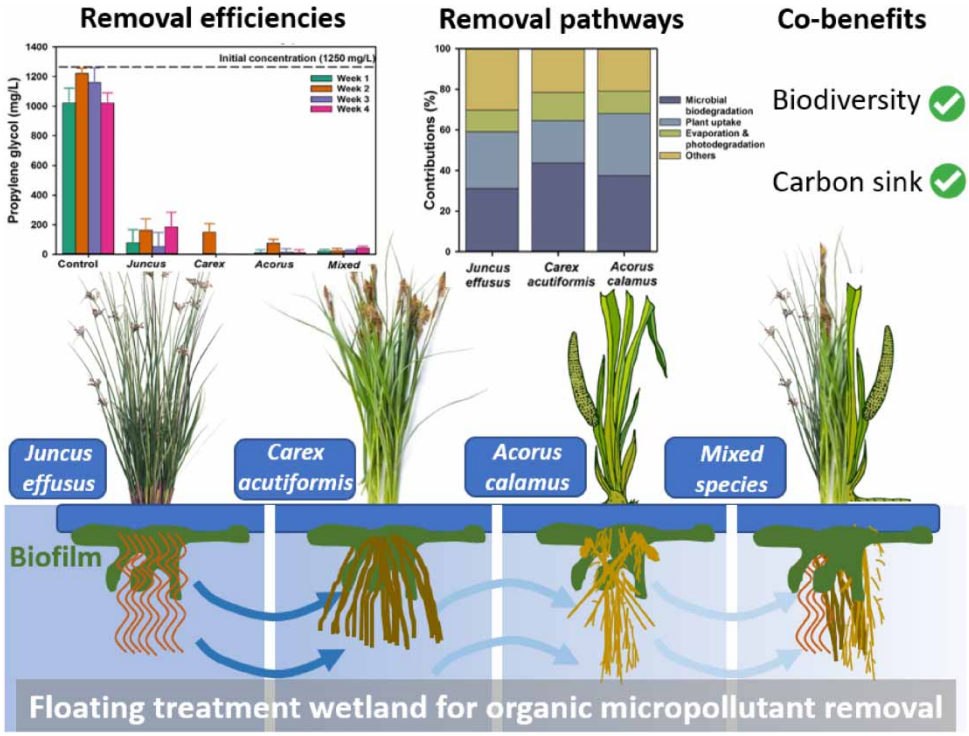
HIGHLIGHTS

- Floating treatment wetland (FTW) can effectively remove organic micropollutants.
- *Acorus* and *Carex* are more efficient to remove propylene glycol than *Juncus*.
- Microbial biodegradation and plant uptake are the major removal pathways.
- Mixed species in FTW can yield high removal efficiency and support biodiversity.

This is an Open Access article distributed under the terms of the Creative Commons Attribution Licence (CC BY 4.0), which permits copying, adaptation and redistribution, provided the original work is properly cited (http://creativecommons.org/licenses/by/4.0/).

Downloaded from http://iwaponline.com/wst/article-pdf/88/11/2751/1340103/wst088112751.pdf
by guest

GRAPHICAL ABSTRACT



1. INTRODUCTION

Constructed wetlands (CWs), as a nature-based solution, have emerged as an important approach for improving water quality while simultaneously delivering co-benefits such as biodiversity enhancement (Oral *et al.* 2020; Castellar *et al.* 2022). Floating treatment wetlands (FTWs) represent one of many diverse applications of CWs, in which emergent wetland plants grow hydroponically on mats or structures floating on the surface of a pond (Pavlineri *et al.* 2017). FTWs offer some advantages over other subsurface flow CWs, such as the absence of requirements for substrate materials, which significantly reduces construction costs and eliminates clogging risks because no substrate pores are present (Colares *et al.* 2020). Moreover, FTWs are advantageous as a stormwater management tool since they can fit easily into existing retention ponds and adapt to varying water depths characteristic of event-driven stormwater systems (Headley & Tanner 2011).

Previous studies have shown the efficacy of FTWs in reducing organics (COD and BOD_5), nutrients, TSS, and toxic metals (e.g. Cd, Cu, and Zn) in wastewater treatment (Afzal *et al.* 2019; Park *et al.* 2019; Colares *et al.* 2020). With increasing concerns about water pollution and stringent regulations, there has been a growing interest in the removal of emerging contaminants (Lv *et al.* 2016a; Zhang *et al.* 2017). One such pollutant is propylene glycol, which is widely used in the manufacture of polyester resins and formulation into functional fluids such as anti-freeze, aircraft anti-icing and de-icing fluids, cosmetics, pharmaceuticals, personal care products, and pesticides (West *et al.* 2014; Rogers *et al.* 2019). Although low concentrations of glycol are not directly toxic to aquatic organisms, elevated concentrations can lead to increased microbial activity that drives a decrease in oxygen levels, which can be very harmful to aquatic life (Freeman *et al.* 2015; Nott *et al.* 2020; Exton *et al.* 2023).

Subsurface flow CWs have demonstrated promising results in the removal of glycol from wastewater, achieving up to 99% removal under initial concentrations of over 1,000 mg/L (Higgins & Maclean 2002; Murphy *et al.* 2015). However, one of the main sources of environmental contamination of glycol is from de-icer applications, such as in airports, which enters into environment through weather-driven storm water runoff (Corsi *et al.* 2012; Nott *et al.* 2020). As such, FTWs have been tested and operated at Heathrow Airport to treat glycol, owing their ability to cope with varying water levels. The average glycol removal efficiency achieved 54% in the initial pilot-scale FTW (3×5 m) trial (Chong *et al.* 1999). However, limited published data were available regarding the treatment performance in their applied FTW system (Richter *et al.* 2003).

Downloaded from http://iwaponline.com/wst/article-pdf/88/11/2751/1340103/wst088112751.pdf
by guest

Therefore, despite numerous studies on the effectiveness of FTWs in removing nutrients and other pollutants (Afzal *et al.* 2019), research on their ability to eliminate glycol remains scarce.

