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# A novel hybrid coagulation-constructed wetland system for the treatment of dairy wastewater

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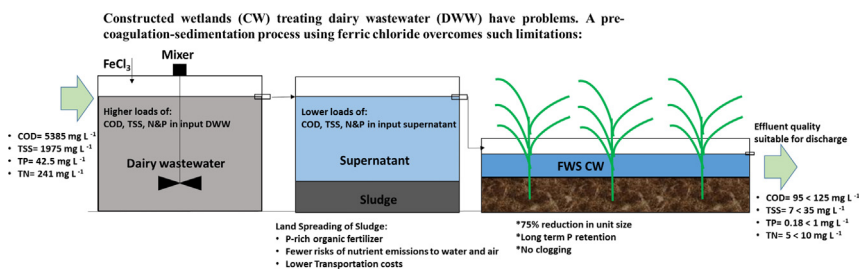
## HIGHLIGHTS

- Performance of hybrid coagulation-CW was compared to conventional CW.
- $FeCl_3$ pre-treatment showed significant reduction of COD, TSS, turbidity, TP, and TN.
- Coagulation-CW produced high effluent quality suitable for discharge.
- Coagulation-CW may be operated at higher HLRs than conventional CWs.
- An OLR of 3.5 g COD $m^{-2}$ $d^{-1}$ was the optimum OLR for constructed wetlands.

## GRAPHICAL ABSTRACT

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## ABSTRACT

Constructed wetlands (CWs) are a cost-effective and sustainable treatment technology that may be used on farms to treat dairy wastewater (DWW). However, CWs require a large area for optimal treatment and have poor long-term phosphorus removal. To overcome these limitations, this study uses a novel, pilot-scale coagulation-sedimentation process prior to loading CWs with DWW. This hybrid system, which was operated on an Irish farm over an entire milking season, performed well at higher hydraulic loading rates than conventional CWs, and obtained removal efficiencies ≥99 % for all measured water quality parameters (chemical oxygen demand, total nitrogen and phosphorus, total suspended solids and turbidity), which complied with EU directives concerning urban wastewater treatment. Overall, the hybrid coagulation-CW is a promising technology that requires a smaller area than conventional CWs and minimal operator input, and produces high effluent quality.

## 1. Introduction

As the world's population is projected to reach 10 billion by 2050, humankind is increasingly facing a burden of hunger and malnutrition (World Health Organization, 2020). Overcoming these challenges will require a sustainable increase of food production globally, taking into consideration biodiversity protection, and delivery of ecosystem and environmental services (Ulian et al., 2020). In many countries, dairy farming is a vital agricultural and economic sector. In Ireland, for example, dairy products amount to one third of all Irish agri-food exports (Irish Food Board, 2019). From 2005 to 2016, the size of dairy herds in Ireland increased fourfold (in farms milking >100 cows; Kelly et al., 2020), and this expansion has caused an increase in the volumes of dairy wastewater (DWW) produced by farms.

Dairy wastewater (also referred to as dairy farm effluent, dairy soiled water, and dairy dirty water) is effluent produced from the washing-down of milking parlours and holding areas, and consists of a dilute mixture of milk, urine, livestock faeces, detergents and sediment. Although DWW has a good value as an organic fertilizer (Minogue et al., 2021), land spreading of DWW can result in contamination of groundwater and deterioration of surface water quality, especially when spread under unfavourable weather and soil conditions (Sommer and Knudsen, 2021). Therefore, to avoid these environmental risks, and to attain good status of surface and

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ground waters, there is a demand for cost-effective and low maintenance treatment processes for DWW.

Constructed wetlands (CWs) have been frequently used for the treatment of agricultural and livestock wastewaters such as piggery wastewater (Li et al., 2020) and dairy wastewater (Schierano et al., 2020). The large surface area of CWs facilitates an environment for physico-chemical retention, biological degradation of organic matter, and the removal of total suspended solids (TSS) and nutrients (Scholz and Lee, 2005; Healy et al., 2007). These systems have been gaining popularity because they are cost-effective, sustainable, and easily operated and maintained. In addition, they provide other social and landscape benefits such as wildlife habitat and biodiversity, ecosystem/ecological services, and recreational uses (Harrington and McInnes, 2009).

Nevertheless, the large surface area required for treatment limits the application of CWs. For example, a low organic loading rate (OLR) of <6 g biochemical oxygen demand (BOD) $m^{-2}$ $d^{-1}$ is required for free water surface (FWS) CWs (US Environmental Protection Agency (USEPA), 1992). Some types of CWs may have higher OLRs. For example, Winter and Goetz (2003) recommend a maximum organic loading rate of 20 g COD $m^{-2}$ $d^{-1}$ for subsurface vertical flow (SSVF) CWs. Foladori et al. (2012) found that hybrid subsurface vertical and horizontal (SSVF + SSHF) CWs treating domestic wastewater can be operated with OLRs up to 87 g COD $m^{-2}$ $d^{-1}$ in summer without comprising effluent quality. CWs also have poor long-term phosphorus (P) removal due to the limited adsorption capacity of the soil, as all active sorbent sites diminish with the time and became exhausted (Kadlec, 2016; Lan et al., 2018).

Concentrations of pollutants in DWW are significantly higher than municipal effluent (Martínez-Suller et al., 2010; Minogue et al., 2015). Therefore, pre-treatment for CWs is recommended (EPA, 2000; Healy et al., 2007) to lessen the strength of the influent wastewater, and therefore avoid their inherent operational problems. Nan et al. (2020) reviewed the performance of 39 systems, incorporating CWs, for agricultural wastewater treatment. Their findings showed that the CWs in combination with other technologies (e.g. anaerobic reactors, UV treatment) can further improve their performance and achieve better removal efficiencies in comparison with conventional CWs. In particular, combined systems had better removals of bacterial indicators and organic matter than single-stage CWs.

Pre-treatment of DWW by a conventional coagulation-sedimentation process may address the shortcomings and operational problems of CWs. These include a large surface area requirement for treatment (Ilyas and Masih, 2017), limited capacity of P removal (Arias and Brix, 2005), clogging of media and substrate, particularly in subsurface flow CWs (de Matos et al., 2018; Wang et al., 2021), and greenhouse gas (GHG) emissions (Ingrao et al., 2020). Cameron and Di (2019) attained high removals of chemical oxygen demand (COD), total phosphorus (TP) and TSS for DWW treated with a ferric-based coagulant at small doses. Consequently, this method can result in a smaller sized downstream CW, with satisfactory long-term P removal, and with a potentially longer operational period without clogging.

Addition of ferric salt coagulants to amend DWW properties also has good P sequestration potential (Fenton et al., 2011; Serrenho et al., 2012) and may reduce GHG and ammonia emissions (Kavanagh et al., 2019). Therefore, the sludge produced from the coagulation-sedimentation process, which accounts for 25 % of the total influent DWW (7.5 ton DM per year per 100 dairy cows; Mohamed et al., 2020), can be spread as an organic fertilizer to agricultural land, with lower risk of nutrient losses to water bodies, and potentially lower GHG emissions to air (Wang et al., 2019; Chisholm et al., 2021).

Nevertheless, there is little knowledge of the efficacy of the coagulation-sedimentation process when combined with CWs, and no guidelines are available to adopt them at farm-scale as a potential pre-treatment method. The expected advantages of a CW configuration employing chemical pre-treatment are (1) high COD reduction during the pre-treatment stage, which reduces the size of a CW and allows an increased hydraulic loading rate (HLR) to be applied to the CW, (2) P retention and N reduction in the pre-treatment stage. The aims of this study were to, for the first time, (1) design and evaluate the performance of conventional CWs and hybrid coagulation-CWs system over a 43-week duration (covering a complete milking season) on an Irish farm (2) identify optimal OLRs and HLRs for a pilot-scale hybrid coagulation-CW system, and (3) provide design and operation guidelines for its successful adoption.

## 2. Materials and methods

### 2.1. Design of constructed wetlands

In July 2020, six shallow FWS CWs (Fig. S1-C) were constructed outdoors, using rectangular polyethylene tanks at the Teagasc Moorepark Dairy farm, Fermoy, Co. Cork, Ireland. In 2021, the mean annual temperature of the study area was 10.5 °C and the total annual rainfall was 1013 mm. The total annual evaporation and potential evapotranspiration were 716 and 519 mm, respectively (Met Éireann, 2021).

The CW design specifications were based on New Zealand guidelines for the treatment of farm dairy wastewaters (Tanner and Kloosterman, 1997) and USEPA guidelines (1992). The internal dimensions of each tank was 1.75 m (length) × 0.5 m (width) × 0.5 m (height), giving a surface area of 0.875 $m^2$ (Fig. 1). The length to width ratio (L:W) was 3.5:1, which generally provides an appropriate balance for surface-flow systems (Tanner and Kloosterman, 1997; US Environmental Protection Agency (USEPA), 2000). Each tank was filled with locally sourced soil to a depth of 0.25 m. The soil was a sandy loam texture comprising 67 % sand (2.00–0.063 mm), 21 % silt (0.063–0.002 mm) and 12 % clay (< 0.002 mm), and had chemical properties as shown in Table 1.