In FTWs, elucidating the removal mechanisms and quantifying different removal pathways of the pollutants through biological processes (e.g. plant uptake and microbial biodegradation) and physicochemical processes (e.g. photodegradation and evaporation) are essential for determining optimal implementation strategies (Barco *et al.* 2021). Sriprapat *et al.* (2011) reported that the aquatic macrophyte (*Echinodorus cordifolius*) was able to uptake diethylene glycol while treating the wastewater. The selection of plant species can substantially impact treatment performance by modulating plant uptake capabilities (Oliveira *et al.* 2021). In terms of removing ethylene glycol from industrial wastewater, *Acorus calamus* demonstrated the best treatment performance when compared to *Phragmites australis* and *Typha latifolia* during phytoremediation experiments (Marecik *et al.* 2013). Apart from plant uptake, the plant rhizosphere provides a conducive environment, oxygen, and carbon sources (root exudates) for microbial growth (Lv *et al.* 2016b; Zhang *et al.* 2017), which can significantly contributes to the biodegradation of glycols (Higgins & Maclean 2002; Murphy *et al.* 2015). Previous FTW research highlighted that the mechanisms underlying nutrient removal in FTWs remain incompletely understood (Colares *et al.* 2020), and this knowledge gap also pertains to glycol removal. We hypothesize that FTWs can effectively remove glycol from contaminated water, and the evaporation process may play a significant role in removal, considering that glycol is a volatile organic compound (VOC). Consequently, a quantitative investigation of removal mechanisms is vital for future FTW implementations.

In this study, three native wetland species, namely *Juncus effusus*, *Carex acutiformis*, and *A. calamus*, and their mixed planting were employed in mesocosm-scale FTWs to evaluate the effects of plant species and planting strategies (monoculture vs. polyculture) on propylene glycol removal from the water column. Moreover, a separate experiment was conducted to quantitatively investigate the removal pathways of propylene glycol, including microbial biodegradation, plant uptake, and the combined effects of photodegradation and evaporation. Additionally, the role of FTWs in supporting the biodiversity and carbon sequestration was discussed.

2. MATERIALS AND METHODS

2.1. Wetland plant and chemicals

Three commonly used native wetland plant species, namely *J. effusus*, *C. acutiformis*, and *A. calamus*, were selected for this study due to their abilities to thrive in wetland environments and their potential for wastewater treatment applications. The leisis floating mat were purchased from a local botanic shop (Beaver Plants Ltd, Lingfield, UK). The propylene glycol and antibiotic sulfamethoxazole were sourced from Fisher Scientific and Scientific Lab Supplies, respectively.

2.2. Experimental setup and operation

The mesocosm-scale experiment was conducted over 6 weeks from 21 July to 1 September 2022, with the initial 2 weeks allocated for stabilisation to facilitate the growth of plants and biofilm. Prior to the experiment, the roots of all plants were washed, the wet biomass was measured and documented. To facilitate the transition of plants from soil to hydroponics, all cleaned plants were submerged in a 1% concentration of Formulex nutrient solution (Growth Technology Ltd, Somerset, UK) for a week. Healthy plants were selected for the experiment. The experimental setup was placed in a large glasshouse to maintain ambient environment while avoiding precipitation.

In this study, two experiments were conducted to assess the effect of planting strategies (monoculture vs. polyculture) on propylene glycol removal and the removal pathways, respectively (Figure 1(a)). The first experiment was carried out in 5-L buckets, each with a surface opening of approximately 0.016 m^2 (Figure 1(b)). Three replicates were used for each plant treatment, including *J. effusus, C. acutiformis, A. calamus* and a mixture of these species. Regardless of the plant species, 500–700 g of initial wet biomass was placed in each bucket. For mixed plant species, an equal wet biomass of each plant species was used in the system. In all systems, two floating mats, each measuring 5 × 5 cm, were used to hold the plants at the water surface (Figure 1(b)). Additionally, a control group was established without plants but with identical floating mats in each bucket. The initial volume of the growth solution was 4 L, and the water loss from each bucket were measured weekly along with the propylene glycol concentration. Water level and propylene glycol concentrations were checked and adjusted weekly in order to align with initial starting conditions.

The second experiment was conducted in smaller 1-L buckets, aimed at quantifying the different pathways involved in propylene glycol removal in FTWs planted with different plant species. Three different FTW configurations (groups) were established within this experiment. A common FTW setup was applied to group 1 (plant + floating mat), with the plant

Downloaded from http://iwaponline.com/wst/article-pdf/88/11/2751/1340103/wst088112751.pdf
by guest

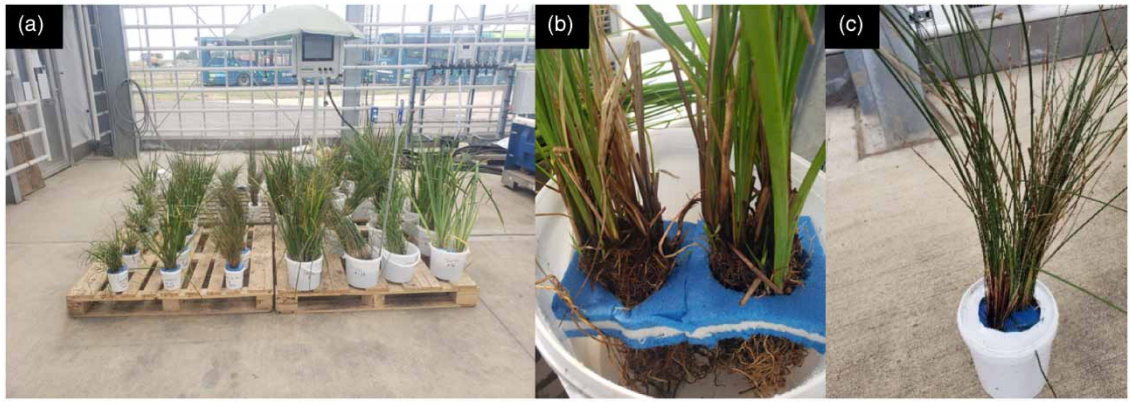


Figure 1 | Experimental setup of the mesocosm-scale floating treatment wetland (FTW) experiment (a). A detailed view of the macrophytes and floating mats for the 1st series experimental unit (b), and the setup employed to evaluate the propylene glycol removal pathway in the 2nd series of experiment (c).

supported by a 5×5 cm floating mat. To reduce propylene glycol loss through evaporation and photodegradation, each bucket in group 2 was covered with a lid, leaving a Ø 4 cm hole for the plant (Figure 1(c), plant + floating mat + lid). To assess the contribution of microbial biofilm on glycol treatment, 30 mg/L of sulfamethoxazole was added to the solution to kill or inhibit bacterial growth in group 3 (plant + floating mat + antibiotics). This antibiotic was chosen because it is frequently present in the environment and has been used to inhibit microbial activity in previous studies (Schmidt *et al.* 2012). The antibiotic dosage used in this experiment was based on a prior study that evaluated the impact of antibiotics on FTWs (Garcia Chance *et al.* 2020). The initial working solution and plant wet biomass were 0.8 L and 100 g in each bucket, respectively. The experimental procedures were identical to those in the first series of experiments.

The initial solution for both experiment series was prepared using local river water (Chicheley Brook, Buckinghamshire, England) and supplemented with propylene glycol. The initial concentration of propylene glycol was set at 1,250 mg/L with a total organic carbon (TOC) of 1,680 mg/L to replicate typical levels of this contaminant in airport runoffs (Murphy *et al.* 2015). Furthermore, a 1% concentration of Formulex nutrient solution (Growth Technology Ltd, Somerset, UK) was added to each bucket weekly to support plant growth.

2.3. Sampling and analysis

2.3.1. Water quality and water loss

Water levels were checked, physicochemical water quality assessed with a multiparameter meter, and water samples taken for glycol concentration at day 2, day 7, and then weekly. Change in water level was used to determine water loss due to evapotranspiration. To assess the physicochemical water quality, HANNA HI-9813-6 and HI-9147-04 probes were used to measure pH, temperature, conductivity, and dissolved oxygen (DO) directly in the buckets. Prior to measurement, all probes were calibrated according to the instrumentation manual, and electrodes were carefully cleaned between measurements to prevent cross-contamination. Water samples (20 mL) were collected using a 25-mL syringe, and prior to each sampling, the buckets were gently shaken for thorough mixing. The samples were than filtered through a 0.2 μm retention cellulose acetate membrane syringe filter in preparation for propylene glycol analysis.

2.3.2. Propylene glycol analysis and calculation

Water samples were analysed on the same day of collection using an HPLC system (Agilent Technologies 1,200 Series, USA) equipped with a Rezex ROA column and a diode array detector (DAD). An injection volume of 100 μL was used, and the analysis was performed under a flow rate of 0.4 mL/min at 60 °C. Standards were prepared using a 10 g/L stock solution of glycol. Both final glycol concentrations and mass removal (Equation (1)) were used to evaluate the treatment performance of different FTW mesocosms.

$$M_{\text{removed}} = C_i \times V_i - C_o \times V_o \quad (1)$$

Downloaded from http://iwaponline.com/wst/article-pdf/88/11/2751/1340103/wst088112751.pdf
by guest

where $M_{removed}$ represents the removed mass of propylene glycol (mg), C_i and C_o are the initial and final concentrations of propylene glycol (mg/L), respectively. V_i and V_o are the initial and final volumes of water in each bucket (mL).

2.3.3. Plant measurement

The wet biomass of each plant species per experimental unit was quantified at the beginning and end of the experiment using a precision weighing scale. Furthermore, the physical changes in root mass and the presence of different species were recorded.

2.4. Statistical analysis

The normality of the mean glycol concentration data was evaluated using the Shapiro-Wilk W test, which was followed by an Analysis of Variance (ANOVA) evaluation to assess the effect of plant species in FTWs on glycol concentration changes. JMP v13 was employed to perform this statistical analysis, and $p \leq 0.05$ were used as the threshold for statistical significance.

3. RESULTS

3.1. The change of water quality

The experiment was carried out in the summer season in the UK, where the water temperature remained between 21 and 25 °C throughout the study period (Table 1). The pH level in all FTW mesocosms significantly increased from approximately 6.4 at the beginning to 7.2–7.4 after 1 week, regardless of the plant species differences in the system. The electrical conductivity (EC) levels in the solution remained at a similar level (1.9–2.0 mS/cm) after 1 week in the control group. However, in the presence of wetland plants (including the mixed planting FTW), the EC significantly dropped to 1.2–1.5 mS/cm after 1 week. The DO concentration was 33.9 ± 0.2 mg/L in the freshly prepared solution. After 1 week, the DO decreased to 1.3–1.7 mg/L in the FTWs with plants, but no significant differences were observed between each group. The final DO level in the control group was significantly lower (0.7 ± 0.3 mg/L) than in all other systems.

3.2. The removal of propylene glycol

To maintain the initial volume and concentration of propylene glycol, corresponding solutions were added to each bucket at the start of each week. This ensured that the experiment could be considered as four-time replicates under the same conditions. The initial glycol concentration was set at 1,250 mg/L, and after 1 week, the concentrations in the control group (static water condition without plant) decreased to 1,020–1,110 mg/L (Figure 2(a)). All planted FTWs showed significantly lower glycol concentrations (<185 mg/L) in the solution at the end of study compared to the control group. In general, no significant differences in removal performance were observed between different weeks in each group (Figure 2(a)). Thus, the results from the 4-week study were combined to evaluate the overall performance of each group (Figure 2(b)). The final concentration of glycol in FTWs planted with *J. effusus* was significantly higher (54–185 mg/L) than those with *C. acutiformis* (0–149 mg/L) and *A. calamus* (11–74 mg/L). Moreover, the polyculture FTW showed significantly lower concentration of glycol (20–42 mg/L) after one week's treatment compared to *J. effusus* but similar to other two types of plants.

Although the final concentration of glycol is always the most important parameter in terms of regulatory and potential environmental risk, water evaporation effects, particularly in the open water surface FTW, may significantly concentrate the pollutant and lead to a misleading understanding of the system's functionality. In this study, the removed mass of propylene glycol from all systems was analysed (Figure 2(c)). Around 11–18% percentage removal of glycol was obtained in the

Table 1 | The changes in water quality at the beginning and after each week's treatment in the floating treatment wetland (FTW) mesocosms and the control group (no plants)

		Temperature (°C)	pH	EC (mS/cm)	DO (mg/L)
Initial level (all system)		24.9 ± 1.2	6.4 ± 0.2[a]	2.0 ± 0.1[a]	3.9 ± 0.2[a]
After 7 days	Control (no plant)	21.2 ± 2.3	7.4 ± 0.4[b]	1.9 ± 0.3[a]	0.7 ± 0.3[c]
	Juncus effusus	18.5 ± 4.3	7.2 ± 0.4[b]	1.2 ± 0.3[b]	1.3 ± 0.1[b]
	Carex acutiformis	19.7 ± 4.0	7.3 ± 0.4[b]	1.5 ± 0.4[b]	1.4 ± 0.3[b]
	Acorus calamus	20.9 ± 3.0	7.3 ± 0.3[b]	1.2 ± 0.2[b]	1.7 ± 0.5[b]
	Mixed species	20.8 ± 2.9	7.2 ± 0.3[b]	1.5 ± 0.3[b]	1.4 ± 0.1[b]

Note: Different letters beside the number for the same parameter represent significant differences.