It is difficult to simulate the operation of full-scale FWS CWs in HDPE tanks over a relatively brief experimental period, as their performance is related to plant development and other natural conditions that may be scale-dependent. To address this limitation, the soil in the experimental units was planted with established and mature *Phragmites australis* (Cav.) Trin. ex Steud. (common reed). The plants were obtained (with roots intact) from a semi-natural wetland on-site at the Moorepark dairy farm, and planted in the tanks at similar density as the original wetland (≈ 200 plants per $m^2$). Following planting, all CWs were flooded with potable water to a depth of 0.15 ± 0.03 m (over soil surface) for four months, to allow establishment and acclimatization of the plants.

The water level in all CWs was maintained at 0.15 ± 0.03 m by installing overflow pipes (diameter = 0.04 m) at the outlet above the soil surface (Fig. 1). Shallow water depths were selected because are associated with the highest ammonia ($NH_3$) diffusion and nitrogen losses (Scholz et al., 2007). The inlet pipe for each CW was located in the left corner, while the outlet pipe was placed in the far right corner, so as to maximise the distance between the inlet and outlet of each system. This helps to increase the hydraulic retention time (HRT) within the CW and prevents the risk of hydraulic short-circuiting and preferential flow.

### 2.2. Wastewater preparation and experimental phases

Fresh DWW was collected once a week from an access chamber on the discharge pipe from the dairy parlour and transferred to a 1 $m^3$ storage container using a submersible pump (Fig. S1-A). The DWW was collected during milking events and comprised washings from the milking parlour, holding yard, and from cleaning the milking plant. The DWW consists of a mixture of water, milk, urine, cow faeces, detergents and sediment, so therefore it had very high contaminant concentrations (Table 2). The DWW was allowed to settle in the storage container for one day in order to remove large TSS, and therefore prevent potential blockages in the system, and avoid damage to downstream pumps. Following this, 200 L of DWW was fed by gravity into a 210 L barrel (henceforth referred to as raw DWW; Fig. 1-A). The raw DWW was mixed frequently to ensure homogeneity.

The remaining DWW in the storage container (800 L) was agitated with 2 L of a chemical coagulant, ferric chloride solution ($FeCl_3$; 40 % *w*/w; Table S1) (10.35 g Fe $g^{-1}$ P; 440 mg Fe $L^{-1}$ of DWW), at a velocity gradient

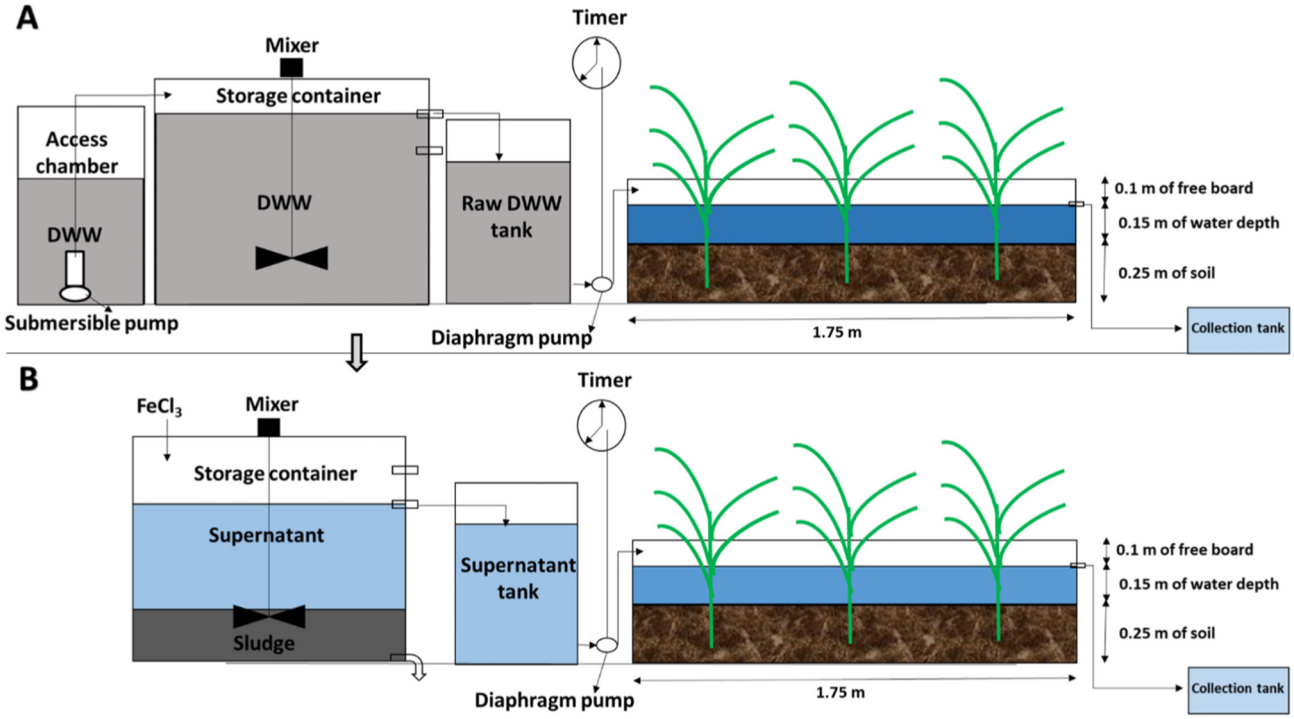


**Fig. 1.** Experimental unit set-up: A) Schematic view of raw dairy wastewater (DWW) system; B) Schematic view of supernatant system.

of 900 $s^{-1}$ for 10 min, using a mixer mounted in the storage container (after Mohamed et al., 2020). After mixing, the mixture was allowed to settle for 3 h, after which 200 L of the supernatant was fed by gravity into another 210 L barrel (henceforth referred to as supernatant; Fig. 1-B). This provided sufficient wastewater to supply the six CWs for an entire week. The storage container, raw and supernatant barrels, and collection containers, were thoroughly cleaned every week before the preparation of a new batch of DWW.

Raw DWW and supernatant (Table 2) were dosed discontinuously (intermittently 4 or 8 times per day, Table 3) onto the replicated CWs ($n = 3$) using waste pumps (type: diaphragm), which was controlled by electronic timers (Fig. 1-B). Hydraulic and organic loading rates were adjusted depending on the experimental phase, using the manual flow control valves that were positioned downstream of the pumps, and/or by altering the pumping duration per dose. The effluent from each CW was collected in a 20 L capacity container.

The experiment consisted of four phases and spanned the entire 2021 milking season (February to December; 43 weeks; Table 3). All CWs were operated with identical OLRs in the first and second phases. In Phase 1 (week 1–week 7), the OLR was set at the higher design limits (7 ± 1.5 g COD $m^{-2}$ $d^{-1}$) (after Tanner and Kloosterman, 1997). However, to increase the treatment efficiency of the CWs, the OLR was halved in Phase 2 to 3.5 ± 1 g COD $m^{-2}$ $d^{-1}$ for both sets of CWs (week 8 to week 29). The HLRs were modified frequently during Phases 1 and 2 to obtain a constant OLR under varying organic concentrations of DWW. In the third and fourth phases, all CWs were operated with identical HLRs. In Phase 3 (week 30 to week 35), the CWs treating supernatant continued to be operated with similar conditions to Phase 2 (OLR = 3.5 ± 1 g COD $m^{-2}$ $d^{-1}$, HLR = 2.3 L $m^{-2}$ $d^{-1}$). The HLR in the raw DWW CWs was raised to 2.3 L $m^{-2}$ $d^{-1}$, producing an OLR of 13 ± 2 g COD $m^{-2}$ $d^{-1}$. In Phase 4, the HLR was further raised to 4.7 L $m^{-2}$ $d^{-1}$ for both sets of CWs, to examine the efficacy of both systems at elevated OLRs.

The HRTs in the current study were relatively high compared to New Zealand guidelines for DWW treatment using FWS CWs (Tanner and Kloosterman, 1997), which recommends HRTs of between 8 and 12 days. This was due to the high concentrations of DWW (Table 2), which meant that, in order to safeguard the long-term operation of the CWs, initially low OLRs were applied (which produced consequently high HRTs). This means that the system performance may be impacted by evapotranspiration rate, during prolonged dry periods, and rainfall, during wet periods.