Downloaded from http://iwaponline.com/wst/article-pdf/88/11/2751/1340103/wst088112751.pdf
by guest

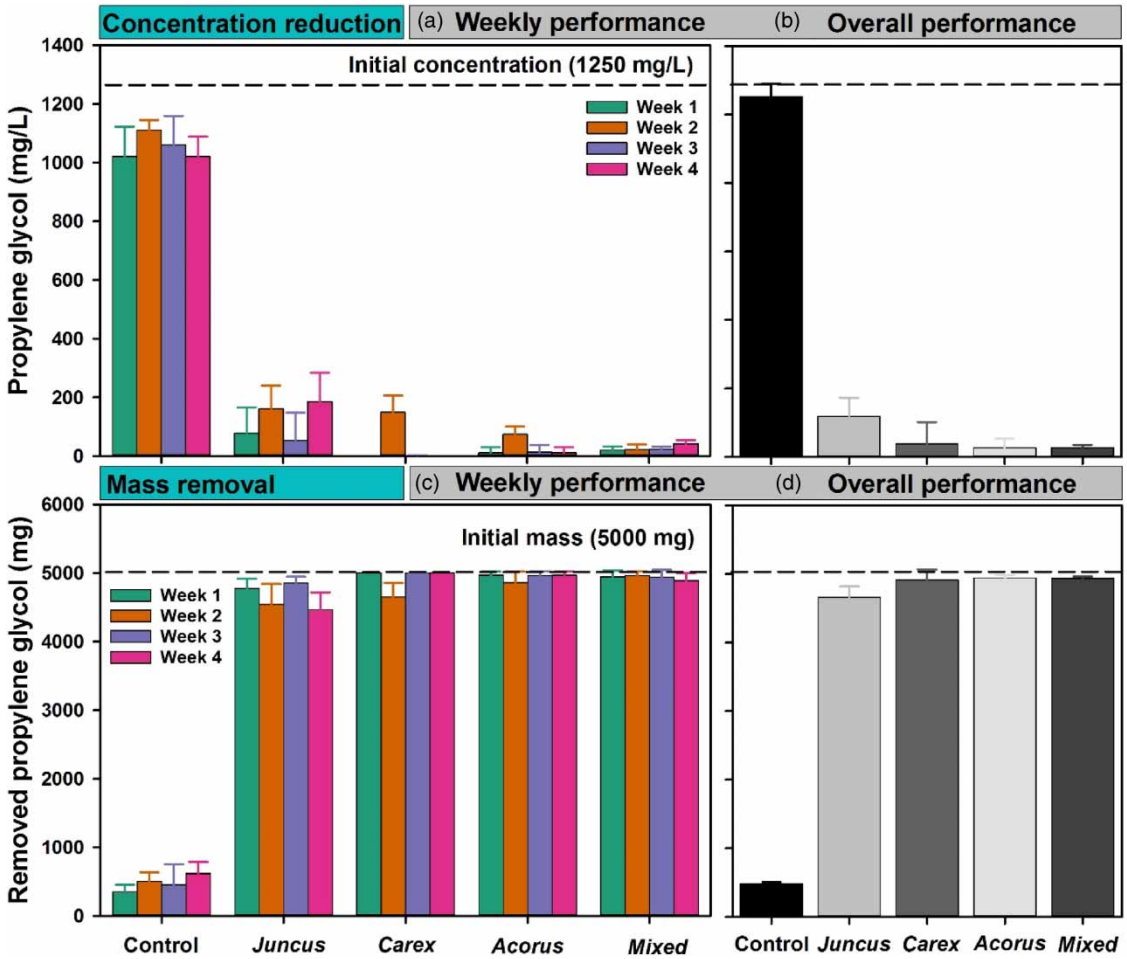


Figure 2 | The concentration changes of propylene glycol after each week (a) and the overall performance along the whole experiment (b) in different floating treatment wetland (FTW) mesocosms with different plant species. The mass removal of propylene glycol after each week (c) and the overall performance (d) in all FTWs.

control group (Figure 2(a)), and 7–11% of mass removal could be accounted for after considering the water loss in the system. Other planted FTWs showed a constant high mass removal (89–99%) of glycol after 7 days of treatment throughout the four-week study (Figure 2(c)). Similar to the results of percentage removal, FTW planted with *J. effusus* had a lower average mass removal on average (4,659 mg, 93%) than with *C. acutiformis* (4,912 mg, 98%), *A. calamus* (4,939 mg, 99%), and polyculture (4,934 mg, 99%) (Figure 2(d)).

3.3. Plant growth and morphology changes

A. calamus had the highest biomass growth (55.6% in wet biomass) during the experiment period, compared to *J. effusus* (13.9%) and *C. acutiformis* (10.6%) (Figure 3(a)). *A. calamus* also exhibited extensive root growth (Figure 3(d)) in comparison to the other plant species. The mixed plants showed a 14.9% increase in wet biomass growth at the end of the experiment. Additionally, the growth of an unidentified wild flora was observed in all experimental units containing *J. effusus* (Figure 3(b)) and the mixed species (Figure 3(e)). Notably, the presence of this wild flora was more extensive within the single species of *J. effusus* units.

3.4. The removal pathways of propylene glycol

Another series of experiment was conducted to evaluate the removal pathways of propylene glycol, including microbial biodegradation, plant uptake, evaporation and photodegradation. Measurements were taken on days 2 and 7 of the experiment. The initial mass of propylene glycol in all systems was 1,000 mg. The addition of antibiotics to inhibit the microbial activity

Downloaded from http://iwaponline.com/wst/article-pdf/88/11/2751/1340103/wst088112751.pdf
by guest

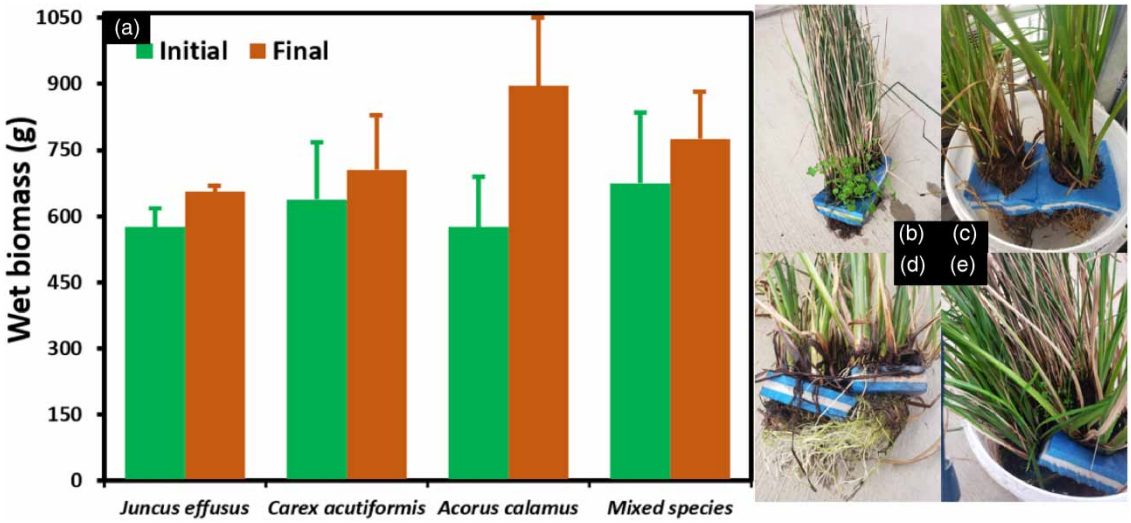


Figure 3 | The plant wet biomass of different plant species at the beginning and end of the experiment (a). The photos of *Juncus effusus* (b), *Carex acutiformis* (c), *Acorus calamus* (d), and mixed plants (e) in the experiment.