**Table 1**
Soil properties ($n = 3$) used in constructed wetland experimental units.

| Parameter | Unit | Mean ± standard deviation |
|---|---|---|
| pH water [1:2.5] | | 7.0 ± 0.08 |
| Organic matter | % *w/w* | 6.6 ± 0.45 |
| Total nitrogen | % w/w | 0.3 ± 0.02 |
| Total iron | mg/kg | 16,392.7 ± 538.3 |
| Total aluminium | mg/kg | 7888.7 ± 332.3 |
| Total calcium | mg/kg | 3284.3 ± 210.3 |
| Total magnesium | mg/kg | 2904.3 ± 94.0 |
| Total phosphorus | mg/kg | 696.7 ± 44.1 |
| Total potassium | mg/kg | 1105.7 ± 60.7 |
| Total carbon | % w/w | 3.50 ± 0.2 |
| Carbon:nitrogen ratio | :1 | 10.9 ± 0.1 |

**Table 2**
Influent raw DWW and supernatant characterization: mean ± standard deviations (SD; $n = 34$).

| Parameter | Raw DWW | Supernatant | Pre-treatment efficiency (%) |
|---|---|---|---|
| COD (mg $L^{-1}$) | 5385 ± 1212 | 1311 ± 375 | 75.7 (***) |
| Turbidity (NTU) | 1976 ± 610 | 85 ± 80 | 95.7 (***) |
| TSS (mg $L^{-1}$) | 1975 ± 825 | 108 ± 96 | 94.6 (***) |
| TP (mg $L^{-1}$) | 42.5 ± 15.0 | 1.14 ± 1.0 | 97.3 (***) |
| DRP (mg $L^{-1}$) | 19.3 ± 12.04 | 0.21 ± 0.51 | 98.9 (***) |
| TN-N (mg $L^{-1}$) | 240.5 ± 60.3 | 129.6 ± 40.3 | 46.1 (***) |
| TNp-N (mg $L^{-1}$) | 75.6 ± 31 | 4.3 ± 7.2 | 94.3 (***) |
| $NH_4$-N (mg $L^{-1}$) | 118.4 ± 40.8 | 87 ± 30.4 | 26.5 (***) |
| DON-N (mg $L^{-1}$) | 49.2 ± 25.3 | 34.8 ± 23 | 29.2 (*) |
| TON-N (mg $L^{-1}$) | 0.8 ± 2.1 | 3.9 ± 4.4 | 390 (***) |
| pH | 7.35 ± 0.57 | 6.1 ± 0.9 | 17.3 (***) |
| Cl (mg $L^{-1}$) | 155 ± 66 | 833 ± 145 | 438 (***) |
| Sludge volume (%) | 100 | 24.6 ± 5.3 | 75.4 (***) |

Statistically significant differences between Raw DWW and supernatant are shown at $P < 0.001$ as ***; $P < 0.01$ as **; $P < 0.05$ as * and no significant difference as NS.

**Table 3**
Experimental phases of different operational regimes of OLR and HLR applied to raw and supernatant CWs during a period of 43 weeks.

| Operation mode | Phase | Weeks | Waste source | OLR (g $m^{-2}$ $d^{-1}$) Mean ± SD | HLR (L $m^{-2}$ $d^{-1}$) Mean ± SD | HRT[a] (days) | Dosing frequency |
|---|---|---|---|---|---|---|---|
| Identical OLR | 1 | 1 to 7 | Raw DWW | 7 ± 1.5 | 1.4 ± 0.4 | 85 | 4 |
| | | | Supernatant | 7 ± 1.5 | 4.7 ± 0.9 | 25 | |
| | 2 | 8 to 29 | Raw DWW | 3.5 ± 1 | 0.7 ± 0.2 | 170 | 4 |
| | | | Supernatant | 3.5 ± 1 | 2.3 ± 0.45 | 50 | |
| Identical HLR | 3 | 30 to 35 | Raw DWW | 13 ± 2 | 2.3 ± 0.45 | 50 | 4 |
| | | | Supernatant | 3.5 ± 1 | 2.3 ± 0.45 | 50 | |
| | 4 | 36 to 43 | Raw DWW | 25 ± 2.5 | 4.7 ± 0.9 | 25 | 8 |
| | | | Supernatant | 7 ± 1.5 | 4.7 ± 0.9 | 25 | |

[a] Treatment volume taken as 80 % of constructed volume for FWS wetlands (i.e. 20 % comprising plant shoots, sludge and litter; Tanner and Kloosterman, 1997).

### 2.3. Sample analyses

Water samples were collected once a week from the influent raw and supernatant barrels ($n = 1$; bulk sample), and from the effluent collection containers (n = 3 for both the raw and supernatant CWs) for analysis. In periods of reduced rainfall in summer, there was no outflow and water samples were taken directly from within the CWs, as close to the outlet as possible. In total, there were 272 samples collected across 34 sampling weeks. The samples were stored at 4 °C for all parameters, except for COD analysis, which required samples to be preserved at −20 °C and analysed within 14 days.

Temperature and pH were recorded instantly using an HQ40d Multi Meter (HACH, USA). Turbidity was measured using an Orion AQUA fast turbidity meter (AQ3010, ThermoFisher Scientific, USA). COD was measured using the dichromate digestion method. TSS was measured using the gravimetric procedure (APHA, 2005). Filtered water samples were analysed for dissolved reactive phosphorus (DRP), ammonium ($NH_4$-N), total oxidised N (TON), nitrite ($NO_2$-N) and chloride (Cl), using a nutrient analyser (Aquakem 600A/Konelab 60, Thermo Clinical Labsystems).

Nitrate ($NO_3$-N) was calculated by deducting $NO_2$-N from TON. Total phosphorus, filtered TP ($TP_F$), total nitrogen (TN) and filtered TN ($TN_F$), were measured using the acid persulphate digestion method. Particulate phosphorus (PP) was calculated by deducting $TP_F$ from TP. Particulate N ($TN_P$) was calculated by deducting $TN_F$ from TN. Dissolved organic N (DON) was calculated by deducting TON and $NH_4$–N from $TN_F$. Dissolved organic P (DOP) was calculated by deducting DRP from $TP_F$. All tests were in accordance with the standard methods (APHA, 2005).

On-site climatic conditions and metrological data (Table S2) were obtained from the Moorepark automatic weather station, which was about 500 m distant from the study area.

### 2.4. Data analysis

Statistical analyses were conducted using SAS 9.4 (SAS Institute Inc., USA). PROC MIXED was used to undertake repeated measures modelling. PROC MIXED overcomes challenges associated with non-normal distribution and enables unbalanced replication associated with the data. The model comprised the following factors: Treatment (four treatments: influent raw DWW, effluent from raw DWW CWs, influent supernatant, effluent from supernatant CWs), Week (various weeks that changed from phase to phase), and the interaction between these factors (Treatment × Week) as fixed terms. The specific CWs were treated as a repeated measure. LSMEANS statement (with a Tukey adjustment) identified significant differences between treatments. Four separate models were established, a separate model for each phase of the experiment, to account for methodological differences between phases as described in Section 2.2. Probability values of $p > 0.05$ were considered not to be significant.

## 3. Results and discussion

### 3.1. Organic matter removal

Pre-treatment by $FeCl_3$ decreased COD significantly ($P < 0.001$) from 5385 ± 1212 mg $L^{-1}$ in the influent raw DWW to 1311 ± 375 mg $L^{-1}$ in the influent supernatant (removal efficiency of 76 %; Fig. 2-A). Mohamed et al. (2020) achieved a similar effluent concentration of 1600 mg $L^{-1}$ (with removal efficiency of 84 %) for DWW amended with $FeCl_3$ at the same dosage (470 mg Fe $L^{-1}$; 10.83 g Fe $g^{-1}$ P). The supernatant concentration was still above the limit value for discharge to urban waters (125 mg $L^{-1}$; 91/271/EEC; EEC, 1991). Mohamed et al. (2020) proposed that removal of particulate COD was a significant mechanism of COD removal by $FeCl_3$, and that the remaining supernatant COD was in dissolved/soluble form, which can be removed by aerobic or anaerobic biological degradation process (Henze et al., 2008).

To operate both sets of CWs with an identical OLR during Phases 1 and 2 (OLRs of 7 and 3.5 g $m^{-2}$ $d^{-1}$, respectively), the HLR of the CWs receiving supernatant was 3.3 times higher than that of CWs receiving raw DWW. This because of the supernatant had a lower COD concentration than raw DWW, following $FeCl_3$ treatment. The systems reached steady-state operation during Phase 2 (OLR of 3.5 g $m^{-2}$ $d^{-1}$; Fig. 2-A), when the CWs were fully established and consistent COD, TSS, P and N effluent concentrations were attained. During the steady-state operation of Phase 2 (Wk 14-Wk 29), the mean effluent COD concentration for supernatant CWs (95 ± 24 mg $L^{-1}$) was lower ($P < 0.001$) than raw DWW CWs (183 ± 31 mg $L^{-1}$), and below the limit values for discharge to urban waters (125 mg $L^{-1}$; 91/271/EEC; EEC, 1991; Fig. 2-A).

In Phase 3, when the HLR of the raw DWW CWs was elevated to equal that of the supernatant CWs (2.3 L $m^{-2}$ $d^{-1}$), the OLR was raised to 13 g COD $m^{-2}$ $d^{-1}$ for the raw DWW CWs (Fig. 3-A). The EPA guidelines (US Environmental Protection Agency (USEPA), 1992) required that the OLR should not exceed 6 g BOD $m^{-2}$ $d^{-1}$ (equivalent to 10 g COD $m^{-2}$ $d^{-1}$). As a result of the increase in OLR, the raw DWW CWs produced a mean effluent COD concentration of 498 ± 159 mg $L^{-1}$ (Fig. 2-A), significantly above the EU directive standard (125 mg $L^{-1}$). The supernatant CWs, which retained the lower OLR of 3.5 g COD $m^{-2}$ $d^{-1}$, performed significantly better ($P < 0.001$) and had a mean effluent COD concentration of 112 ± 22 mg $L^{-1}$ (Fig. 2-A).

In Phase 4, when the HLR was elevated to 4.7 L $m^{-2}$ $d^{-1}$ for both set of CWs, the OLR was increased to 25 g COD $m^{-2}$ $d^{-1}$ for the raw DWW CWs and 6 g COD $m^{-2}$ $d^{-1}$ for the supernatant CWs (Fig. 3-A). Because of the high OLR on the raw DWW CWs, their capability to remove COD further deteriorated, with effluent COD mean concentrations of 1070 ± 282 mg $L^{-1}$, higher than the supernatant CWs' effluent COD concentration (144 ± 31 mg $L^{-1}$, $P < 0.001$; Fig. 2-A), surpassing the threshold value of 125 mg COD $L^{-1}$, and approaching the influent supernatant.

Towards the end of Phase 4 an increase in effluent COD from the supernatant CWs was recorded. The low temperature recorded in November and December (Table S2) may have reduced the bacterial activity, and therefore the effluent COD from the supernatant CWs slightly exceeded the threshold. Furthermore, the OLR for supernatant CWs was at the upper design limit specified by New Zealand guidelines for the treatment of farm wastewaters (Tanner and Kloosterman, 1997), which recommends that the OLR should not exceed 3 g BOD $m^{-2}$ $d^{-1}$ (equivalent to 5 g COD $m^{-2}$ $d^{-1}$) for FWS CWs.

Overall, the pre-treatment step by $FeCl_3$ had a significant impact on reducing the OLR in the supernatant CWs (Fig. 3-A), and therefore facilitated higher HLR operation while obtaining good effluent quality.

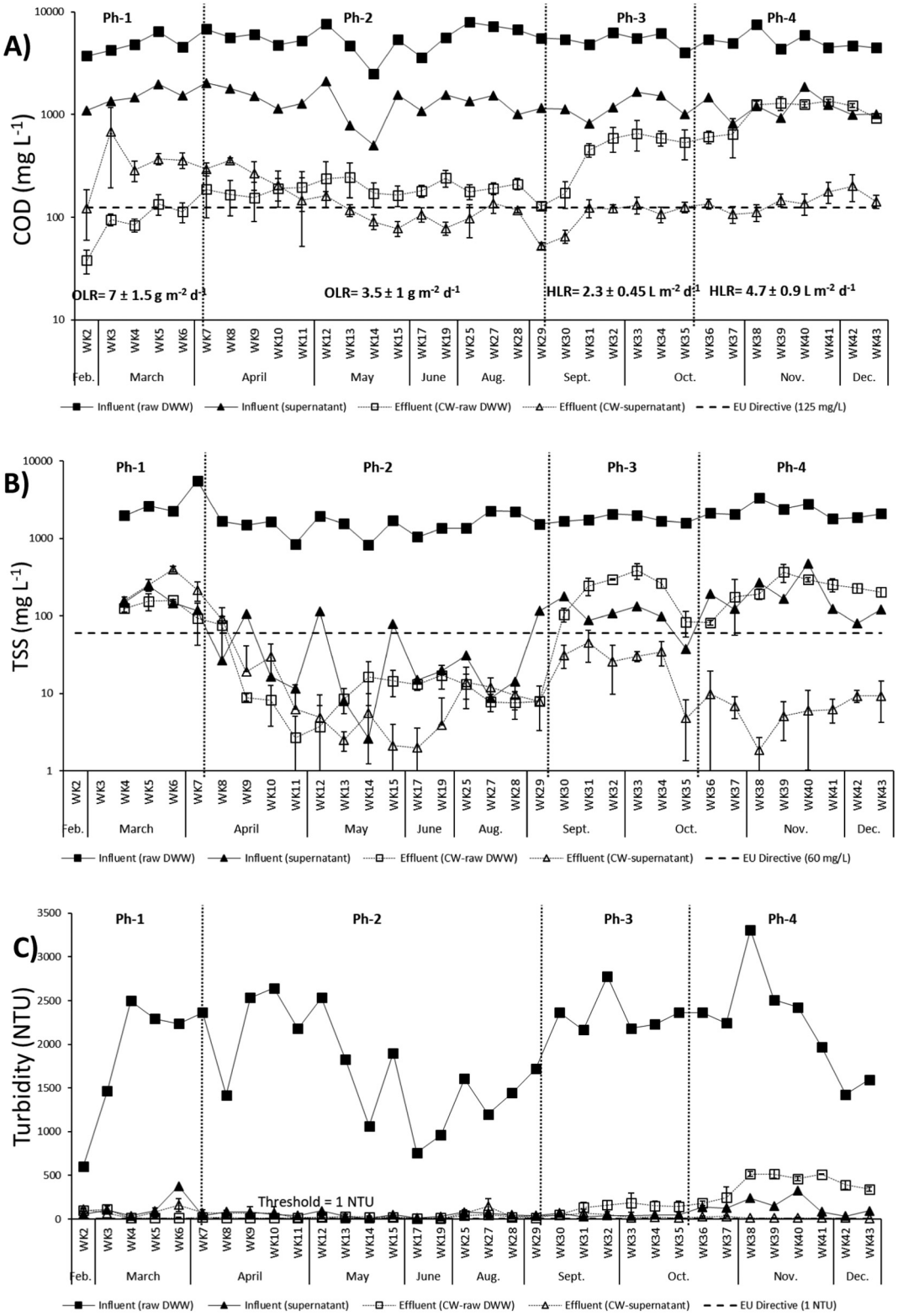


**Fig. 2.** Influent and effluent concentrations of raw DWW and supernatant constructed wetlands and EU directive limits for a study period of 43 weeks: A) chemical oxygen demand (COD); B) total suspended solids (TSS); and C) turbidity.

### *3.2. Suspended solids and turbidity removal*

The $FeCl_3$ reduced TSS and turbidity significantly ($P < 0.0001$) from 1975 ± 825 mg $L^{-1}$ and 1976 ± 610 NTU in the influent raw DSW to 108 ± 96 mg $L^{-1}$ and 85 ± 80 NTU in the influent supernatant, respectively (removal efficiency of 96 %; Fig. 2-B and -C). Cameron and Di (2019) and Mohamed et al. (2020) reported similar removals (97 %) using poly-ferric sulphate and $FeCl_3$ coagulants to treat DWW.

Throughout the steady-state/stable operation of Phase 2 (Wk 14-Wk 29), both raw DWW and supernatant CWs were efficient at reducing TSS to below the standard limit of 60 mg $L^{-1}$ stated by the EU directive for urban water discharge (91/271/EEC; EEC, 1991). Nevertheless, the