(Figure 4(a)) resulted in significantly higher propylene glycol mass in the solution (583–653 mg, Group 3 in Figure 4) after 2 days of treatment compared to conventional floating treatment wetland (FTW) setups (210–303 mg, Group 1 in Figure 4). An additional lid was employed to reduce propylene glycol removal through evaporation and photodegradation processes (Group 2 in Figure 4), leading to a higher residual mass (319–410 mg) compared to standard FTWs. Based on mass balance analyses (Figure 4(b)), the combined effects of evaporation and photodegradation accounted for 10.8–13.8% of the total mass removal. In contrast, microbial biodegradation (31.2–43.7%) and plant uptake (20.9–30.7%) were identified as the primary removal pathways. The removal of glycols through plant uptake followed the order of *A. calamus* (30.7%) > *J. effusus* (27.8%) > *C. acutiformis* (20.9%). Microbial biodegradation processes contributed more to glycol removal than plant uptake, with percentages of 37.4, 31.1, and 43.7% for *A. calamus*, *J. effusus*, and *C. acutiformis*, respectively.

A similar trend was observed after 7 days of treatment across all groups (Figure 4(c)), wherein 90–98% of the glycol mass was removed from the FTWs with standard setups (Group 1 in Figure 4(c)). However, groups with the additional lid (Group 2) and antibiotic treatment (Group 3) exhibited residual glycol masses of 169–237 mg (76–83% removal) and 381–425 mg (58–62% removal), respectively. The contributions of microbial biodegradation, plant uptake and evaporation increased to 37.9, 45.2, and 15.5% for *A. calamus*, 33.3, 43.1, and 18.9% for *J. effusus*, and 39.7, 37.9, and 19.5% for *C. acutiformis*, respectively (Figure 4(d)). The results confirmed that microbial biodegradation and plant uptake persist as the primary pathways for propylene glycol removal.

4. DISCUSSION

4.1. The effect of different plant species and planting strategies

The observed reduction in EC (Table 1) in the solution was attributed to the removal of dissolved glycols (Figure 2) and nutrients, such as organics, nitrogen, and phosphorus (Wang *et al.* 2021). In the absence of plants in the control group, the EC remained similar to the initial level, with less glycol removal (Table 1). The relatively low DO levels observed in this experiment (Table 1) highlight the occurrence of oxygen-consuming processes required for glycol removal through microbial biodegradation, which can have toxic effects on aquatic biota (Freeman *et al.* 2015). The DO in the control group dropped to an extremely low level (0.7 mg/L), however, wetland plant roots can release oxygen into the solution (Brix 1997), and the DO in the planted FTWs remained at a relatively higher level (1.3–1.7 mg/L) compared to the control group.

This study demonstrated that all of the selected plant species could remove glycol up to 99% within 7 days (Figure 2). The results were generally consistent with previous findings in glycol phytoremediation, which were supported by earlier studies showing above 91% removal within 12–15 day under a higher initial concentration of 2,000 mg/L (Teamkao & Thiravetyan 2010; Sriprapat *et al.* 2011). *A. calamus* was the most efficient species, especially when planted as a single species, despite

Downloaded from http://iwaponline.com/wst/article-pdf/88/11/2751/1340103/wst088112751.pdf
by guest