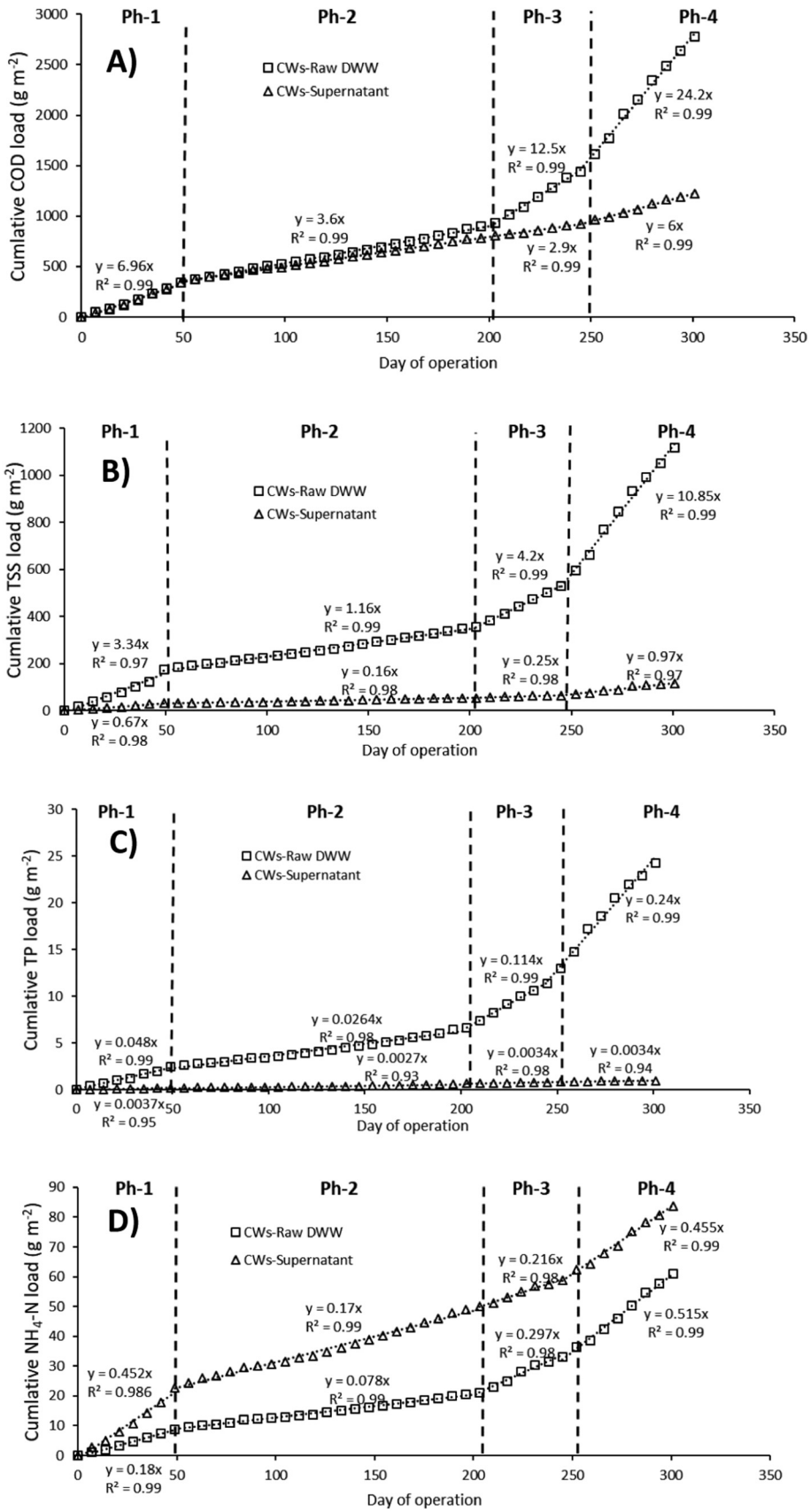


**Fig. 3.** Cumulative loads of pollutants on raw DWW and supernatant constructed wetlands for a study period of 43 weeks: A) chemical oxygen demand (COD); B) total suspended solids (TSS); C) total phosphorus; and D) Ammonium (NH4-N). Slopes denote loading rates in g $m^{-2}$ $d^{-1}$ for different parameters in different phases.

supernatant CWs (7.2 ± 4.2 mg $L^{-1}$) performed slightly better ($P < 0.05$) for the removal of TSS than raw DWW CWs (12.2 ± 3.7 mg $L^{-1}$; Fig. 2-B). During this phase, the suspended solids loading rate (SSLR) for raw DWW CWs (1.2 g SS $m^{-2}$ $d^{-1}$) was 7–8 times higher than the supernatant CWs (0.16 g SS $m^{-2}$ $d^{-1}$; Fig. 3-B).

In Phase 3, the SSLR of raw DWW CWs was elevated to 4.2 g SS $m^{-2}$ $d^{-1}$ compared to 0.25 g SS $m^{-2}$ $d^{-1}$ for supernatant CWs (Fig. 3-B). Consequently, the effluent TSS concentration from the raw DWW CWs (231 ± 106 mg $L^{-1}$) was significantly higher ($P < 0.001$) than the supernatant CWs (29 ± 12 mg $L^{-1}$; Fig. 2-B), and exceeded the EU limit value of 60 mg $L^{-1}$. In Phase 4, the SSLR increased to 11 g SS $m^{-2}$ $d^{-1}$ for raw DWW CWs and 1 g SS $m^{-2}$ $d^{-1}$ for supernatant CWs (Fig. 3-B). The EPA guidelines (US Environmental Protection Agency (USEPA), 1992) required that the SSLR should not exceed 5 g TSS $m^{-2}$ $d^{-1}$. Despite this increase in SSLR for raw DWW CWs, the mean effluent TSS concentrations was similar to Phase 3 (225.5 ± 79 mg $L^{-1}$; $P > 0.05$), but was significantly higher than the supernatant CWs (6.8 ± 2.4 mg $L^{-1}$; $P < 0.001$; Fig. 2-B) and exceeded the EU limit value of 60 mg $L^{-1}$.

Similar to TSS trends, the mean effluent turbidity in Phases 3 and 4 from supernatant CWs (36 ± 22 and 16 ± 7 NTU) was consistent and significantly lower ($P < 0.001$) than those from raw DWW CWs (137 ± 42 and 396 ± 121 NTU), respectively (Fig. 2-C). Overall, the pre-treatment step by $FeCl_3$ had a significant impact on reducing the SSLR (Fig. 3-B) in the supernatant CWs, and therefore prevented the losses of particulate matter/TSS in the effluent.

### 3.3. Phosphorus retention

Total P and DRP were reduced significantly ($P < 0.0001$) by $FeCl_3$ treatment from 42.5 ± 15 and 19.3 ± 12.04 mg $L^{-1}$ in the influent raw DWW to 1.14 ± 1.0 and 0.21 ± 0.51 mg $L^{-1}$ in the influent supernatant, respectively (removal efficiency of 98 %; Fig. 4-A). Similarly, Fenton et al. (2011) attained P effluent concentration <1 mg $L^{-1}$ using $FeCl_3$ at a dosage of 200 g Fe $g^{-1}$ P for DWW treatment.

Throughout the steady-state/stable operation of Phase 2 (Wk 14-Wk 29), both sets of CWs were effective at removing TP to below the EU standard limit of 2 mg $L^{-1}$ (91/271/EEC; EEC, 1991; Fig. 4-A). Nevertheless, supernatant CWs (0.18 ± 0.15 mg $L^{-1}$) performed significantly better ($P < 0.001$) than raw DWW CWs (1.17 ± 0.5 mg $L^{-1}$; Fig. 4-A). For DRP, both effluents were below the EU threshold value of 1 mg $L^{-1}$, but the effluent DRP from the supernatant CWs (0.013 ± 0.01 mg $L^{-1}$) was significantly lower ($P < 0.001$) than raw DWW CWs (0.26 ± 0.15 mg $L^{-1}$; Fig. 4-B).

In Phases 3 and 4, the TP loading rates on the raw DWW CWs were higher, by up to two orders of magnitude, than the supernatant CWs (Fig. 3-C). Because of these high TP loading rates applied in raw DWW CWs, the effluent TP concentration increased sharply starting from Wk 30, and exceeded the supernatant influent and the threshold value of 2 mg $L^{-1}$ (Fig. 4-A). In Phases 3 and 4, the corresponding mean effluent TP concentrations for raw DWW CWs were 3.86 ± 1.17 and 14.55 ± 4.7 mg $L^{-1}$, and were significantly higher ($P < 0.001$) than supernatant CWs, which achieved concentrations of 0.12 ± 0.034 and 0.16 ± 0.032 mg $L^{-1}$, respectively (Fig. 4-A). There was also surge in the effluent DRP of raw DWW CWs in Phase 4 (3.8 ± 1.3 mg $L^{-1}$ > 1 mg $L^{-1}$; Fig. 4-B). During Phases 3 and 4, the effluent TP from raw DWW CWs comprised mainly PP (60 %), followed by DRP (22 %), then DOP (18 %) (Fig. 4-C). The high content of PP in the effluent TP implied that the losses of TSS and particulate matter in the effluent of raw DWW CWs was the main reason for deteriorating effluent P in Phase 3 and 4.

At P loadings below 5 g P $m^{-2}$ $yr^{-1}$, a wetland soil can adsorb >90 % of the total received P (Healy et al., 2007). However, the applied P loading rates on the current raw DWW CWs exceeded this recommended value across the phases (10, 40 and 80 g P $m^{-2}$ $yr^{-1}$ in Phases 2, 3 and 4, respectively; Fig. 3-C). Lu et al. (2009) calculated a TP removal efficiency of only 59 % for a FWS CW treating agricultural runoff at TP loads of 12 g P $m^{-2}$ $yr^{-1}$. Vymazal (2007) indicated that the P removal by plant uptake (with successive harvesting) is trivial but it could be only significant for low loaded systems (e.g. 10–20 g P $m^{-2}$ $yr^{-1}$). The cumulative P mass loaded in the current raw DWW CWs until Wk 43 was approximately 25 g P $m^{-2}$ versus 1 g P $m^{-2}$ for supernatant CWs (Fig. 3-C). Therefore, using $FeCl_3$ coagulation as a pre-treatment step facilitates the possibility to reduce P load significantly and prevent P losses in the effluent.