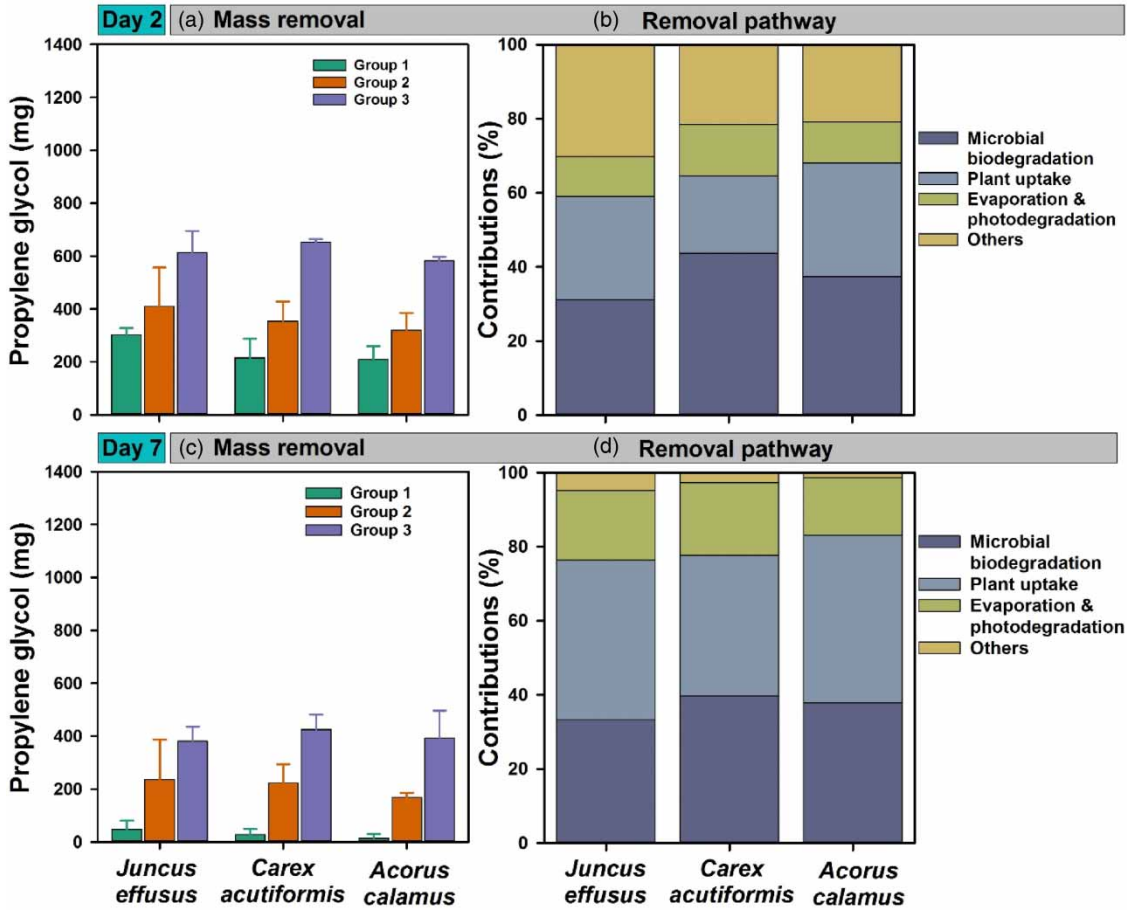


Figure 4 | The mass of propylene glycol after 2 (a) and 7 (c) days of the experiment in different floating treatment wetland (FTW) mesocosms. The contributions of different removal pathways of propylene glycol in three FTW mesocosms planted with individual species at day 2 (b) and 7 (d). Group 1 represent the standard FTW setup, group 2 consists of FTWs with an additional surface lid, and group 3 incorporates antibiotics to inhibit microbial activity.

having a smaller wet biomass and root structure at the beginning of the experiment (Figure 3(a)). However, the *A. calamus* achieved the highest biomass growth (Figure 4(a)) and root density (Figure 4(d)), which may contribute to the higher removal performance of glycol through plant uptake and microbial biodegradation processes. In contrast, the polyculture FTW did not exhibit such high biomass growth during the experiment but still achieved nearly complete removal of glycol. Previous studies have shown that mixed cultures of plants can enhance microbial biodiversity and activity in wetlands (Button *et al.* 2016; Wang *et al.* 2022), which may have contributed to the high removal efficiency of glycol observed in this study. In summary, this study confirms that the selected UK native macrophytes are suitable and effective for the removal of propylene glycol and that *A. calamus* or mixed species could provide the best removal performance in FTWs.

4.2. Propylene glycol removal pathways in FTWs

Propylene glycol is a VOC that should be removed via evaporation and photodegradation (Chen *et al.* 2017). Nevertheless, only 10.8–13.8% (Figure 4(b)) and 15.5–19.5% (Figure 4(d)) of the glycol mass were removed via these processes after 2 days and 7 days of treatment, respectively. These low values may be due to the low exposed water surface, as much of the water surface was covered by the floating mat and plant within the experimental mesocosms. Previous studies have reported that FTWs can effectively reduce evaporation and inhibit phytoplankton growth through shielding and nutrient uptake (Jones *et al.* 2017). Additionally, lower removal rates within mesocosm experiments may be attributed to lower heat transfer between air and stagnant water compared to flowing systems, which can lower evaporation rates (Garcia Chance *et al.* 2020).

The contributions of plant uptake and microbial biodegradation of glycol in this study, accounting for over 80% of the total, were supported by previous studies that claimed that microbial biodegradation and plant's effect could be the key factors in

Downloaded from http://iwaponline.com/wst/article-pdf/88/11/2751/1340103/wst088112751.pdf
by guest

removing glycols (Chong *et al.* 1999; Higgins & Maclean 2002; Murphy *et al.* 2015). Moreover, the contributions of microbial biodegradation and plant uptake exhibited similar patterns during both the 2- and 7-day investigations (Figure 4). The reduced removal efficiencies of glycol (Figure 4(a)) compared to normally operated FTWs (Figure 2(b)) indicated that bacterial composition had been altered as a result of the antibiotic addition. Tong *et al.* (2020) also reported that exposure to ofloxacin and tetracycline can suppress the growth and activity of overall microbial bacteria. The substantial removal of glycol through plant uptake may be attributed to the rapid growth of the plants during that specific period. With regard to the long-term operation of the FTW in the applied system, as the plant biomass reaches a stable period, microbial biodegradation is expected to become the dominant process for glycol removal in the system. Long-term monitoring, with the support of microbial community analysis, is essential to gain a more accurate understanding of the contributions, which is recommended for future studies.

4.3. Co-benefits and FTW implementations

A whole lake experiment showed that the application of FTWs can aesthetically improve the lake environment and increase biodiversity by attracting more local fish and other wildlife (Henny *et al.* 2020). In this study, the presence of small wild plant species did not affect the FTW's performance as averagely 93% glycol removal was achieved throughout the observation period (Figure 2). The results supported the diverse benefits of macrophytes in increasing biodiversity while removing glycols. Moreover, FTWs are a feasible net zero carbon strategy that should be encouraged within WWTPs, as they have a low carbon footprint with 50% fewer greenhouse gas (GHG) emissions (Nahlik & Mitsch 2006). The previous study has demonstrated that FTWs can act as sinks for carbon, considering different types of GHGs, with carbon storage capability up to 25 g CO_2-eq/m^2/d (Sun *et al.* 2019). As GHG emissions from wetlands vary widely, depending mostly on location, the source of water received by the wetlands and wetland vegetation (D'Acunha & Johnson 2019). Therefore, directly monitoring at larger scale applied FTWs in the context of glycol removal is needed to quantify the co-benefits.

5. CONCLUSION

This study provides evidence that FTWs are highly effective in mitigating the contaminant propylene glycol. All plant species tested in the FTWs were able to remove up to 99% of the glycol within 1 week under a high initial concentration of 1,250 mg/L. *A. calamus* and the mixed species were found to be the most effective planting strategy for rapidly reducing glycol concentrations compared to other plant species. The key mechanisms responsible for glycol removal were found to be microbial biodegradation and plant uptake, which together accounted for nearly 80% of glycol removal. The contributions of evaporation and photodegradation were responsible for the remaining glycol mass removal; however, their contributions are estimated to be higher in applied FTWs with moving waters. Overall, this study demonstrates the potential of FTWs for treating glycol-contaminated water while potentially delivering additional co-benefits such as supporting biodiversity and carbon sequestration.

ACKNOWLEDGEMENTS

This research was funded by Heathrow Airport Ltd and conducted at Cranfield University. The authors gratefully acknowledge the support of Andy Freeman and Clodagh Murphy from ARM Ltd, who provided valuable insights and consultation based on their experience with treatment wetlands and glycol management.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

REFERENCES

Afzal, M., Arslan, M., Müller, J. A., Shabir, G., Islam, E., Tahseen, R., Anwar-ul-Haq, M., Hashmat, A. J., Iqbal, S. & Khan, Q. M. 2019 Floating treatment wetlands as a suitable option for large-scale wastewater treatment. *Nat. Sustainability* **29** (2), 863–871. https://doi.org/10.1038/s41893-019-0350-y.