### 3.4. Nitrogen removal and retention

Pre-treatment by $FeCl_3$ reduced total nitrogen significantly ($P < 0.001$) from 240.5 ± 60.3 mg $L^{-1}$ in the influent raw DWW to 129.6 ± 40.3 mg $L^{-1}$ in the influent supernatant (Fig. 5-A), but the removal efficiency (46 %) was lower than that of COD, turbidity, TSS and TP. The supernatant concentration was above the limits for urban water discharge (15 mg $L^{-1}$; 91/271/EEC; EEC, 1991). Similar to this study, Cameron and Di (2019) measured an effluent TN concentration of 87 mg $L^{-1}$ (removal efficiency of 57 %) using poly-ferric sulphate coagulant to treat DWW at an optimum dosage of 214 mg Fe $L^{-1}$ (6.1 g Fe $g^{-1}$ P). The residual N in the supernatant was mainly $NH_4$-N (87 ± 30.4 mg $L^{-1}$, 27 % removal, Fig. 5-B) and DON (34.8 ± 23 mg $L^{-1}$, 29 % removal, Fig. 5-C), which can be only removed through other biological and chemical transformation processes such as nitrification-denitrification, bio-adsorption, volatilization and plant uptake (Vymazal, 2007; Henze et al., 2008). The main mechanism of N removal by $FeCl_3$ was due to particulate N removal through sedimentation (94 % removal; Fig. 5-D).

During the start-up operation (Phase 1), the CWs treating raw DWW performed better in TN removal than CWs treating the supernatant (Fig. 5-A). The effluent TN from supernatant CWs comprised mainly $NH_4$-N in Phase 1 (Fig. 6). The raw DWW CWs had a better $NH_4$-N removal than supernatant CWs (Fig. 5-B) likely because the raw DWW CWs operated with a lower $NH_4$-N loading rate than supernatant CWs (Fig. 3-D). The effluent TN (including $NH_4$-N and DON) from supernatant CWs started to decrease linearly in Phase 2 (From Wk 8 - Wk 14) when the OLR was reduced by half (Fig. 5-A, B and C). This reduction also coincided with the period when the plants started to grow rapidly (growing season of reeds), which indicated that plant uptake was likely the main mechanism of $NH_4$-N removal.

During the steady-state of Phase 2, both raw DWW and supernatant CWs were effective at removing TN to below the EU standard limit of 15 mg $L^{-1}$ (91/271/EEC; EEC, 1991) as both achieved effluent concentrations of 7.83 ± 2. 5 mg $L^{-1}$ and 5.1 ± 1.8 mg $L^{-1}$ (Fig. 5-A). The effluent TN from raw DWW CWs comprised mainly DON, while the effluent TN from supernatant CWs comprised mainly both DON and $NH_4$-N (Fig. 6-B). Both sets of CWs were effective in removing particulate N in Phase 2 (Fig. 5-D).

In Phase 3, when the operational HLR of the raw DWW CWs was increased to match that of the supernatant CWs (2.3 L $m^{-2}$ $d^{-1}$), the effluent TN from the raw DWW CWs started to deteriorate (18.6 ± 5.95 mg $L^{-1}$) and was significantly higher ($P < 0.001$) than those from supernatant CWs (4.1 ± 0.7 mg $L^{-1}$) and exceeded the threshold value of 15 mg $L^{-1}$. The majority of the effluent TN from raw DWW CWs was in dissolved organic and particulate forms (Fig. 6; Fig. S2), which indicated that the retention of TN in the raw DWW CWs was limited by the sedimentation process (loss of TSS in the effluent) and limited by adsorption process of DON as well. However, the effluent $NH_4$-N from raw DWW CWs remained low and the same as in Phase 2 (0.51 ± 0.47 mg $L^{-1}$), with no differences ($P > 0.05$) from the effluent of supernatant CWs (0.57 ± 0.47 mg $L^{-1}$; Fig. 5-B).

At the higher HLR of 4.7 L $m^{-2}$ $d^{-1}$ applied in Phase 4, both effluents of TN from raw DWW and supernatant CWs deteriorated and increased linearly, surpassing the threshold value of 15 mg $L^{-1}$ (Fig. 5-A). This mainly due to the high N loading rates applied on the CWs (Fig. 3-D).The applied TN on raw DWW CWs (439 g N $m^{-2}$ $yr^{-1}$) and supernatant CWs (277 g N $m^{-2}$ $yr^{-1}$) in Phase 4 were higher than the average TN removal rate recorded for 85 FWS CWs (247 g N $m^{-2}$ $yr^{-1}$; Vymazal, 2007). These were also higher than the median TN removal rate recorded for 116 FWS CWs (129 g N $m^{-2}$ $yr^{-1}$) by Kadlec and Wallace (2008). The deterioration in

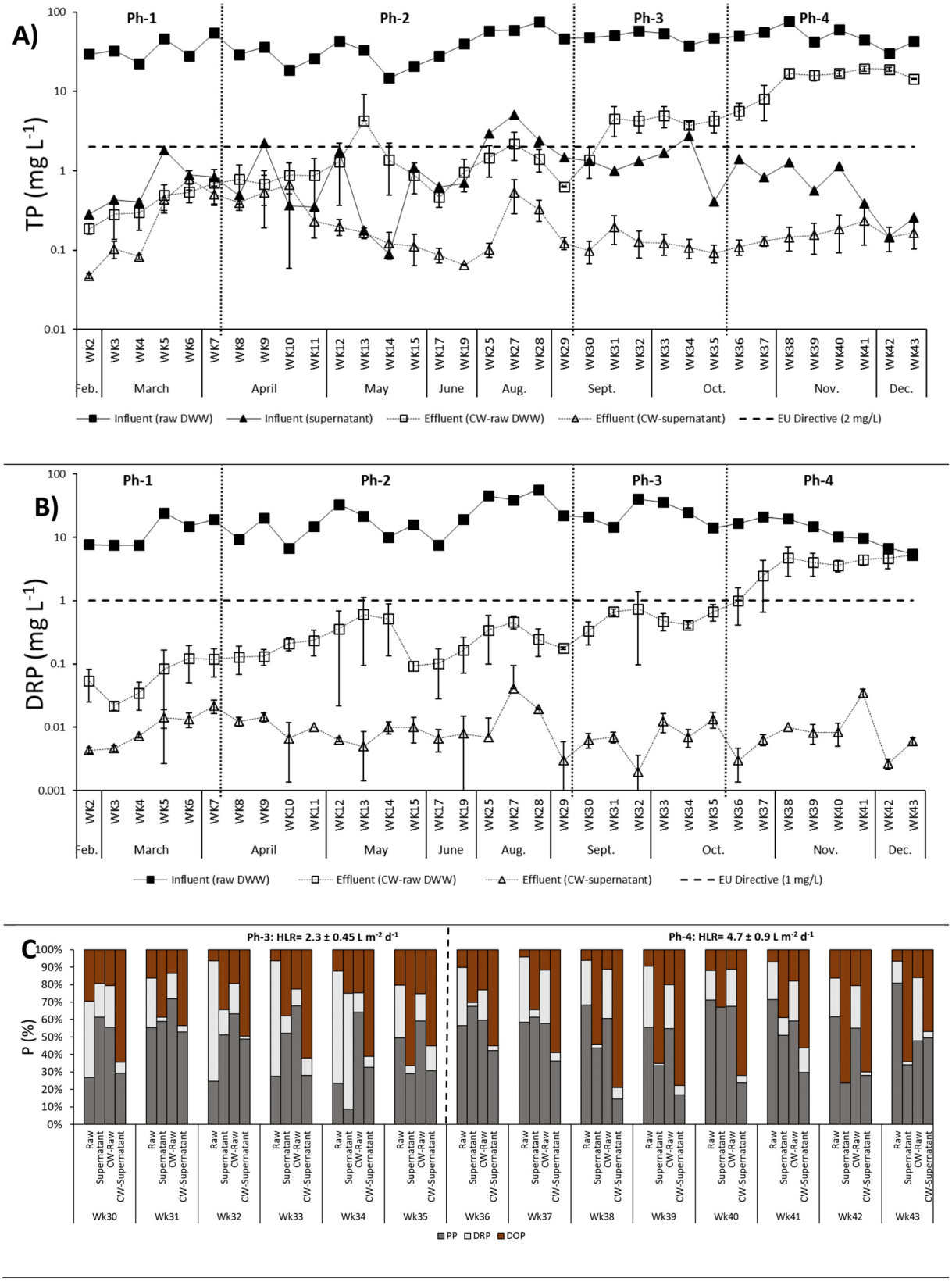


**Fig. 4.** Influent and effluent concentrations of raw DWW and supernatant constructed wetlands and EU directive limits for a study period of 43 weeks: A) total phosphorus (TP); B) dissolved reactive phosphorus (DRP); and C) phosphorus fractionation percentage during phase 3 and 4.

the effluent TN could also be due to the cold weather conditions in Phase 4 (Table S2), which may have resulted in decay of the vegetation and reduced microbial activity. The die-back of macrophytes during cold seasons may release organic N back into the CWs, which may then undergo ammonification (Kadlec and Wallace, 2008). Therefore, CWs may become a source of $NH_4$-N. Variability in nitrogen removal rates has been shown to be