Downloaded from http://iwaponline.com/wst/article-pdf/88/11/2751/1340103/wst088112751.pdf
by guest

Barco, A., Bona, S. & Borin, M. 2021 Plant species for floating treatment wetlands: A decade of experiments in North Italy. *Sci. Total Environ.* **751**, 141666. https://doi.org/10.1016/J.SCITOTENV.2020.141666.

Brix, H. 1997 Do macrophytes play a role in constructed treatment wetlands? *Water Sci. Technol.* **35**, 11–17. https://doi.org/10.1016/S0273-1223(97)00047-4.

Button, M., Rodriguez, M., Brisson, J. & Weber, K. P. 2016 Use of two spatially separated plant species alters microbial community function in horizontal subsurface flow constructed wetlands. *Ecol. Eng.* **92**, 18–27. https://doi.org/10.1016/J.ECOLENG.2016.03.044.

Castellar, J. A. C., Torrens, A., Buttiglieri, G., Monclús, H., Arias, C. A., Carvalho, P. N., Galvao, A. & Comas, J. 2022 Nature-based solutions coupled with advanced technologies: An opportunity for decentralized water reuse in cities. *J. Cleaner Prod.* **340**, 130660. https://doi.org/10.1016/J.JCLEPRO.2022.130660.

Chen, Z., Reiche, N., Vymazal, J. & Kuschk, P. 2017 Treatment of water contaminated by volatile organic compounds in hydroponic root mats. *Ecol. Eng.* **98**, 339–345. https://doi.org/10.1016/J.ECOLENG.2016.08.012.

Chong, S., Garelick, H., Revitt, D. M., Shutes, R. B. E., Worrall, P. & Brewer, D. 1999 The microbiology associated with glycol removal in constructed wetlands. *Water Sci. Technol.* **40**, 99–107. https://doi.org/10.1016/S0273-1223(99)00445-X.

Colares, G. S., Dell'Osbel, N., Wiesel, P. G., Oliveira, G. A., Lemos, P. H. Z., da Silva, F. P., Lutterbeck, C. A., Kist, L. T. & Machado, Ê. L. 2020 Floating treatment wetlands: A review and bibliometric analysis. *Sci. Total Environ.* **714**, 136776. https://doi.org/10.1016/J.SCITOTENV.2020.136776.

Corsi, S. R., Mericas, D. & Bowman, G. T. 2012 Oxygen demand of aircraft and airfield pavement deicers and alternative freezing point depressants. *Water Air Soil Pollut.* **223**, 2447–2461. https://doi.org/10.1007/s11270-011-1036-x.

D'Acunha, B. & Johnson, M. S. 2019 Water quality and greenhouse gas fluxes for stormwater detained in a constructed wetland. *J. Environ. Manage.* **231**, 1232–1240. https://doi.org/10.1016/J.JENVMAN.2018.10.106.

Exton, B., Hassard, F., Medina Vaya, A. & Grabowski, R. 2023 Polybacterial shift in benthic river biofilms attributed to organic pollution – a prospect of a new biosentinel? *Hydrol. Res.* **54**, 348–359. https://doi.org/10.2166/nh.2023.114.

Freeman, A. I., Surridge, B. W. J., Matthews, M., Stewart, M. & Haygarth, P. M. 2015 Understanding and managing de-icer contamination of airport surface waters: a synthesis and future perspectives. *Environ. Technol. Innov.* **3**, 46–62. https://doi.org/10.1016/J.ETI.2015.01.001.

Garcia Chance, L. M., Majsztrik, J. C., Bridges, W. C., Willis, S. A., Albano, J. P. & White, S. A. 2020 Comparative nutrient remediation by monoculture and mixed species plantings within floating treatment wetlands. *Environ. Sci. Technol.* **54**, 8710–8718. https://doi.org/10.1021/ACS.EST.0C00198/SUPPL_FILE/ES0C00198_SI_001.PDF.

Headley, T. R. & Tanner, C. C. 2011 Constructed wetlands with floating emergent macrophytes: An innovative stormwater treatment technology. **42**, 2261–2310. https://doi.org/10.1080/10643389.2011.574108.

Henny, C., Jasalesmana, T., Kurniawan, R., Melati, I., Suryono, T., Susanti, E., Yoga, G. P., Rosidah & Sudiono, B. T. 2020 The effectiveness of integrated floating treatment wetlands (FTWs) and lake fountain aeration systems (LFAS) in improving the landscape ecology and water quality of a eutrophic lake in Indonesia. *IOP Conf. Ser. Earth Environ. Sci.* **535**, 012018. https://doi.org/10.1088/1755-1315/535/1/012018.

Higgins, J. & Maclean, M. 2002 Technical note – the use of a very large constructed sub-surface flow wetland to treat glycol-contaminated stormwater from aircraft de-icing operations. *Water Qual. Res. J.* **37**, 785–792. https://doi.org/10.2166/WQRJ.2002.053.

Jones, T. G., Willis, N., Gough, R. & Freeman, C. 2017 An experimental use of floating treatment wetlands (FTWs) to reduce phytoplankton growth in freshwaters. *Ecol. Eng.* **99**, 316–323. https://doi.org/10.1016/J.ECOLENG.2016.11.002.

Lv, T., Zhang, Y., Casas, M. E., Carvalho, P. N., Arias, C. A., Bester, K. & Brix, H. 2016a Phytoremediation of imazalil and tebuconazole by four emergent wetland plant species in hydroponic medium. *Chemosphere* **148**, 459–466. https://doi.org/10.1016/J.CHEMOSPHERE.2016.01.064.

Lv, T., Zhang, Y., Zhang, L., Carvalho, P. N., Arias, C. A. & Brix, H. 2016b Removal of the pesticides imazalil and tebuconazole in saturated constructed wetland mesocosms. *Water Res.* **91**, 126–136. https://doi.org/10.1016/J.WATRES.2016.01.007.

Marecik, R., Bieganska-Marecik, R., Cyplik, P., Lawniczak, L. & Chrzanowski, L. 2013 Phytoremediation of industrial wastewater containing nitrates, nitroglycerin, and nitroglycol. *Polish J. Environ. Stud.* **22**, 773–780.

Murphy, C., Wallace, S., Knight, R., Cooper, D. & Sellers, T. 2015 Treatment performance of an aerated constructed wetland treating glycol from de-icing operations at a UK airport. *Ecol. Eng.* **80**, 117–124. https://doi.org/10.1016/J.ECOLENG.2014.05.032.

Nahlik, A. M. & Mitsch, W. J. 2006 Tropical treatment wetlands dominated by free-floating macrophytes for water quality improvement in Costa Rica. *Ecol. Eng.* **28**, 246–257. https://doi.org/10.1016/J.ECOLENG.2006.07.006.