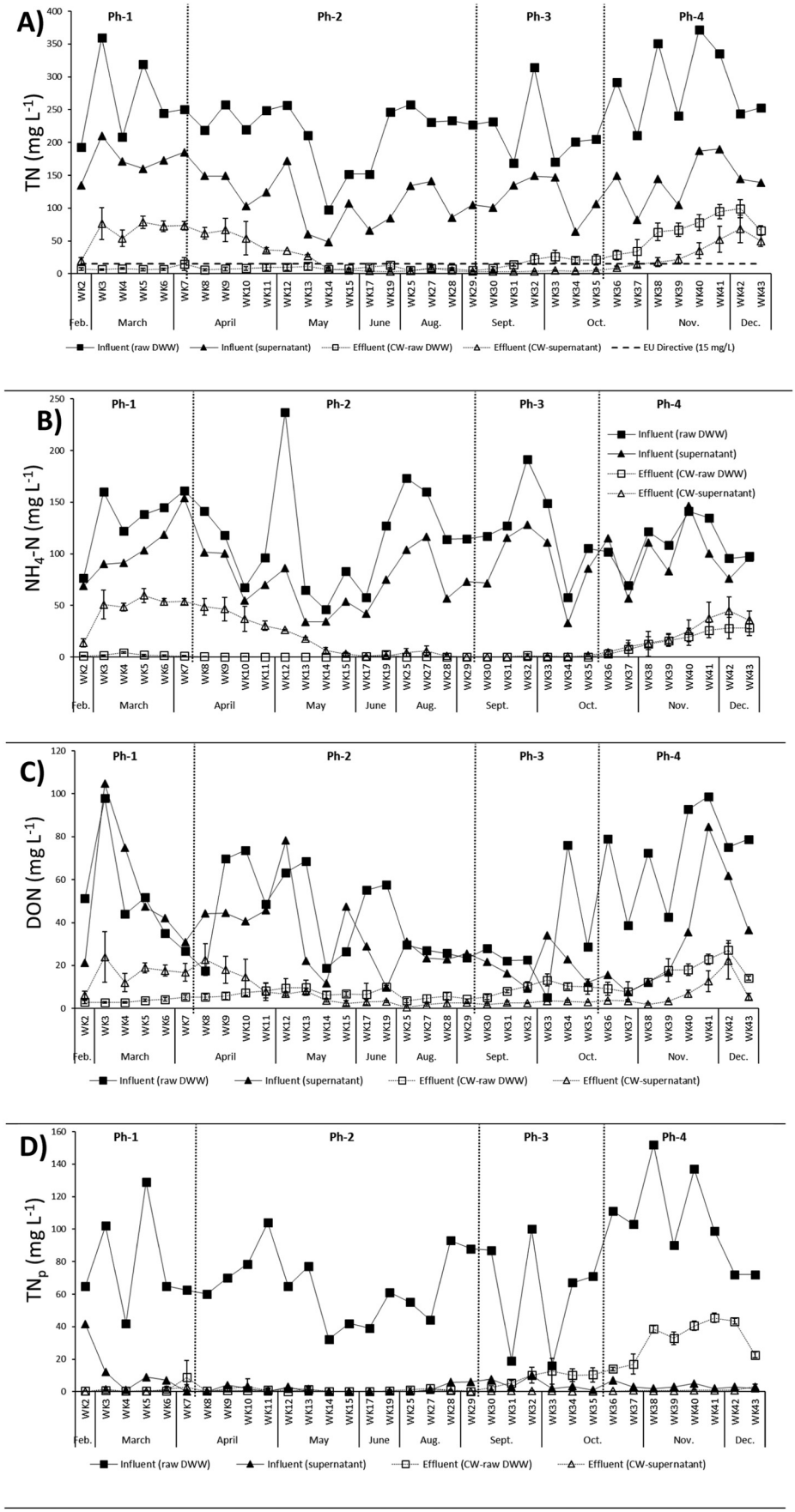


**Fig. 5.** Influent and effluent concentrations of raw DWW and supernatant constructed wetlands for a study period of 43 weeks: A) total nitrogen (TN); B) ammonium-N ($NH_4$-N); C) dissolved organic nitrogen (DON); and D) particulate nitrogen ($TN_p$).

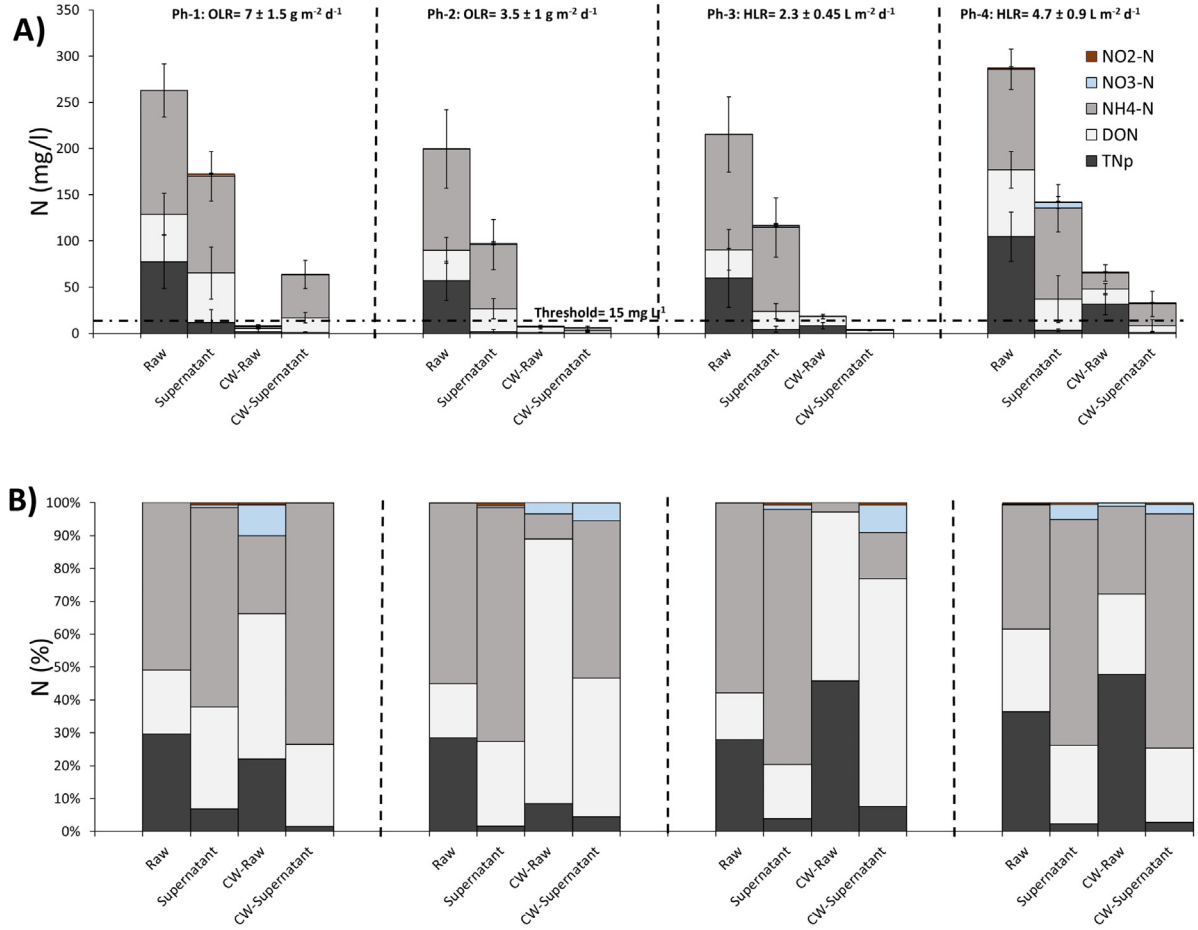


**Fig. 6.** Means influent and effluent total nitrogen concentrations of raw DWW and supernatant constructed wetlands for Phase 1 ($n = 6$), steady state of Phase 2 ($n = 8$), Phase 3 (n = 6) and Phase 4 (n = 8): A) Total nitrogen fractions in mg $L^{-1}$ (particulate N (TNp), dissolved organic nitrogen (DON), ammonium-N ($NH_4$-N), nitrate-N ($NO_3$-N) and nitrite ($NO_2$-N)); B) % of relative total nitrogen fractions.

temperature dependent, thus resulting in seasonal variation (Harrington et al., 2012; Uusheimo et al., 2018).

The effluent TN from supernatant CWs (33.6 ± 18.9 mg $L^{-1}$) was significantly lower ($P < 0.001$) than those from raw DWW CWs (66.4 ± 23.7 mg $L^{-1}$; Fig. 5-A). This difference was mainly due to the advantages provided by the pre-treatment step, which reduced the particulate and dissolved organic N load on supernatant CWs. The effluents $TN_p$ and DON from the supernatant CWs were significantly lower ($P < 0.001$) than those from raw DWW CWs (Figs. 5-C and -D; 6). In contrast, the raw DWW CW performed slightly better than supernatant CWs for $NH_4$-N removal (Figs. 5-B; 6), although $NH_4$-N load on raw DWW CWs was higher than that of supernatant CWs (Fig. 3-D).