Nott, M. A., Driscoll, H. E., Takeda, M., Vangala, M., Corsi, S. R. & Tighe, S. W. 2020 Advanced biofilm analysis in streams receiving organic deicer runoff. *PLoS One* **15**, 1–27. https://doi.org/10.1371/journal.pone.0227567.

Oliveira, G. A., Colares, G. S., Lutterbeck, C. A., Dell'Osbel, N., Machado, Ê. L. & Rodrigues, L. R. 2021 Floating treatment wetlands in domestic wastewater treatment as a decentralized sanitation alternative. *Sci. Total Environ.* **773**, 145609. https://doi.org/10.1016/J.SCITOTENV.2021.145609.

Oral, H. V., Carvalho, P., Gajewska, M., Ursino, N., Masi, F., van Hullebusch, E. D., Kazak, J. K., Exposito, A., Cipolletta, G., Andersen, T. R., Finger, D. C., Simperler, L., Regelsberger, M., Rous, V., Radinja, M., Buttiglieri, G., Krzeminski, P., Rizzo, A., Dehghanian, K., Nikolova, M. & Zimmermann, M. 2020 A review of nature-based solutions for urban water management in European circular cities: A critical assessment based on case studies and literature. *Blue-Green Syst.* **2**, 112–136. https://doi.org/10.2166/BGS.2020.932.

Downloaded from http://iwaponline.com/wst/article-pdf/88/11/2751/1340103/wst088112751.pdf
by guest

Park, J. B. K., Sukias, J. P. S. & Tanner, C. C. 2019 Floating treatment wetlands supplemented with aeration and biofilm attachment surfaces for efficient domestic wastewater treatment. *Ecol. Eng.* **139**, 105582. https://doi.org/10.1016/J.ECOLENG.2019.105582.

Pavlineri, N., Skoulikidis, N. T. & Tsihrintzis, V. A. 2017 Constructed floating wetlands: A review of research, design, operation and management aspects, and data meta-analysis. *Chem. Eng. J.* **308**, 1120–1132. https://doi.org/10.1016/J.CEJ.2016.09.140.

Richter, K. M., Margetts, J. R., Saul, A. J., Guymer, I. & Worrall, P. 2003 Baseline hydraulic performance of the Heathrow constructed wetlands subsurface flow system. *Water Sci. Technol.* **47**, 177–181. https://doi.org/10.2166/WST.2003.0687.

Rogers, J. D., Thurman, E. M., Ferrer, I., Rosenblum, J. S., Evans, M. V., Mouser, P. J. & Ryan, J. N. 2019 Degradation of polyethylene glycols and polypropylene glycols in microcosms simulating a spill of produced water in shallow groundwater. *Environ. Sci. Process. Impacts* **21**, 256–268. https://doi.org/10.1039/C8EM00291F.

Schmidt, S., Winter, J. & Gallert, C. 2012 Long-term effects of antibiotics on the elimination of chemical oxygen demand, nitrification, and viable bacteria in laboratory-scale wastewater treatment plants. *Arch. Environ. Contam. Toxicol.* **63**, 354–364. https://doi.org/10.1007/S00244-012-9773-4/FIGURES/6.

Sriprapat, W., Kullavanijaya, S., Techkarnjanaruk, S. & Thiravetyan, P. 2011 Diethylene glycol removal by *Echinodorus cordifolius* (L.): The role of plant–microbe interactions. *J. Hazard. Mater.* **185**, 1066–1072. https://doi.org/10.1016/J.JHAZMAT.2010.10.015.

Sun, S., Liu, J., Zhang, M. & He, S. 2019 Simultaneous improving nitrogen removal and decreasing greenhouse gas emission with biofilm carriers addition in ecological floating bed. *Bioresour. Technol.* **292**, 121944. https://doi.org/10.1016/J.BIORTECH.2019.121944.

Teamkao, P. & Thiravetyan, P. 2010 Phytoremediation of ethylene glycol and its derivatives by the burhead plant (Echinodorus cordifolius (L.)): Effect of molecular size. *Chemosphere* **81**, 1069–1074. https://doi.org/10.1016/J.CHEMOSPHERE.2010.09.049.

Tong, X. N., Wang, X. Z., He, X. J., Wang, Z. & Li, W. X. 2020 Effects of antibiotics on microbial community structure and microbial functions in constructed wetlands treated with artificial root exudates. *Environ. Sci. Process. Impacts* **22**, 217–226. https://doi.org/10.1039/C9EM00458K.

Wang, H., Sun, J., Xu, J. & Sheng, L. 2021 Study on clogging mechanisms of constructed wetlands from the perspective of wastewater electrical conductivity change under different substrate conditions. *J. Environ. Manage.* **292**, 112813. https://doi.org/10.1016/J.JENVMAN.2021.112813.

Wang, Z., Zhang, Y., Li, X., Li, J., Zhao, Z. & Hou, X. 2022 Mixed culture of plants improved nutrient removal in constructed wetlands: Response of microbes and root exudates. *Environ. Sci. Pollut. Res.* **30**, 5861–5872. https://doi.org/10.1007/S11356-022-22305-4/FIGURES/6.

West, R., Banton, M., Hu, J. & Klapacz, J. 2014 The distribution, fate, and effects of propylene glycol substances in the environment. *Rev. Environ. Contam. Toxicol.* **232**, 107–138. https://doi.org/10.1007/978-3-319-06746-9_5/COVER.

Zhang, Y., Lv, T., Carvalho, P. N., Zhang, L., Arias, C. A., Chen, Z. & Brix, H. 2017 Ibuprofen and iohexol removal in saturated constructed wetland mesocosms. *Ecol. Eng.* **98**, 394–402. https://doi.org/10.1016/J.ECOLENG.2016.05.077.

First received 23 May 2023; accepted in revised form 21 November 2023. Available online 1 December 2023

Downloaded from http://iwaponline.com/wst/article-pdf/88/11/2751/1340103/wst088112751.pdf
by guest



https://dspace.org/

Assessing the efficacy and mechanisms of glycol-contaminated water treatment through floating treatment wetlands

Lyu, Tao; Williams, Randy; Exton, Benjamin; Grabowski, Robert C.

2023-12-01
IWA Publishing
https://doi.org/10.2166/wst.2023.389
Lyu T, Williams R, Exton B, Grabowski RC. (2023) Assessing the efficacy and mechanisms of glycol-contaminated water treatment through floating treatment wetlands. Water Science & Technology, Volume 88, Issue 11, December 2023, pp. 2751-2761

Downloaded from a DSpace repository. https://dspace.org/