Effluent $NO_3$-N and $NO_2$-N concentrations from both sets of CWs were minimal or negligible across the phases (Fig. 6), which either indicate that nitrification in CWs was low, or simultaneous denitrification may have occurred and therefore masked the real amount of $NH_4$-N nitrified. Matheson et al. (2002) suggested that 60 % of TN is removed from FWS CWs through denitrification and 15 % through plant uptake, and the remainder is stored in the soil. Vymazal (2007) found that nitrification-denitrification and ammonium volatilization are the main nitrogen transformation processes that take place in FWS CWs and that they occur at rates greater than plant uptake.

The pH of the effluent raw DWW and supernatant CWs was slightly alkaline during rainfall periods (Phase 1 and 4; pH = 8; Fig. S3), which may have encouraged very minor ammonia volatilization. The losses of ammonia ($NH_3$) through volatilization from CWs are insignificant if pH value is below 7.5 (Vymazal, 2007).

### 3.5. Summary results and implication of the study

The optimum conditions for each parameter to achieve best performance are summarised in Table 4. The optimum mass loading rate for each parameter was almost identical for both sets of CWs. However, the optimum HLR for supernatant CWs was lower than raw DWW CWs. This was due to the fact that the supernatant had lower contaminant concentrations than raw DWW, following $FeCl_3$ amendment. Unlike other parameters, raw DWW CWs did not meet the EU directive standard limit for COD even at the optimum OLR (Table 4). The removal of COD and TSS was more cost-effective than TP and TN removals (Table 4). The decision to select the coagulation treatment as a pre-treatment step depends on the wastewater characterization. For example, a hybrid system may only be feasible if the wastewater mainly comprises TSS, COD or TP, because smaller surface areas are required to treat these parameters (Table 4). In contrast, no saving in treatment area can be expected if the wastewater is only enriched with ammonium (Table 4).

Overall, the hybrid coagulation-CWs achieved superior effluent quality with high removal efficiencies (≥ 99 %) for all parameters, and consistently complied with EU directives concerning urban wastewater treatment (91/271/EEC; EEC, 1991) during Phases 2 and 3. The performance achieved by the hybrid system in the current study was better than those

**Table 4**
Showing optimum gravimetrical and hydraulic loading rates for different parameters, the average concentration of the final effluent (mg $L^{-1}$) at the optimum loading rate for each water quality parameter examined, cost of chemical per kg mass removal of each parameter, and expected saving in treatment area (when chemical dosing is used versus raw dairy wastewater) for each parameter.

| | Optimum mass loading rate (g $m^{-2}$ d) | | Optimum hydraulic loading rate (L $m^{-2}$ d) | | Final effluent (mg $L^{-1}$) | | | Cost[a] | Saving in area |
|---|---|---|---|---|---|---|---|---|---|
| | ICW-raw DWW | ICW-supernatant | ICW-raw DWW | ICW-supernatant | ICW-raw DWW | ICW-supernatant | EU Standard limits | (€ $kg^{-1}$ removed) | (%) |
| COD | 3.5 | 3.5 | 0.7 | 2.3 | 183 | 95 | 125 | 0.24 | 76 |
| TSS | 1.16 | 1 | 0.7 | 4.7 | 12.2 | 6.8 | 60 | 0.52 | 96 |
| TP-P | 0.0264 | 0.0034 | 0.7 | 4.7 | 1.17 | 0.16 | 2 | 23.6 | 98 |
| $NH_4$-N | 0.297 | 0.216 | 2.3 | 2.3 | 0.51 | 0.57 | 10 | 31 | 27 |
| TN-N | 0.152 | 0.277 | 0.7 | 2.3 | 7.83 | 4.1 | 15 | 8.8 | 46 |

[a] Cost was estimated using the prices of commercial $FeCl_3$. (520 € $m^{-3}$).

achieved by conventional CWs reported in the literature. For example, Healy and O'Flynn (2011) reviewed the performance of seven FWS constructed wetlands treating DWW on Irish farms and reported an average COD, TSS, $NH_4$-N and P removals of 91 %, 94 %, 88 % and 80 %, respectively, and final effluents were frequently in excess of the discharge limits (91/271/EEC; EEC, 1991).

The authors acknowledge issues with developing (and extrapolating) design criteria based on results achieved in relatively small-scale pilot-scale CWs such as those used in this study. However, with this caveat, it is recommended that a hybrid system should be operated with similar conditions to Phases 2 and 3 in the current study (OLR of 3.5 g $m^{-2}$ $d^{-1}$; HLR of 2.3 L $m^{-2}$ $d^{-1}$), because at higher OLRs and HLRs, the system will fail to meet COD and TN effluent standard as shown in Phase 4. For example, on a 100 cow dairy farm, with each cow producing 33 L $d^{-1}$ (Minogue et al., 2015), and an OLR of 3.5 g $m^{-2}$ $d^{-1}$ for the FWS CWs; an area of 1242 $m^2$ ($\approx$20 m $*$ 60 m) would be required for a hybrid system versus 5142 $m^2$ ($\approx$40 m $*$ 130 m) for a conventional CWs system.

Coagulation pre-treatment could bring more benefits if the type of wetlands examined were subsurface flow (SSF) CWs. With pre-treatment, subsurface horizontal flow (SSHF) and SSVF CWs can be operated with higher OLRs and HLRs than FWS CWs. Furthermore, a pre-treatment step will eliminate all TSS upfront, and therefore prevent clogging of SSHF and SSVF CWs, which is a major operational issue on these type of CWs (de Matos et al., 2018; Wang et al., 2021). This was obvious from the experiment of Mohamad et al. (2022), who combined coagulation treatment with an intermittent sand filter (ISF), which had characteristics and properties similar to SSVF CWs. In their experiment, a hybrid ISF was able to operate at an OLR of 30 COD g $m^{-2}$ $d^{-1}$ without showing any evidence of clogging, and performed significantly better than a conventional ISF.

The generated sludge from the pre-treatment step, which is enriched with nutrients and P-sorbing coagulants, may have agronomic value as an organic fertilizer, and therefore could be spread on farmland. The formation of ferric-phosphate chemical bonds in the amended sludge will reduce the solubility and mobility of P, thereby minimizing the risk of P losses to water via runoff or drainage (Che et al., 2022). Mohamed et al. (2020) examined the properties of the sludge generated from DWW amended with $FeCl_3$ and found that it had TP and TN contents three and two times higher than the influent raw DWW, respectively. In addition, their study found that the produced sludge had an *E. coli* concentration significantly lower than the influent raw DWW due to the acidity of the chemical coagulant. This means that the produced sludge is safer than raw DWW and can be spread to the farmland with fewer health risks.

Separation of the milking plant effluent from the other waste in raw DWW (i.e. cow faeces/slurry and urine) could provide the possibility to recover resources from the residual milk, which can be used for many purposes on the farm. This could also be considered as a management practice to reduce influent COD in raw DWW, and to increase the HLR on CWs.

## 4. Conclusion

A hybrid coagulation-CW system combines the advantages of both technologies and performs better than a conventional CW system. Combined systems may be operated at higher HLRs than conventional FWS CW systems, and therefore require a smaller sized footprint to treat the same volume of DWW. The pilot-scale hybrid coagulation-CW in this study was able to produce high effluent quality at optimum OLR of 3.5 g COD $m^{-2}$ $d^{-1}$ (HLR = 2.3 $L^{-1}$ $m^{-2}$ $d^{-1}$) and achieved removal efficiencies $\geq$99 % for all parameters, which complied with EU directives concerning urban wastewater treatment, and hence is suitable for discharge. Future studies should explore the possibility of testing coagulation pre-treatment method in combination with other type of CWs such as SSHF and SSVF CWs. Future research should also focus on the agronomic potential and value of reusing the sludge generated from the hybrid system as an organic bio-based fertilizer (Shi et al., 2021).

## CRediT authorship contribution statement

**A.Y.A. Mohamed**: Conceptualization; Data curation; Formal analysis; Investigation; Methodology; Validation; Visualization; Roles/Writing – original draft; Writing – review & editing. **A. Siggins:** Conceptualization; Funding acquisition; Investigation; Methodology; Supervision; Validation; Visualization; Writing – review & editing. **M.G. Healy:** Conceptualization; Funding acquisition; Investigation; Methodology; Supervision; Validation; Visualization; Writing – review & editing. **D. Ó hUallacháin:** Conceptualization; Funding acquisition; Investigation; Methodology; Supervision; Validation; Visualization; Writing – review & editing. **O. Fenton:** Conceptualization; Funding acquisition; Investigation; Methodology; Software; Supervision; Validation; Visualization;– review & editing. **P. Tuohy:** Conceptualization; Funding acquisition; Investigation; Methodology; Project administration; Resources; Supervision; Validation; Visualization; Writing – review & editing.

## Declaration of competing interest

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

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